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# The Structure-Property Relationship of Metallocene-based Ethylene Oligomerization Catalysts Using DFT and Graph Neural Networks

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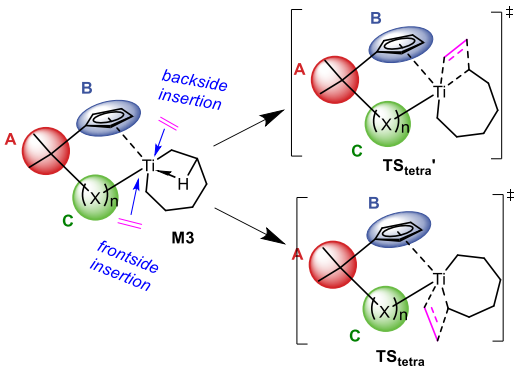
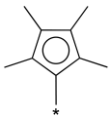
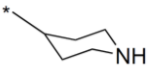
**Table S1.** The relative energy of $\mathbf{TS_{tetra}}$ $\Delta G_{ethylene-insertion}$ and the relative energy of $\mathbf{TS_{tetra}}$' $\Delta G_{ethylene-insertion}{}'$ responding to coordination-insertion occurred at the side of agnostic insertion and its opposite side for A1B1C9, A1B2C11, A1B7C9, A2B7C10, and A3B7C19.

| Index | ID |  | | | The relative energy of $\mathbf{TS_{tetra}}$ (frontside) $\Delta G_{ethylene-inser}$ (kcal/mol) | The relative energy of $\mathbf{TS_{tetra}}$' (backside) $\Delta G_{ethylene-insertion}$ (kcal/mol) |
|---|---|---|---|---|---|---|
| | | A | B | C | | |
| 1 | A1B1C9 | | | | 3.62 | 6.52 |
| 2 | A1B2C11 | |  |  | 9.06 | 14.04 |
| 3 | A1B7C9 | | | | 2.34 | 7.87 |
| 4 | A2B7C10 | | | | 9.21 | 10.07 |

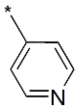
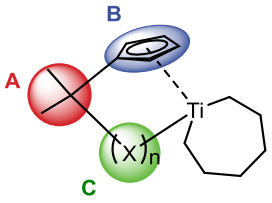
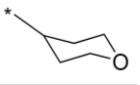
| | | | | | | |
|---|---|---|---|---|---|---|
| 5 | A3B7C19 | | |  | 7.71 | 11.66 |

**Table S2.** The energy barrier difference $\Delta\Delta G$ of the constructed 621 catalyst datasets. $\Delta\Delta G$ equals the fourth ethylene insertion energy barrier $\Delta G_{\text{ethylene insertion}}$ minus the $\beta$-hydrogen transfer (after the third ethylene insertion) energy barrier $\Delta G_{\beta-\text{H transfer}}$.

| Index | ID | Substructures in  | | | the energy barrier difference $\Delta\Delta G$ (kcal/mol) |
|---|---|---|---|---|---|
| | | A | B | C | |
| 1 | A1B1C1 | | | | -0.818 |
| 2 | A1B1C2 | | | | 0.548 |
| 3 | A1B1C3 | | | | -1.037 |
| 4 | A1B1C4 | | | | 6.745 |
| 5 | A1B1C5 | | | | 2.358 |
| 6 | A1B1C6 | | | | -17.536 |
| 7 | A1B1C7 | | | | -16.112 |
| 8 | A1B1C8 | | | | -14.625 |
| 9 | A1B1C9 | | |  | -16.781 |

| | | | | | |
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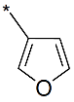
| 11 | A1B1C11 | | | | -15.674 |
| 12 | A1B1C12 | | |  | -3.245 |
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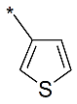
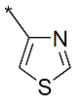
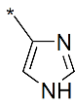
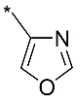
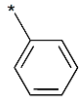
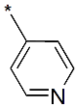
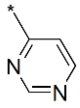
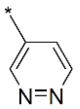
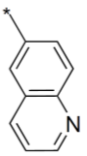
| 14 | A1B1C14 | | |  | -2.253 |
| 15 | A1B1C15 | | |  | 18.125 |
| 16 | A1B1C16 | | |  | 19.815 |
| 17 | A1B1C17 | | |  | 17.353 |
| 18 | A1B1C18 | | |  | 2.44 |
| 19 | A1B1C19 | | |  | -7.406 |
| 20 | A1B1C20 | | |  | -16.109 |
| 21 | A1B1C21 | | |  | -9.506 |
| 22 | A1B1C22 | | |  | -4.965 |
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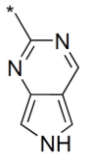
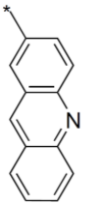
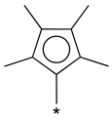
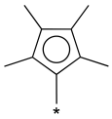
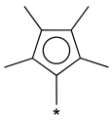
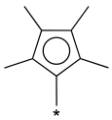
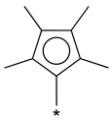
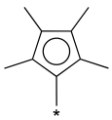
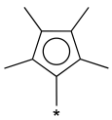
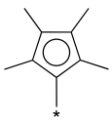
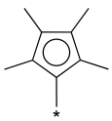
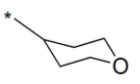
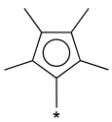
| 25 | A1B1C25 | | |  | 17.615 |
| 26 | A1B1C26 | | |  | -1.947 |
| 27 | A1B2C1 | |  | | 2.885 |
| 28 | A1B2C2 | |  | | 4.073 |
| 29 | A1B2C3 | |  | | 0.781 |
| 30 | A1B2C4 | |  | | 10.74 |
| 31 | A1B2C5 | |  | | 4.818 |
| 32 | A1B2C6 | |  | | -10.214 |
| 33 | A1B2C7 | |  | | -5.222 |
| 34 | A1B2C8 | |  | | -8.908 |
| 35 | A1B2C9 | |  |  | -7.374 |
| 36 | A1B2C10 | |  | | -6.559 |

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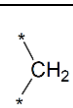
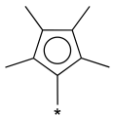
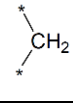
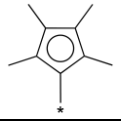
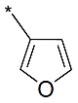
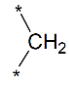
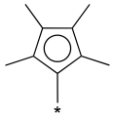
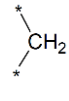
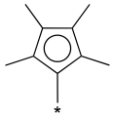
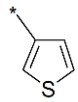
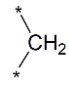
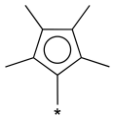
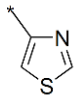
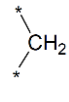
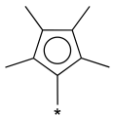
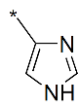
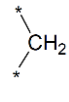
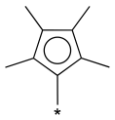
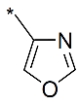
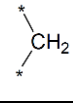
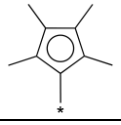
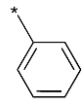
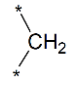
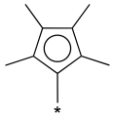
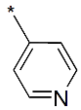
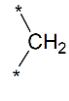
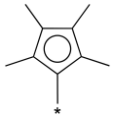
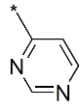
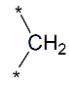
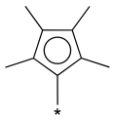
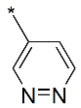
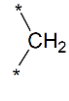
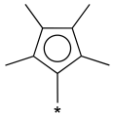
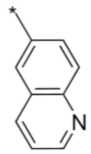
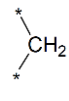
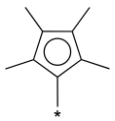
| 37 | A1B2C11 |  |  | | -6.098 |
| 38 | A1B2C12 |  |  |  | 0.98 |
| 39 | A1B2C13 |  |  | | -0.566 |
| 40 | A1B2C14 |  |  |  | 1.856 |
| 41 | A1B2C15 |  |  |  | 21.298 |
| 42 | A1B2C16 |  |  |  | 22.814 |
| 43 | A1B2C17 |  |  |  | 20.905 |
| 44 | A1B2C18 |  |  |  | -0.676 |
| 45 | A1B2C19 |  |  |  | -2.472 |
| 46 | A1B2C20 |  |  |  | -8.114 |
| 47 | A1B2C21 |  |  |  | -5.082 |
| 48 | A1B2C22 |  |  |  | -0.986 |
| 49 | A1B2C23 |  |  | | -0.83 |

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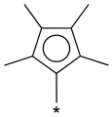
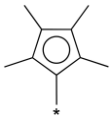
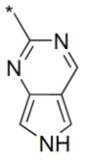
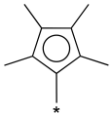
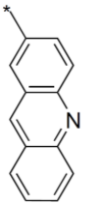
| 50 | A1B2C24 | |  | | 4.622 |
| 51 | A1B2C25 | |  |  | 20.779 |
| 52 | A1B2C26 | |  |  | 0.707 |
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| 54 | A1B3C2 | | | | -1.396 |
| 55 | A1B3C3 | | | | -2.126 |
| 56 | A1B3C4 | | | | 5.737 |
| 57 | A1B3C5 | | | | 0.011 |
| 58 | A1B3C6 | | | | -13.839 |
| 59 | A1B3C7 | | | | -15.764 |

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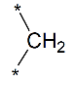
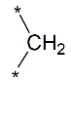
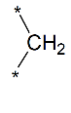
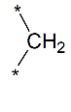
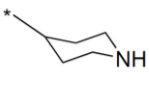
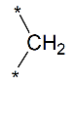
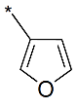
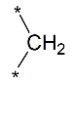
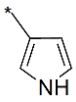
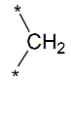
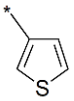
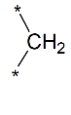
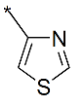
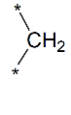
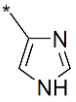
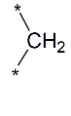
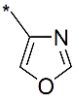
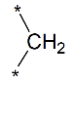
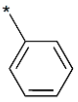
| 60 | A1B3C8 |  | | | -18.805 |
| 61 | A1B3C9 |  | | | -14.121 |
| 62 | A1B3C10 |  | | | -13.325 |
| 63 | A1B3C11 |  | |  | -12.661 |
| 64 | A1B3C12 |  | |  | -5.505 |
| 65 | A1B3C13 |  | |  | -4.185 |
| 66 | A1B3C14 |  | |  | -4.744 |
| 67 | A1B3C15 |  | |  | 14.875 |
| 68 | A1B3C16 |  | |  | 16.571 |
| 69 | A1B3C17 |  | |  | 14.396 |
| 70 | A1B3C18 |  | |  | -7.809 |

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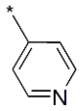
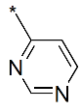
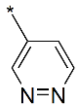
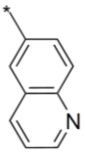
| 71 | A1B3C19 | | |  | -9.824 |
| 72 | A1B3C20 | | |  | -18.113 |
| 73 | A1B3C21 | | |  | -11.783 |
| 74 | A1B3C22 | | |  | -7.661 |
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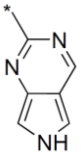
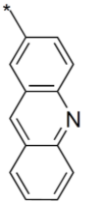
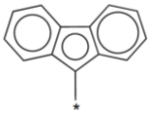
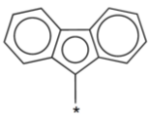
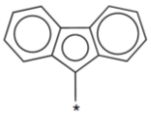
| 76 | A1B3C24 | | | | -1.653 |
| 77 | A1B3C25 | | |  | 14.405 |
| 78 | A1B3C26 | | |  | -5.604 |
| 79 | A1B4C1 | |  | | 5.367 |
| 80 | A1B4C2 | |  | | 5.934 |
| 81 | A1B4C3 | |  | | 4.169 |

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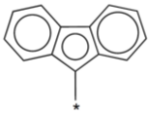
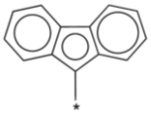
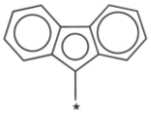
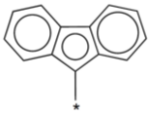
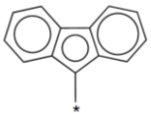
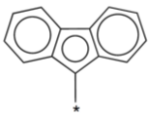
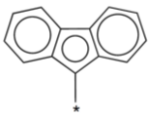
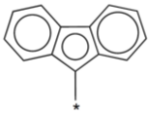
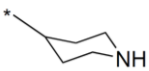
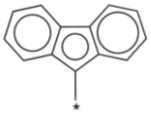
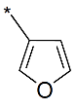
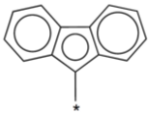
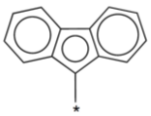
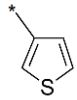
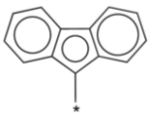
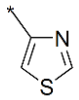
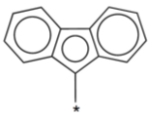
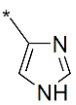
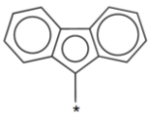
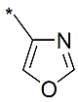
| 82 | A1B4C4 | |  | | 13.546 |
| 83 | A1B4C5 | |  | | 5.351 |
| 84 | A1B4C6 | |  | | -2.525 |
| 85 | A1B4C7 | |  | | -11.63 |
| 86 | A1B4C8 | |  | | -10.15 |
| 87 | A1B4C9 | |  | | -1.921 |
| 88 | A1B4C10 | |  | | -8.522 |
| 89 | A1B4C11 | |  |  | -0.39 |
| 90 | A1B4C12 | |  |  | 4.276 |
| 91 | A1B4C13 | |  | | -0.867 |
| 92 | A1B4C14 | |  |  | 3.98 |
| 93 | A1B4C15 | |  |  | 17.873 |
| 94 | A1B4C16 | |  |  | 18.997 |
| 95 | A1B4C17 | |  |  | 17.833 |

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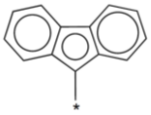
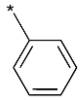
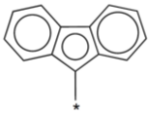
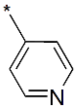
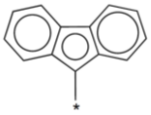
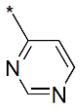
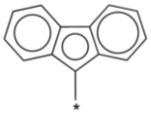
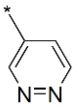
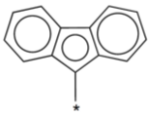
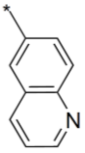
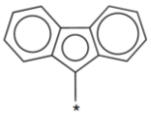
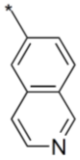
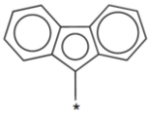
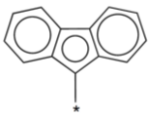
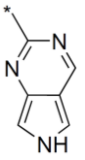
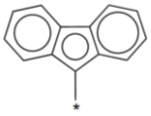
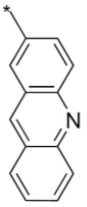
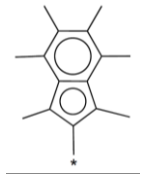
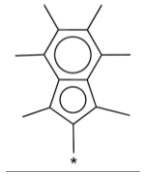
| 96 | A1B4C18 | |  |  | 1.33 |
| 97 | A1B4C19 | |  |  | -2.79 |
| 98 | A1B4C20 | |  |  | -4.163 |
| 99 | A1B4C21 | |  |  | -1.101 |
| 100 | A1B4C22 | |  |  | 1.507 |
| 101 | A1B4C23 | |  |  | -3.747 |
| 102 | A1B4C24 | |  | | 6.181 |
| 103 | A1B4C25 | |  |  | 16.623 |
| 104 | A1B4C26 | |  |  | 2.579 |
| 105 | A1B5C1 | |  | | 1.111 |
| 106 | A1B5C2 | |  | | 2.109 |

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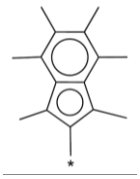
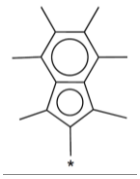
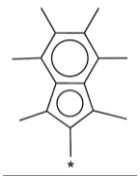
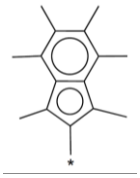
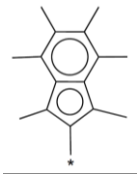
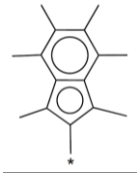
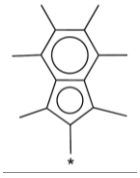
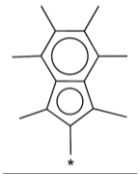
| 107 | A1B5C3 | |  | | 1.048 |
| 108 | A1B5C4 | |  | | 8.941 |
| 109 | A1B5C5 | |  | | 2.719 |
| 110 | A1B5C6 | |  | | -6.36 |
| 111 | A1B5C7 | |  | | -7.917 |
| 112 | A1B5C8 | |  | | -13.9 |
| 113 | A1B5C9 | |  | | -6.022 |
| 114 | A1B5C10 | |  | | -5.971 |

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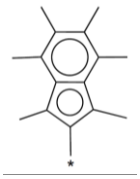
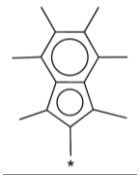
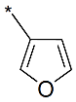
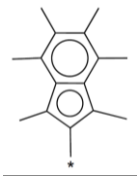
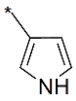
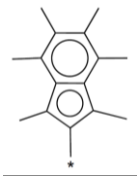
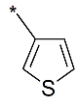
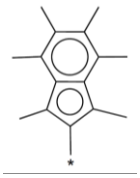
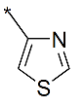
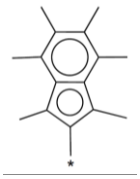
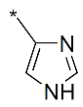
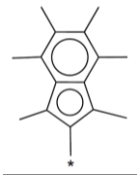
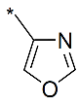
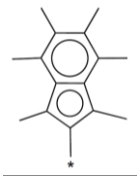
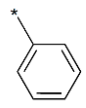
| 115 | A1B5C11 | |  | | -4.479 |
| 116 | A1B5C12 | |  |  | 0.585 |
| 117 | A1B5C13 | |  |  | -1.812 |
| 118 | A1B5C14 | |  |  | 0.959 |
| 119 | A1B5C15 | |  |  | 18.955 |
| 120 | A1B5C16 | |  |  | 20.134 |
| 121 | A1B5C17 | |  |  | 17.735 |
| 122 | A1B5C18 | |  |  | -6.632 |

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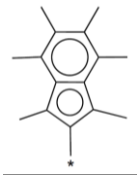
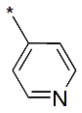
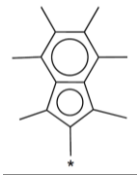
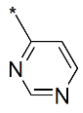
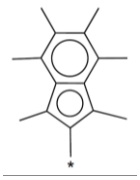
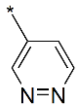
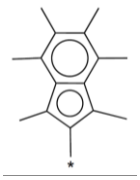
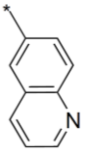
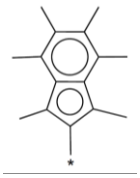
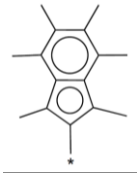
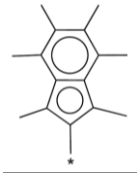
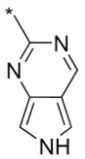
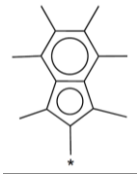
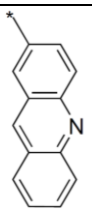
| 123 | A1B5C19 | |  |  | -8.055 |
| 124 | A1B5C20 | |  |  | -13.698 |
| 125 | A1B5C21 | |  |  | -10.114 |
| 126 | A1B5C22 | |  |  | -2.689 |
| 127 | A1B5C23 | |  | | -0.298 |
| 128 | A1B5C24 | |  | | 2.57 |
| 129 | A1B5C25 | |  |  | 18.683 |
| 130 | A1B5C26 | |  |  | 0.504 |
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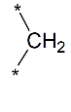
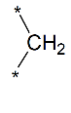
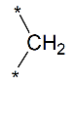
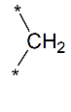
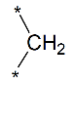
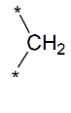
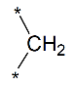
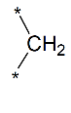
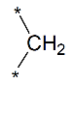
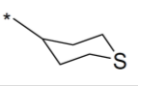
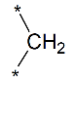
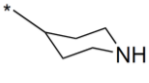
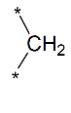
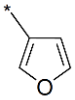
| 132 | A1B6C2 |  | | | 0.724 |
| 133 | A1B6C3 |  | | | -1.168 |
| 134 | A1B6C4 |  | | | 7.807 |
| 135 | A1B6C5 |  | | | 3.052 |
| 136 | A1B6C6 |  | | | -16.436 |
| 137 | A1B6C7 |  | | | -16.541 |
| 138 | A1B6C8 |  | | | -16.028 |
| 139 | A1B6C9 |  | | | -17.457 |
| 140 | A1B6C10 |  | |  | -12.095 |
| 141 | A1B6C11 |  | |  | -15.767 |
| 142 | A1B6C12 |  | |  | -4.187 |

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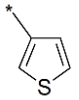
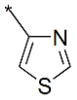
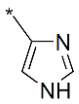
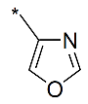
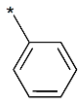
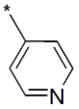
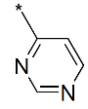
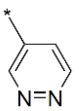
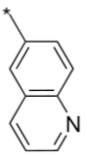
| 144 | A1B6C14 | | |  | -3.119 |
| 145 | A1B6C15 | | |  | 14.266 |
| 146 | A1B6C16 | | |  | 15.524 |
| 147 | A1B6C17 | | |  | 13.354 |
| 148 | A1B6C18 | | |  | -5.761 |
| 149 | A1B6C19 | | |  | -7.911 |
| 150 | A1B6C20 | | |  | -16.611 |
| 151 | A1B6C21 | | |  | -10.262 |
| 152 | A1B6C22 | | |  | -5.373 |
| 153 | A1B6C23 | | | | -5.132 |

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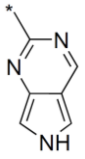
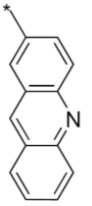
| 154 | A1B6C24 | | | | 0.96 |
| 155 | A1B6C25 | | |  | 15.35 |
| 156 | A1B6C26 | | |  | -1.591 |
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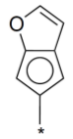
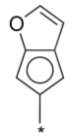
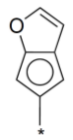
| 158 | A1B7C2 | |  | | 1.451 |
| 159 | A1B7C3 | |  | | -1.106 |
| 160 | A1B7C4 | |  | | 7.669 |
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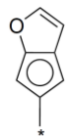
| 162 | A1B7C6 | |  | | -13.433 |
| 163 | A1B7C7 | | | | -16.197 |

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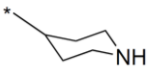
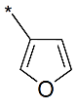
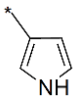
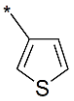
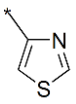
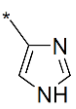
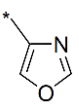
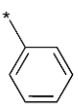
| 164 | A1B7C8 | | | | -16.601 |
| 165 | A1B7C9 | | | | -12.517 |
| 166 | A1B7C10 | | | | -11.761 |
| 167 | A1B7C11 | | |  | -11.197 |
| 168 | A1B7C12 | | |  | -3.752 |
| 169 | A1B7C13 | | |  | -1.08 |
| 170 | A1B7C14 | | |  | -2.77 |
| 171 | A1B7C15 | | |  | 14.986 |
| 172 | A1B7C16 | | |  | 16.28 |
| 173 | A1B7C17 | | |  | 14.118 |
| 174 | A1B7C18 | | |  | -5.542 |

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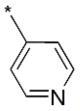
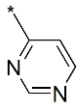
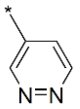
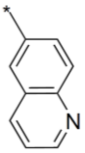
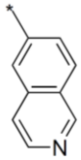
| 175 | A1B7C19 | | |  | -7.717 |
| 176 | A1B7C20 | | |  | -16.059 |
| 177 | A1B7C21 | | |  | -10.015 |
| 178 | A1B7C22 | | |  | -5.483 |
| 179 | A1B7C23 | | |  | -5.39 |
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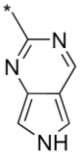
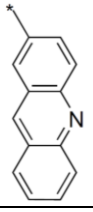
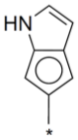
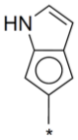
| 181 | A1B7C25 | | |  | 15.725 |
| 182 | A1B7C26 | | |  | -1.428 |
| 183 | A1B8C1 | |  | | 0.636 |
| 184 | A1B8C2 | |  | | 1.857 |

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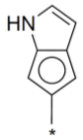
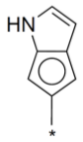
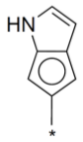
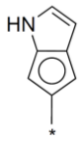
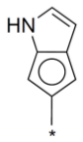
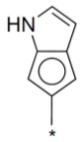
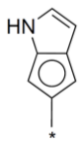
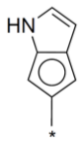
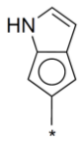
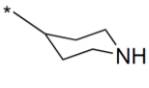
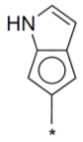
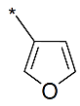
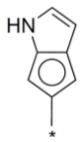
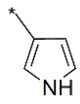
| 185 | A1B8C3 | |  | | -0.23 |
| 186 | A1B8C4 | |  | | 9.209 |
| 187 | A1B8C5 | |  | | 2.923 |
| 188 | A1B8C6 | |  | | -11.443 |
| 189 | A1B8C7 | |  | | -14.814 |
| 190 | A1B8C8 | |  | | -16.902 |
| 191 | A1B8C9 | |  | | -11.399 |
| 192 | A1B8C10 | |  | | -10.68 |
| 193 | A1B8C11 | |  |  | -10.017 |
| 194 | A1B8C12 | |  |  | -3.396 |
| 195 | A1B8C13 | |  |  | -1.564 |

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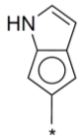
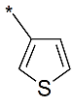
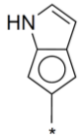
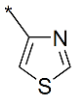
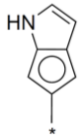
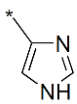
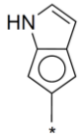
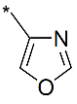
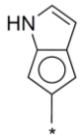
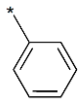
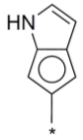
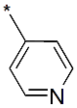
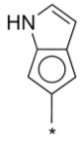
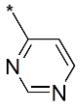
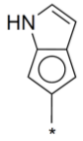
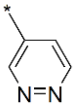
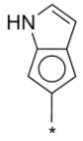
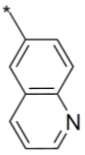
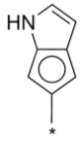
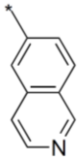
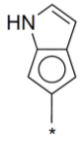
| 196 | A1B8C14 | |  |  | -2.511 |
| 197 | A1B8C15 | |  |  | 16.072 |
| 198 | A1B8C16 | |  |  | 17.586 |
| 199 | A1B8C17 | |  |  | 15.493 |
| 200 | A1B8C18 | |  |  | -5.228 |
| 201 | A1B8C19 | |  |  | -7.174 |
| 202 | A1B8C20 | |  |  | -14.924 |
| 203 | A1B8C21 | |  |  | -9.277 |
| 204 | A1B8C22 | |  |  | -5.316 |
| 205 | A1B8C23 | |  |  | -5.256 |
| 206 | A1B8C24 | |  | | 0.508 |

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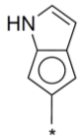
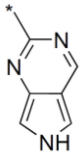
| 207 | A1B8C25 | |  |  | 16.677 |
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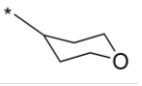
| 209 | A2B1C2 | | | | 7.521 |
| 210 | A2B1C3 | | | | 3.491 |
| 211 | A2B1C4 | | | | 12.41 |
| 212 | A2B1C5 | | | | 8.394 |
| 213 | A2B1C6 | | | | -10.301 |
| 214 | A2B1C7 | | | | -13.005 |
| 215 | A2B1C8 | | | | -3.549 |
| 216 | A2B1C9 | | |  | -11.33 |
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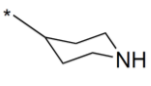
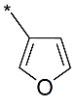
| 218 | A2B1C11 | | |  | -10.068 |
| 219 | A2B1C12 | | |  | 0.693 |
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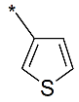
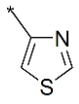
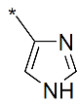
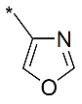
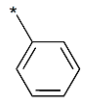
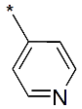
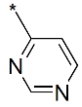
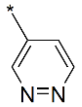
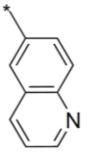
| 221 | A2B1C14 | | |  | 1.963 |
| 222 | A2B1C15 | | |  | 17.674 |
| 223 | A2B1C16 | | |  | 19.102 |
| 224 | A2B1C17 | | |  | 16.735 |
| 225 | A2B1C18 | | |  | 5.574 |
| 226 | A2B1C19 | | |  | -2.394 |
| 227 | A2B1C20 | | |  | -9.74 |
| 228 | A2B1C21 | | |  | -4.763 |
| 229 | A2B1C22 | | |  | -0.139 |
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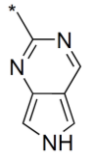
| 231 | A2B1C24 | | | | 5.808 |
| 232 | A2B1C25 | | |  | 21.234 |

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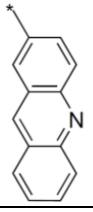
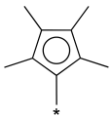
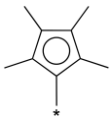
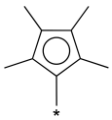
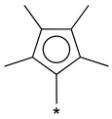
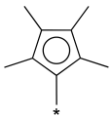
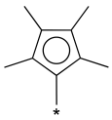
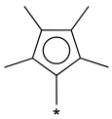
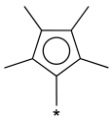
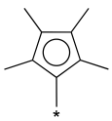
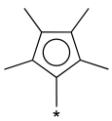
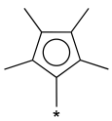
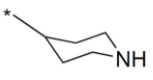
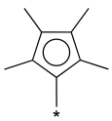
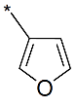
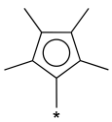
| 233 | A2B1C26 | | |  | 2.63 |
| 234 | A2B2C1 | |  | | 5.522 |
| 235 | A2B2C2 | |  | | 8.279 |
| 236 | A2B2C3 | |  | | 3.656 |
| 237 | A2B2C4 | |  | | 11.699 |
| 238 | A2B2C5 | |  | | 8.052 |
| 239 | A2B2C6 | |  | | -6.613 |
| 240 | A2B2C7 | |  | | -3.943 |
| 241 | A2B2C8 | |  | | -1.431 |
| 242 | A2B2C9 | |  | | -5.289 |
| 243 | A2B2C10 | |  | | -4.568 |
| 244 | A2B2C11 | |  |  | -3.965 |
| 245 | A2B2C12 | |  |  | 2.803 |
| 246 | A2B2C13 | |  | | 2.046 |

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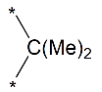
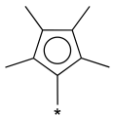
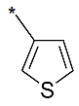
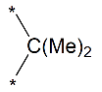
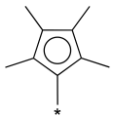
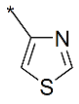
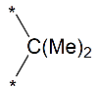
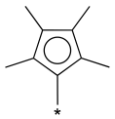
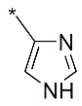
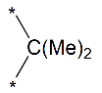
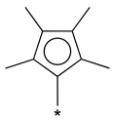
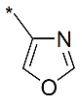
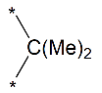
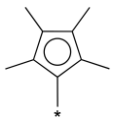
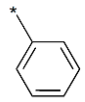
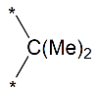
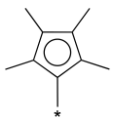
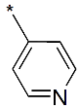
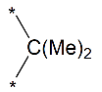
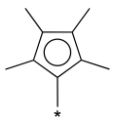
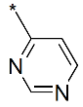
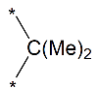
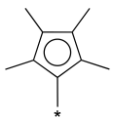
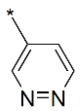
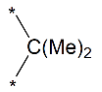
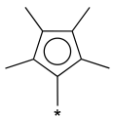
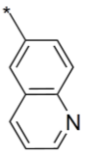
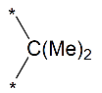
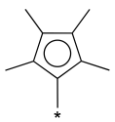
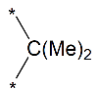
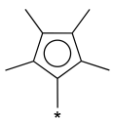
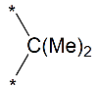
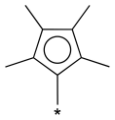
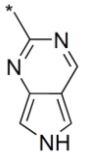
| 247 | A2B2C14 |  |  |  | 3.94 |
| 248 | A2B2C15 |  |  |  | 20.023 |
| 249 | A2B2C16 |  |  |  | 21.581 |
| 250 | A2B2C17 |  |  |  | 19.547 |
| 251 | A2B2C18 |  |  |  | 2.297 |
| 252 | A2B2C19 |  |  |  | 0.462 |
| 253 | A2B2C20 |  |  |  | -2.901 |
| 254 | A2B2C21 |  |  |  | -0.555 |
| 255 | A2B2C22 |  |  |  | 1.087 |
| 256 | A2B2C23 |  |  | | 1.678 |
| 257 | A2B2C24 |  |  | | 6.852 |
| 258 | A2B2C25 |  |  |  | 21.46 |

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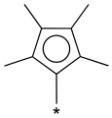
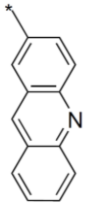
| 259 | A2B2C26 | |  |  | 2.679 |
| 260 | A2B3C1 | | | | 2.846 |
| 261 | A2B3C2 | | | | 5.284 |
| 262 | A2B3C3 | | | | 2.924 |
| 263 | A2B3C4 | | | | 10.944 |
| 264 | A2B3C5 | | | | 5.939 |
| 265 | A2B3C6 | | | | -11.528 |
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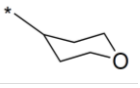
| 267 | A2B3C8 | | | | -8.043 |
| 268 | A2B3C9 | | |  | -13.376 |

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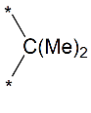
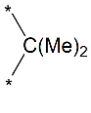
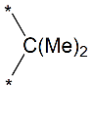
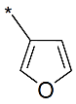
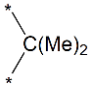
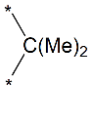
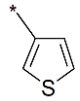
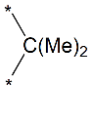
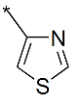
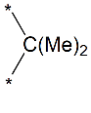
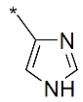
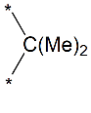
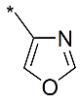
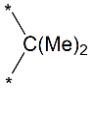
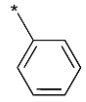
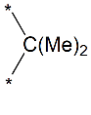
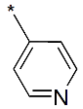
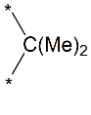
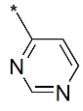
| 269 | A2B3C10 |  | | | -8.654 |
| 270 | A2B3C11 |  | | | -12.159 |
| 271 | A2B3C12 |  | |  | -0.625 |
| 272 | A2B3C13 |  | | | 1.535 |
| 273 | A2B3C14 |  | |  | -0.044 |
| 274 | A2B3C15 |  | |  | 14.716 |
| 275 | A2B3C16 |  | |  | 16.168 |
| 276 | A2B3C17 |  | |  | 13.923 |
| 277 | A2B3C18 |  | |  | -2.428 |
| 278 | A2B3C19 |  | |  | -4.381 |
| 279 | A2B3C20 |  | |  | -11.673 |

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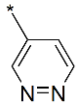
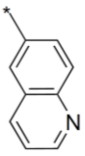
| 280 | A2B3C21 | | |  | -6.185 |
| 281 | A2B3C22 | | |  | -2.199 |
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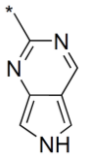
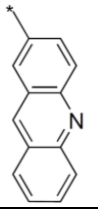
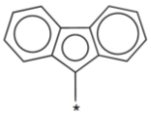
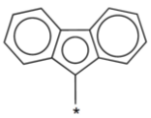
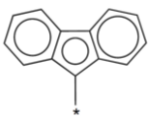
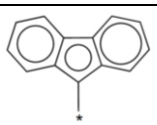
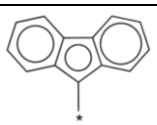
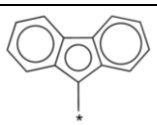
| 283 | A2B3C24 | | | | 3.562 |
| 284 | A2B3C25 | | |  | 18.546 |
| 285 | A2B3C26 | | |  | -0.661 |
| 286 | A2B4C1 | |  | | 5.798 |
| 287 | A2B4C2 | |  | | 8.646 |
| 288 | A2B4C3 | |  | | 7.396 |
| 289 | A2B4C4 | |  | | 13.857 |
| 290 | A2B4C5 | |  | | 7.485 |
| 291 | A2B4C6 | |  | | 0.838 |

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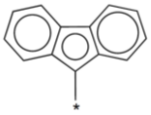
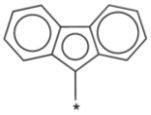
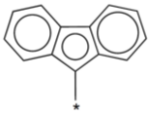
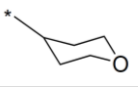
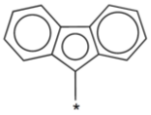
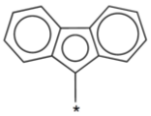
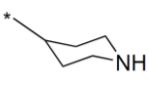
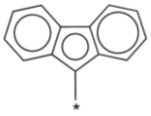
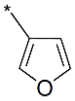
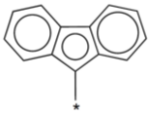
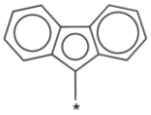
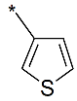
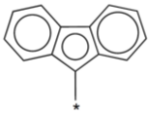
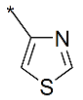
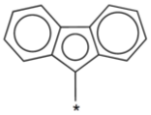
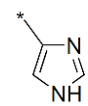
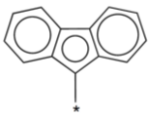
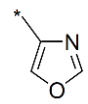
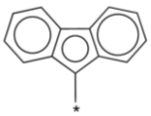
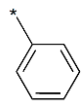
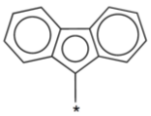
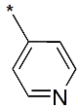
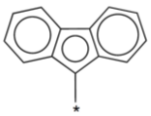
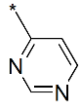
| 292 | A2B4C7 | |  | | -4.925 |
| 293 | A2B4C8 | |  | | -6.825 |
| 294 | A2B4C9 | |  |  | 0.8 |
| 295 | A2B4C10 | |  | | 1.997 |
| 296 | A2B4C11 | |  |  | 2.322 |
| 297 | A2B4C12 | |  |  | 4.335 |
| 298 | A2B4C13 | |  | | 0.171 |
| 299 | A2B4C14 | |  |  | 5.028 |
| 300 | A2B4C15 | |  |  | 15.693 |
| 301 | A2B4C16 | |  |  | 16.646 |
| 302 | A2B4C17 | |  |  | 15.472 |
| 303 | A2B4C18 | |  |  | 3.889 |
| 304 | A2B4C19 | |  |  | 2.497 |
| 305 | A2B4C20 | |  |  | -0.723 |

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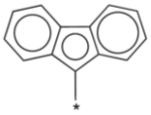
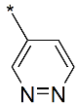
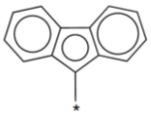
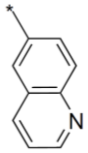
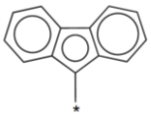
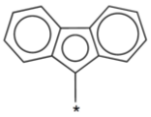
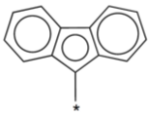
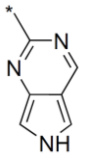
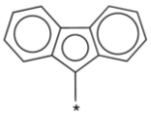
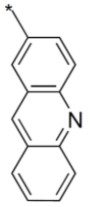
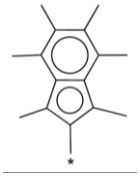
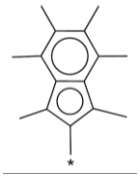
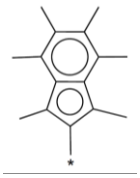
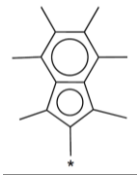
| 306 | A2B4C21 | |  |  | 1.855 |
| 307 | A2B4C22 | |  |  | 3.163 |
| 308 | A2B4C23 | |  | | 4.046 |
| 309 | A2B4C24 | |  | | 8.456 |
| 310 | A2B4C25 | |  |  | 14.248 |
| 311 | A2B4C26 | |  |  | 4.179 |
| 312 | A2B5C1 | |  | | 3.122 |
| 313 | A2B5C2 | |  | | 4.884 |
| 314 | A2B5C3 | |  | | 1.344 |
| 315 | A2B5C4 | |  | | 8.362 |

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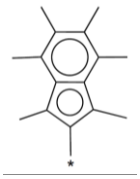
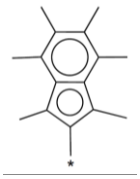
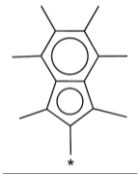
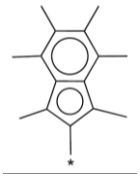
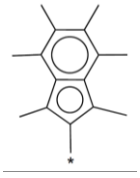
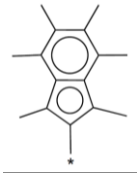
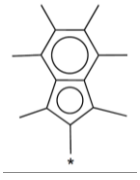
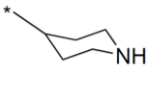
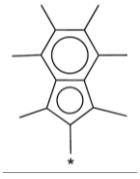
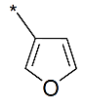
| 316 | A2B5C5 | |  | | 2.354 |
| 317 | A2B5C6 | |  | | -5.833 |
| 318 | A2B5C7 | |  | | -11.278 |
| 319 | A2B5C8 | |  | | -8.413 |
| 320 | A2B5C9 | |  | | -4.111 |
| 321 | A2B5C10 | |  | | -4.152 |
| 322 | A2B5C11 | |  |  | -2.828 |
| 323 | A2B5C12 | |  |  | 1.206 |

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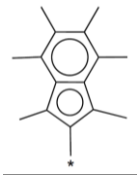
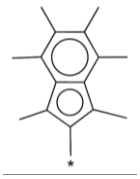
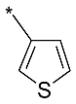
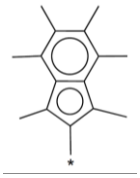
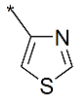
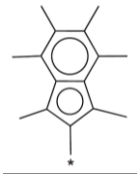
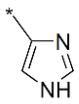
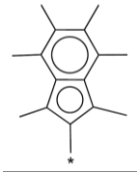
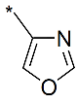
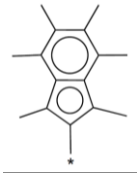
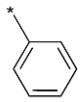
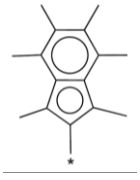
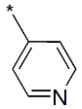
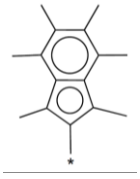
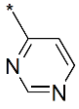
| 324 | A2B5C13 | |  | | -1.867 |
| 325 | A2B5C14 | |  |  | 1.846 |
| 326 | A2B5C15 | |  |  | 18.816 |
| 327 | A2B5C16 | |  |  | 19.795 |
| 328 | A2B5C17 | |  |  | 18.127 |
| 329 | A2B5C18 | |  |  | 0.381 |
| 330 | A2B5C19 | |  |  | -1.38 |
| 331 | A2B5C20 | |  |  | 2.322 |

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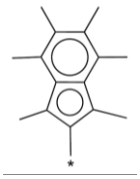
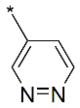
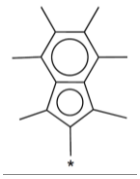
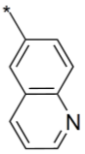
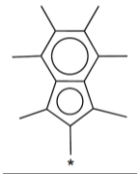
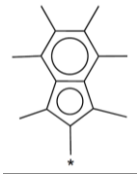
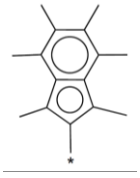
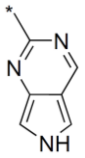
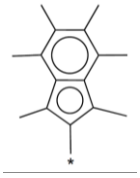
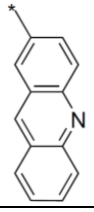
| 332 | A2B5C21 | |  |  | -2.038 |
| 333 | A2B5C22 | |  |  | -1.045 |
| 334 | A2B5C23 | |  | | -0.49 |
| 335 | A2B5C24 | |  | | 3.663 |
| 336 | A2B5C25 | |  |  | 20.565 |
| 337 | A2B5C26 | |  |  | 0.074 |
| 338 | A2B6C1 | | | | 4.94 |
| 339 | A2B6C2 | | | | 8.089 |
| 340 | A2B6C3 | | | | 3.954 |

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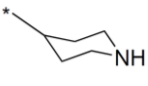
| 341 | A2B6C4 | | | | 13.592 |
| 342 | A2B6C5 | | | | 9.714 |
| 343 | A2B6C6 | | | | -10.713 |
| 344 | A2B6C7 | | | | -11.808 |
| 345 | A2B6C8 | | | | -4.927 |
| 346 | A2B6C9 | | | | -11.633 |
| 347 | A2B6C10 | | | | -11.704 |
| 348 | A2B6C11 | | |  | -10.447 |
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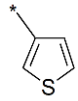
| 350 | A2B6C13 | | | | 5.725 |
| 351 | A2B6C14 | | |  | 2.351 |

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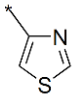
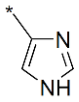
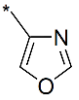
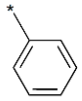
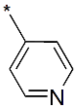
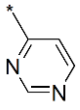
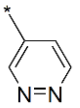
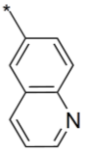
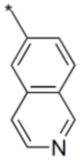
| 352 | A2B6C15 | | |  | 13.505 |
| 353 | A2B6C16 | | |  | 14.835 |
| 354 | A2B6C17 | | |  | 12.614 |
| 355 | A2B6C18 | | |  | 0.412 |
| 356 | A2B6C19 | | |  | -1.996 |
| 357 | A2B6C20 | | |  | -10.437 |
| 358 | A2B6C21 | | |  | -4.725 |
| 359 | A2B6C22 | | |  | 0.397 |
| 360 | A2B6C23 | | |  | 0.421 |
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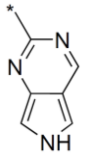
| 362 | A2B6C25 | | |  | 16.025 |

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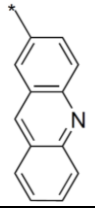
| 363 | A2B6C26 | | |  | 3.747 |
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| 365 | A2B7C2 | | | | 8.591 |
| 366 | A2B7C3 | | | | 3.95 |
| 367 | A2B7C4 | | | | 13.219 |
| 368 | A2B7C5 | | | | 9.384 |
| 369 | A2B7C6 | | | | -9.802 |
| 370 | A2B7C8 | | | | -5.767 |
| 371 | A2B7C9 | | | | -10.733 |
| 372 | A2B7C10 | | | | -11.289 |

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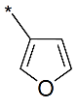
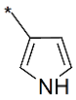
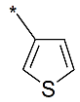
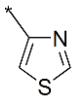
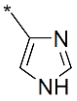
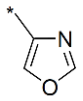
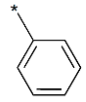
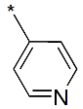
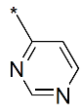
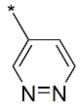
| 374 | A2B7C12 | | |  | 1.021 |
| 375 | A2B7C13 | | |  | 4.701 |
| 376 | A2B7C14 | | |  | 2.172 |
| 377 | A2B7C15 | | |  | 14.219 |
| 378 | A2B7C16 | | |  | 15.825 |
| 379 | A2B7C17 | | |  | 13.412 |
| 380 | A2B7C18 | | |  | 0.384 |
| 381 | A2B7C19 | | |  | -2.065 |
| 382 | A2B7C20 | | |  | -9.614 |
| 383 | A2B7C21 | | |  | -4.619 |

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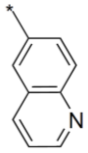
| 384 | A2B7C22 | | |  | 0.39 |
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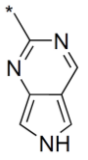
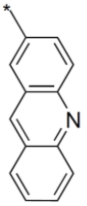
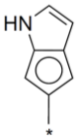
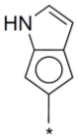
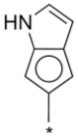
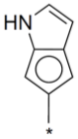
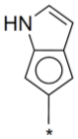
| 387 | A2B7C25 | | |  | 16.812 |
| 388 | A2B7C26 | | |  | 3.997 |
| 389 | A2B8C1 | |  | | 6.36 |
| 390 | A2B8C2 | |  | | 8.855 |
| 391 | A2B8C3 | |  | | 4.688 |
| 392 | A2B8C4 | |  | | 14.956 |
| 393 | A2B8C5 | |  | | 9.098 |

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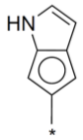
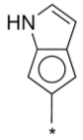
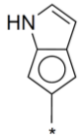
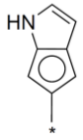
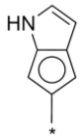
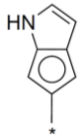
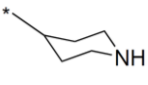
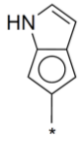
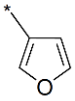
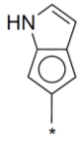
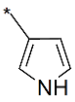
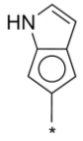
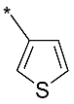
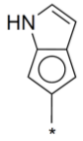
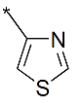
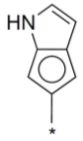
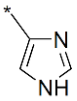
| 394 | A2B8C6 | |  | | -8.953 |
| 395 | A2B8C7 | |  | | -11.098 |
| 396 | A2B8C8 | |  | | -6.157 |
| 397 | A2B8C9 | |  | | -6.239 |
| 398 | A2B8C10 | |  | | -5.014 |
| 399 | A2B8C11 | |  |  | -4.811 |
| 400 | A2B8C12 | |  |  | 2.658 |
| 401 | A2B8C13 | |  |  | 4.026 |
| 402 | A2B8C14 | |  |  | 2.207 |
| 403 | A2B8C15 | |  |  | 15.465 |
| 404 | A2B8C16 | |  |  | 16.984 |

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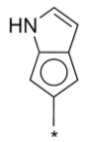
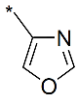
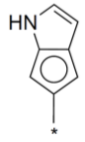
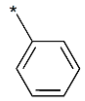
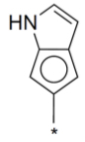
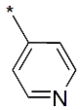
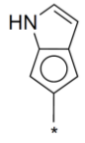
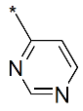
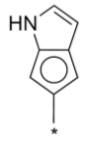
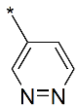
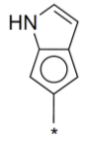
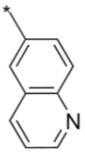
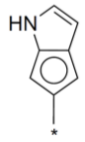
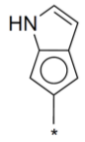
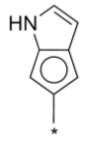
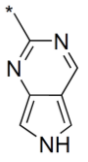
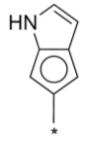
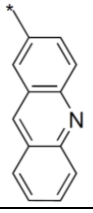
| 405 | A2B8C17 | |  |  | 14.828 |
| 406 | A2B8C18 | |  |  | 0.206 |
| 407 | A2B8C19 | |  |  | -1.583 |
| 408 | A2B8C20 | |  |  | -8.487 |
| 409 | A2B8C21 | |  |  | -3.92 |
| 410 | A2B8C22 | |  |  | 0.173 |
| 411 | A2B8C23 | |  | | 0.415 |
| 412 | A2B8C24 | |  | | 5.948 |
| 413 | A2B8C25 | |  |  | 17.546 |
| 414 | A2B8C26 | |  |  | 4.2 |
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| 417 | A3B1C3 | | | | 0.898 |
| 418 | A3B1C4 | | | | 9.207 |
| 419 | A3B1C5 | | | | 4.637 |
| 420 | A3B1C6 | | | | -12.335 |
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| 422 | A3B1C8 | | | | -7.803 |
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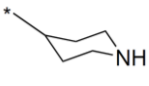
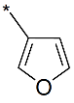
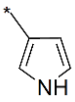
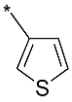
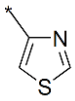
| 425 | A3B1C11 | | |  | -13.677 |
| 426 | A3B1C12 | | |  | -3.786 |
| 427 | A3B1C13 | | |  | -0.417 |
| 428 | A3B1C14 | | |  | -2.293 |
| 429 | A3B1C15 | | |  | 20.893 |

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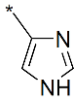
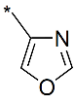
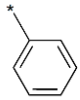
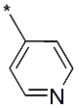
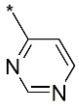
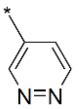
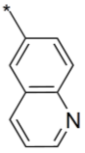
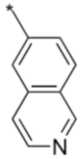
| 430 | A3B1C16 | | |  | 22.289 |
| 431 | A3B1C17 | | |  | 19.886 |
| 432 | A3B1C18 | | |  | -4.497 |
| 433 | A3B1C19 | | |  | -6.473 |
| 434 | A3B1C20 | | |  | -14.718 |
| 435 | A3B1C21 | | |  | -9.242 |
| 436 | A3B1C22 | | |  | -5.195 |
| 437 | A3B1C23 | | |  | -5.196 |
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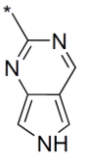
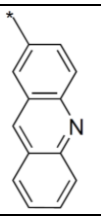
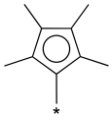
| 439 | A3B1C25 | | |  | 15.74 |
| 440 | A3B1C26 | | |  | -2.816 |
| 441 | A3B2C1 | |  | | 6.504 |

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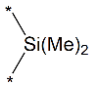
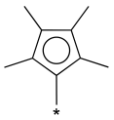
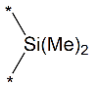
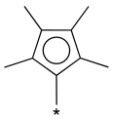
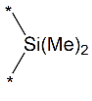
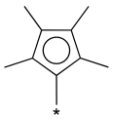
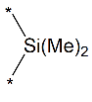
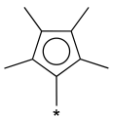
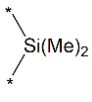
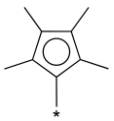
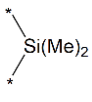
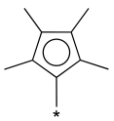
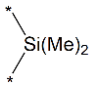
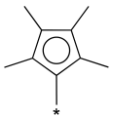
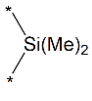
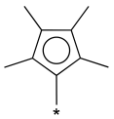
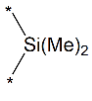
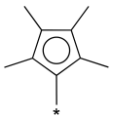
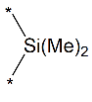
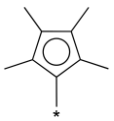
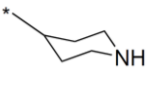
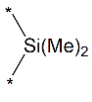
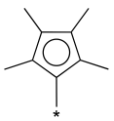
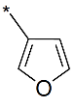
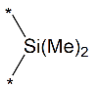
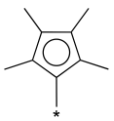
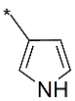
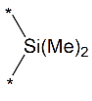
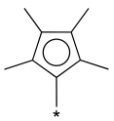
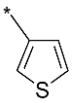
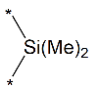
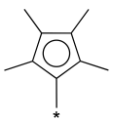
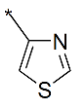
| 442 | A3B2C2 |  |  | | 8.066 |
| 443 | A3B2C3 |  |  | | 1.925 |
| 444 | A3B2C4 |  |  | | 8.71 |
| 445 | A3B2C5 |  |  | | 4.265 |
| 446 | A3B2C6 |  |  | | -9.734 |
| 447 | A3B2C7 |  |  | | -6.809 |
| 448 | A3B2C8 |  |  | | -7.509 |
| 449 | A3B2C9 |  |  | | -9.137 |
| 450 | A3B2C10 |  |  | | -8.134 |
| 451 | A3B2C11 |  |  |  | -7.542 |
| 452 | A3B2C12 |  |  |  | -2.145 |
| 453 | A3B2C13 |  |  |  | -0.072 |
| 454 | A3B2C14 |  |  |  | -1.261 |
| 455 | A3B2C15 |  |  |  | 22.988 |

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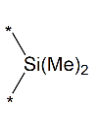
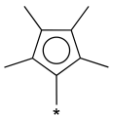
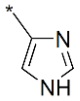
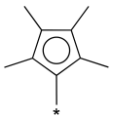
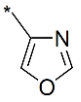
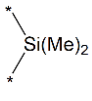
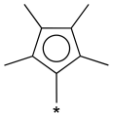
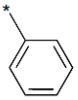
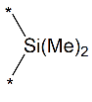
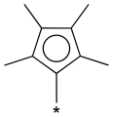
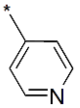
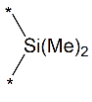
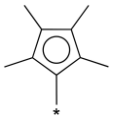
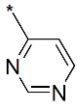
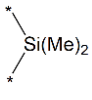
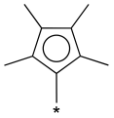
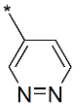
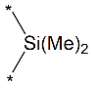
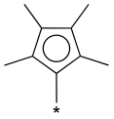
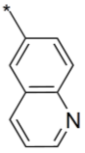
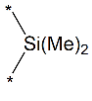
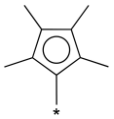
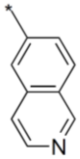
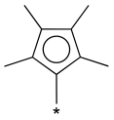
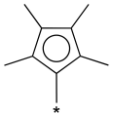
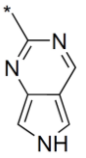
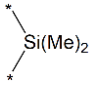
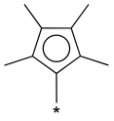
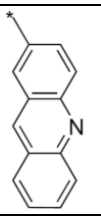
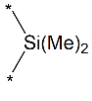
| 456 | A3B2C16 |  |  |  | 24.979 |
| 457 | A3B2C17 | |  |  | 22.851 |
| 458 | A3B2C18 |  |  |  | -2.786 |
| 459 | A3B2C19 |  |  |  | -4.346 |
| 460 | A3B2C20 |  |  |  | -12.727 |
| 461 | A3B2C21 |  |  |  | -5.161 |
| 462 | A3B2C22 |  |  |  | -3.962 |
| 463 | A3B2C23 |  |  |  | -3.902 |
| 464 | A3B2C24 | |  | | 0.966 |
| 465 | A3B2C25 | |  |  | 15.671 |
| 466 | A3B2C26 |  |  |  | -2.611 |
| 467 | A3B3C1 |  | | | 2.41 |

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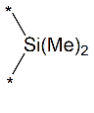
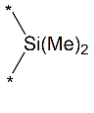
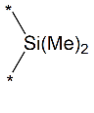
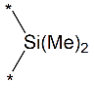
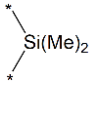
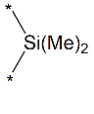
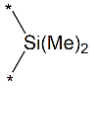
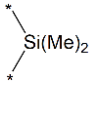
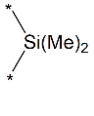
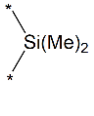
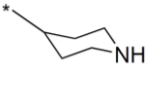
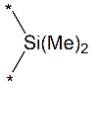
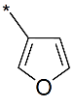
| 468 | A3B3C2 |  | | | 4.276 |
| 469 | A3B3C3 |  | | | -0.971 |
| 470 | A3B3C4 |  | | | 7.075 |
| 471 | A3B3C5 |  | | | 1.738 |
| 472 | A3B3C6 |  | | | -17.476 |
| 473 | A3B3C7 |  | | | -7.229 |
| 474 | A3B3C8 |  | | | -12.642 |
| 475 | A3B3C9 |  | | | -17.27 |
| 476 | A3B3C10 |  | | | -12.101 |
| 477 | A3B3C11 |  | |  | -16.018 |
| 478 | A3B3C12 |  | |  | -6.201 |

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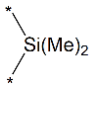
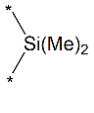
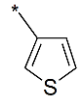
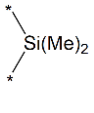
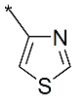
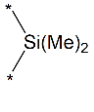
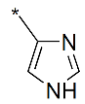
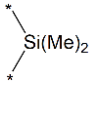
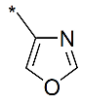
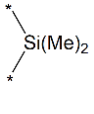
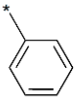
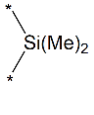
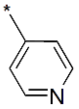
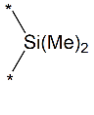
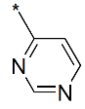
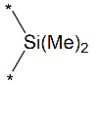
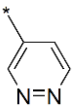
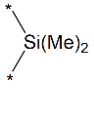
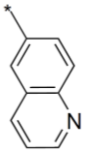
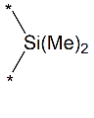
| 479 | A3B3C13 |  | | | -4.219 |
| 480 | A3B3C14 |  | |  | -5.686 |
| 481 | A3B3C15 |  | |  | 17.898 |
| 482 | A3B3C16 |  | |  | 19.662 |
| 483 | A3B3C17 |  | |  | 17.43 |
| 484 | A3B3C18 |  | |  | -8.114 |
| 485 | A3B3C19 |  | |  | -9.874 |
| 486 | A3B3C20 |  | |  | -16.773 |
| 487 | A3B3C21 |  | |  | -11.537 |
| 488 | A3B3C22 |  | |  | -8.126 |
| 489 | A3B3C23 |  | | | -8.485 |

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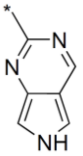
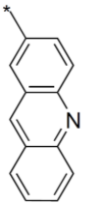
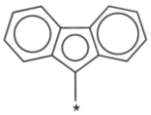
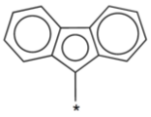
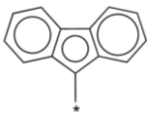
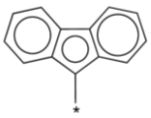
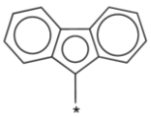
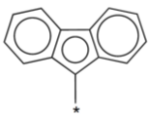
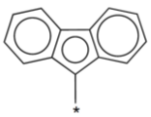
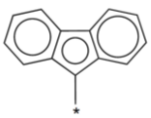
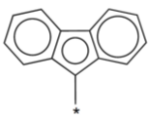
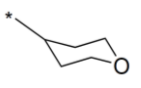
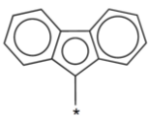
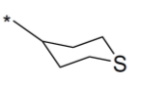
| 491 | A3B3C25 | | |  | 13.293 |
| 492 | A3B3C26 | | |  | -6.81 |
| 493 | A3B4C1 | |  | | 6.146 |
| 494 | A3B4C2 | |  | | 7.711 |
| 495 | A3B4C3 | |  | | 4.794 |
| 496 | A3B4C4 | |  | | 11.353 |
| 497 | A3B4C5 | |  | | 2.686 |
| 498 | A3B4C6 | |  | | -5.957 |
| 499 | A3B4C7 | |  | | -8.51 |
| 500 | A3B4C8 | |  | | -13.8 |
| 501 | A3B4C9 | |  |  | -5.177 |
| 502 | A3B4C10 | |  |  | -5.792 |

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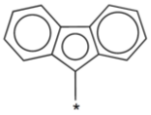
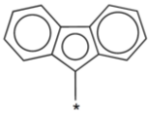
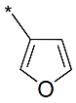
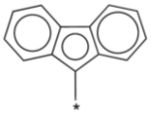
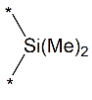
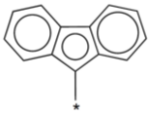
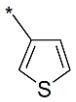
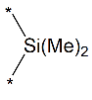
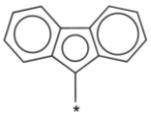
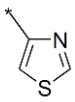
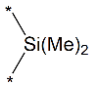
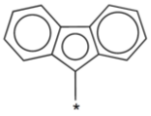
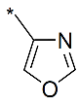
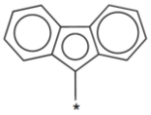
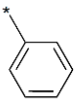
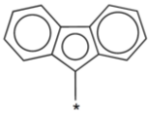
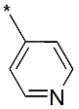
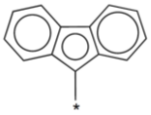
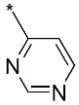
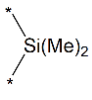
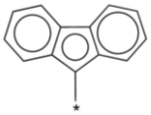
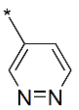
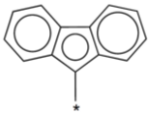
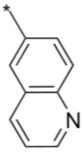
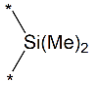
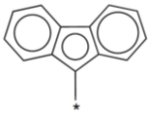
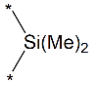
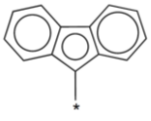
| 503 | A3B4C11 | |  | | -5.188 |
| 504 | A3B4C12 | |  |  | -1.854 |
| 505 | A3B4C13 | |  | | -6.052 |
| 506 | A3B4C14 |  |  |  | -2.012 |
| 507 | A3B4C15 |  |  |  | 15.867 |
| 508 | A3B4C17 |  |  |  | 16.179 |
| 509 | A3B4C18 | |  |  | -4.131 |
| 510 | A3B4C19 | |  |  | -5.137 |
| 511 | A3B4C20 | |  |  | -9.367 |
| 512 | A3B4C21 |  |  |  | -2.634 |
| 513 | A3B4C22 | |  |  | -4.419 |
| 514 | A3B4C23 |  |  | | -4.661 |
| 515 | A3B4C24 |  |  | | -0.536 |

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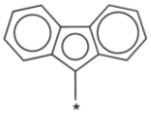
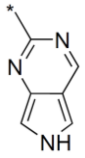
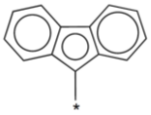
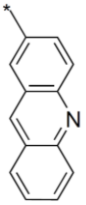
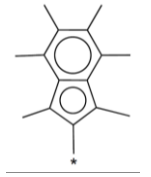
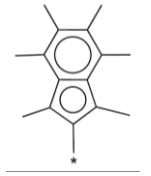
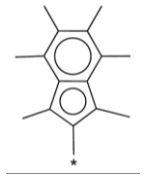
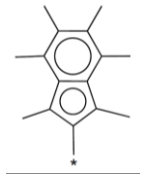
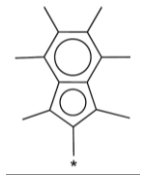
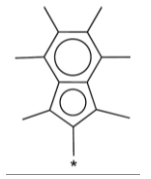
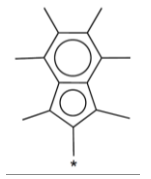
| 516 | A3B4C25 | |  |  | 12.073 |
| 517 | A3B4C26 | |  |  | -1.606 |
| 518 | A3B5C1 | |  | | 3.274 |
| 519 | A3B5C2 | |  | | 4.127 |
| 520 | A3B5C3 | |  | | -0.446 |
| 521 | A3B5C4 | |  | | 8.214 |
| 522 | A3B5C5 | |  | | -0.014 |
| 523 | A3B5C6 | |  | | -10.425 |
| 524 | A3B5C7 | |  | | -11.29 |

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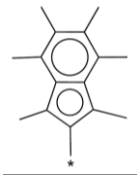
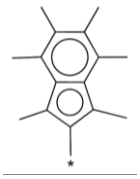
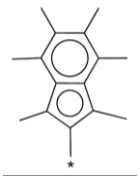
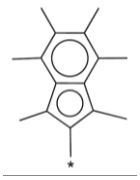
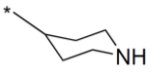
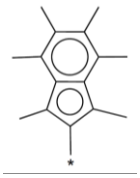
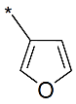
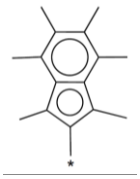
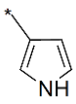
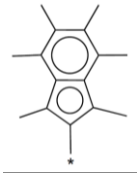
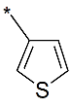
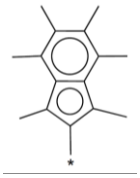
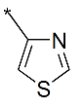
| 525 | A3B5C8 | |  | | -13.026 |
| 526 | A3B5C9 | |  | | -9.226 |
| 527 | A3B5C10 | |  | | -8.853 |
| 528 | A3B5C11 | |  |  | -8.316 |
| 529 | A3B5C12 | |  |  | -4.029 |
| 530 | A3B5C13 | |  |  | -2.46 |
| 531 | A3B5C14 | |  |  | -3.633 |
| 532 | A3B5C15 | |  |  | 21.853 |

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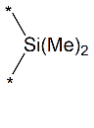
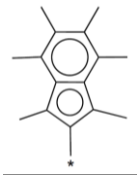
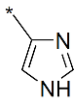
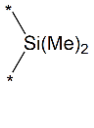
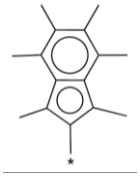
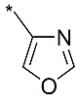
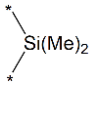
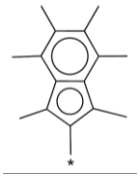
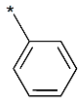
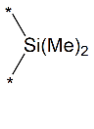
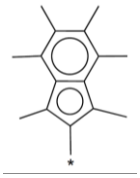
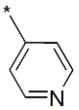
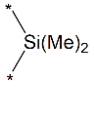
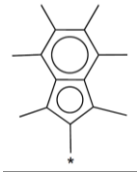
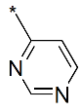
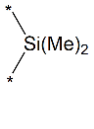
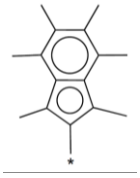
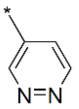
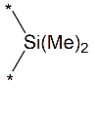
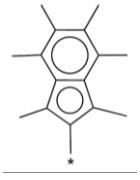
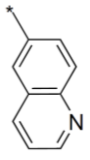
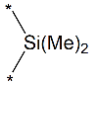
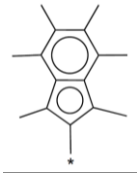
| 533 | A3B5C16 |  |  |  | 23.692 |
| 534 | A3B5C17 |  |  |  | 21.911 |
| 535 | A3B5C18 |  |  |  | -5.432 |
| 536 | A3B5C19 |  |  |  | -7.293 |
| 537 | A3B5C20 |  |  |  | -11.943 |
| 538 | A3B5C21 |  |  |  | -8.197 |
| 539 | A3B5C22 |  |  |  | -6.791 |
| 540 | A3B5C23 |  |  | | -6.069 |

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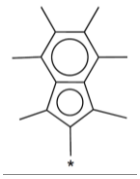
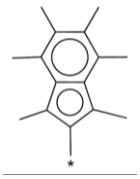
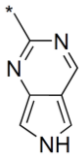
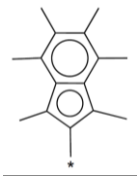
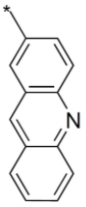
| 541 | A3B5C24 | |  | | -3.63 |
| 542 | A3B5C25 | |  |  | 15.434 |
| 543 | A3B5C26 | |  |  | -5.498 |
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| 548 | A3B6C5 | | | | 5.496 |
| 549 | A3B6C6 | | | | -14.64 |
| 550 | A3B6C7 | | | | -13.983 |

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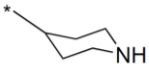
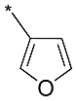
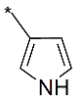
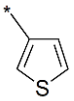
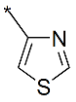
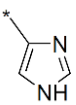
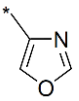
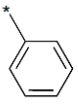
| 554 | A3B6C11 | | |  | -14.15 |
| 555 | A3B6C12 | | |  | -4.737 |
| 556 | A3B6C13 | | |  | -0.137 |
| 557 | A3B6C14 | | |  | -3.108 |
| 558 | A3B6C15 | | |  | 16.455 |
| 559 | A3B6C16 | | |  | 18.159 |
| 560 | A3B6C17 | | |  | 15.703 |
| 561 | A3B6C18 | | |  | -5.366 |

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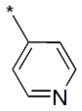
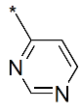
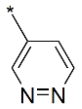
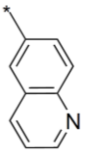
| 562 | A3B6C19 | | |  | -7.355 |
| 563 | A3B6C20 | | |  | -15.604 |
| 564 | A3B6C21 | | |  | -9.612 |
| 565 | A3B6C22 | | |  | -5.284 |
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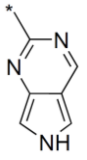
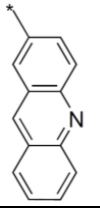
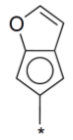
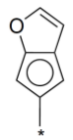
| 568 | A3B6C25 | | |  | 16.693 |
| 569 | A3B6C26 | | |  | -3.516 |
| 570 | A3B7C1 | |  | | 4.844 |
| 571 | A3B7C2 | |  | | 7.12 |

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| 577 | A3B7C8 | | | | -10.649 |
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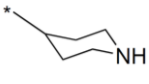
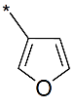
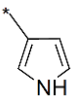
| 580 | A3B7C11 | | |  | -14.128 |
| 581 | A3B7C12 | | |  | -4.977 |
| 582 | A3B7C13 | | |  | -1.817 |

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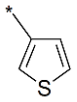
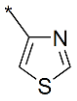
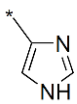
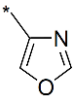
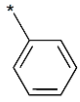
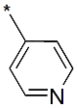
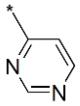
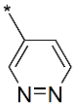
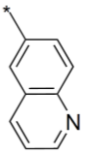
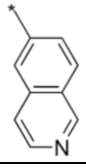
| 583 | A3B7C14 | | |  | -3.611 |
| 584 | A3B7C15 | | |  | 17.002 |
| 585 | A3B7C16 | | |  | 18.835 |
| 586 | A3B7C17 | | |  | 16.561 |
| 587 | A3B7C18 | | |  | -5.859 |
| 588 | A3B7C19 | | |  | -7.99 |
| 589 | A3B7C20 | | |  | -15.359 |
| 590 | A3B7C21 | | |  | -9.595 |
| 591 | A3B7C22 | | |  | -6.455 |
| 592 | A3B7C23 | | |  | -6.415 |
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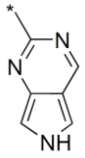
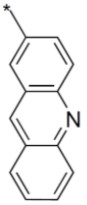
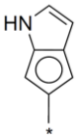
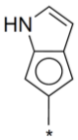
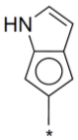
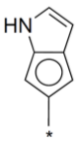
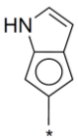
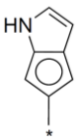
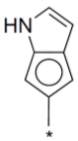
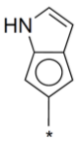
| 594 | A3B7C25 | | |  | 17.041 |
| 595 | A3B7C26 | | |  | -3.261 |
| 596 | A3B8C1 | |  | | 5.171 |
| 597 | A3B8C2 | |  | | 7.168 |
| 598 | A3B8C3 | |  | | 0.623 |
| 599 | A3B8C4 | |  | | 9.433 |
| 600 | A3B8C5 | |  | | 4.43 |
| 601 | A3B8C6 | |  | | -15.771 |
| 602 | A3B8C7 | |  | | -13.33 |
| 603 | A3B8C8 | |  | | -11.301 |

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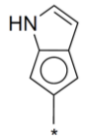
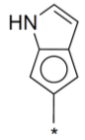
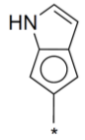
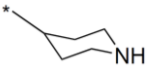
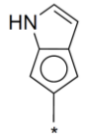
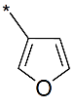
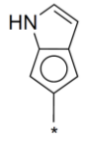
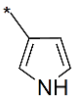
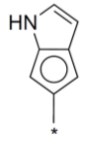
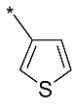
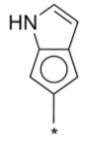
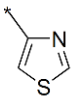
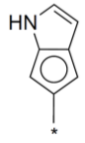
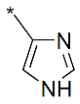
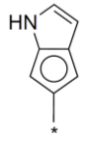
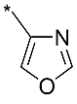
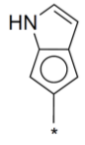
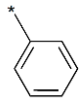
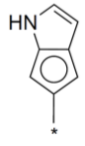
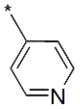
| 604 | A3B8C9 | |  | | -9.96 |
| 605 | A3B8C10 | |  | | -10.131 |
| 606 | A3B8C11 | |  |  | -9.658 |
| 607 | A3B8C12 | |  |  | -2.711 |
| 608 | A3B8C13 | |  |  | -2.299 |
| 609 | A3B8C14 | |  |  | -3.667 |
| 610 | A3B8C15 | |  |  | 17.934 |
| 611 | A3B8C16 | |  |  | 19.648 |
| 612 | A3B8C17 | |  |  | 17.506 |
| 613 | A3B8C18 | |  |  | -5.969 |
| 614 | A3B8C19 | |  |  | -7.698 |

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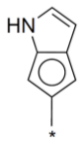
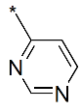
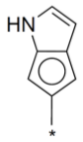
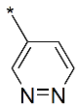
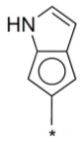
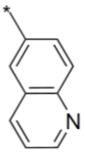
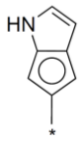
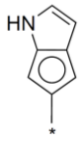
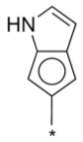
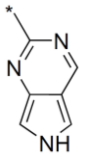
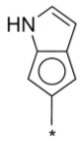
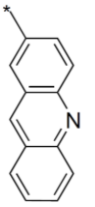
| 615 | A3B8C20 | |  |  | -14.436 |
| 616 | A3B8C21 | |  |  | -9.336 |
| 617 | A3B8C22 | |  |  | -5.82 |
| 618 | A3B8C23 | |  | | -6.29 |
| 619 | A3B8C24 | |  | | -1.182 |
| 620 | A3B8C25 | |  |  | 17.669 |
| 621 | A3B8C26 | |  |  | -3.301 |

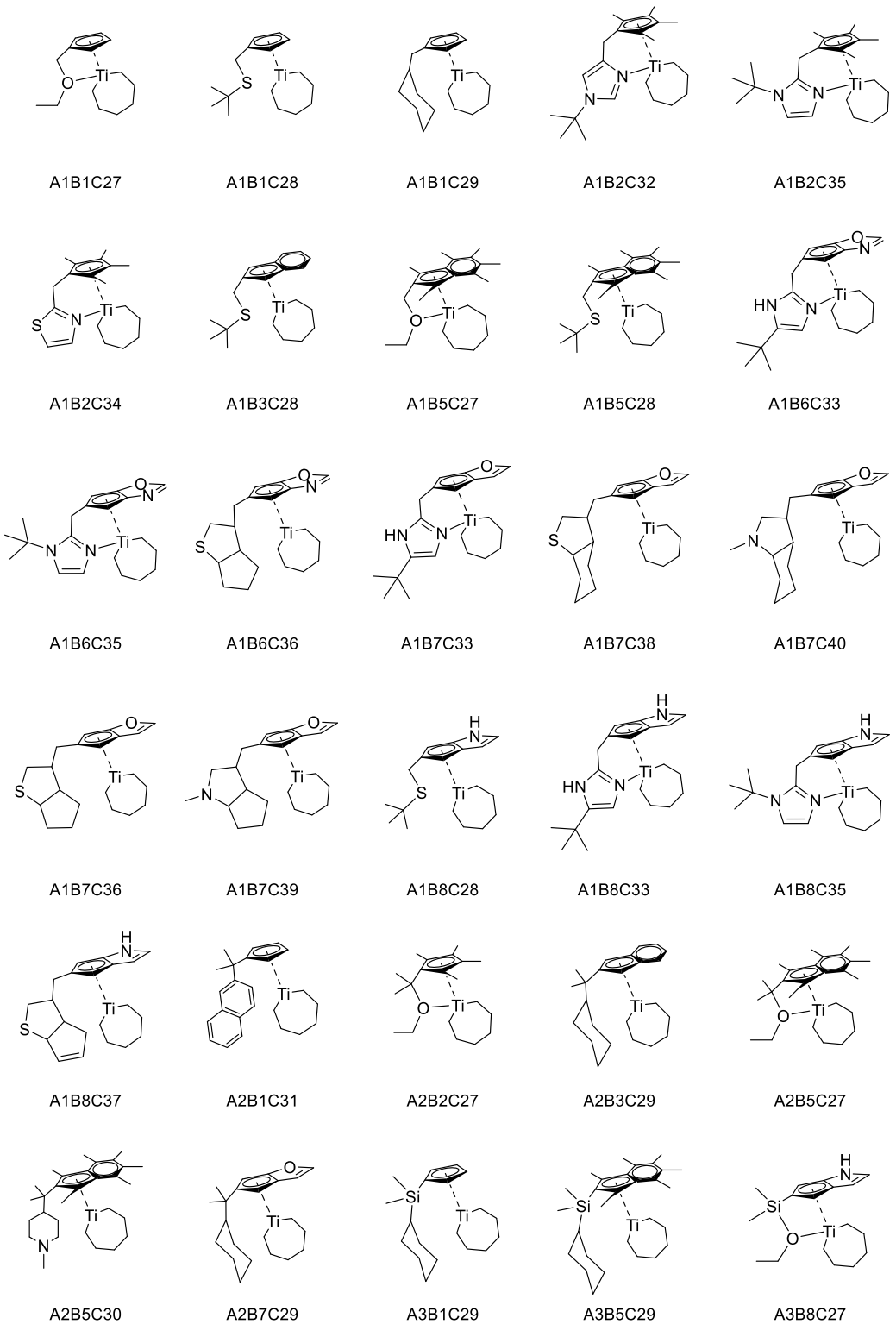



**Figure S1.** Different ligand structure catalysts intermediates in the external validation set.

**Table S3.** The energy barrier difference $\Delta\Delta G$ of the external validation set. $\Delta\Delta G$ equals the fourth ethylene insertion energy barrier $\Delta G_{\text{ethylene insertion}}$ minus the $\beta$-hydrogen transfer (after the third ethylene insertion) energy barrier $\Delta G_{\beta-\text{H transfer}}$.

| Index | ID (structures shown in Figure S1) | The energy barrier difference $\Delta\Delta G$ (kcal/mol) |
|---|---|---|
| 1 | A1B1C27 | -2.066 |
| 2 | A1B1C28 | 1.008 |
| 3 | A1B1C29 | -15.508 |
| 4 | A1B2C32 | 21.240 |
| 5 | A1B2C34 | 19.946 |
| 6 | A1B2C35 | 21.037 |
| 7 | A1B3C28 | 0.246 |
| 8 | A1B5C27 | 0.051 |
| 9 | A1B5C28 | 3.803 |
| 10 | A1B6C33 | 15.009 |
| 11 | A1B6C35 | 15.543 |
| 12 | A1B6C36 | -19.597 |
| 13 | A1B7C33 | 15.766 |
| 14 | A1B7C36 | -19.578 |
| 15 | A1B7C38 | -8.128 |
| 16 | A1B7C39 | -16.767 |
| 17 | A1B7C40 | -7.053 |
| 18 | A1B8C28 | 1.983 |
| 19 | A1B8C33 | 16.988 |
| 20 | A1B8C35 | 17.688 |
| 21 | A1B8C37 | -12.518 |
| 22 | A2B1C31 | 0.987 |
| 23 | A2B2C27 | 7.535 |
| 24 | A2B3C29 | -8.386 |
| 25 | A2B5C27 | 1.496 |
| 26 | A2B5C30 | -3.086 |
| 27 | A2B7C29 | -9.971 |
| 28 | A3B1C29 | -12.694 |
| 29 | A3B5C29 | -8.745 |
| 30 | A3B8C27 | 5.326 |