

Table 1s Total energies (E , in a.u.) of 1*H*-, 2*H*-, and 4*H*- azide tautomers at different levels

		NAzTA	AAzTA	AzTA
B3LYP/6-31G*	$E(1H-AZ)$	-610.26050	-461.11944	-405.77667
	$E(2H-AZ)$	-610.25985	-461.11800	-405.77563
	$E(4H-AZ)$	-610.24425	-461.09863	-405.75784
B3LYP/6-311++G**	$E(1H-AZ)$	-610.43682	-461.25794	-405.89101
	$E(2H-AZ)$	-610.43556	-461.25580	-405.88977
	$E(4H-AZ)$	-610.41952	-461.23563	-405.87156
B3LYP/AUG-cc-PVDZ	$E(1H-AZ)$	-610.36000	-461.19781	-405.83890
	$E(2H-AZ)$	-610.35976	-461.19598	-405.83823
	$E(4H-AZ)$	-610.34484	-461.17682	-405.82106

Table 2s Total energies (E , in a.u.) in different solutions predicted at the B3LYP/6-311++G** level

		Benzene	Dichloromethane	Acetone	Methanol	Water
NAzTA	$E(1H-AZ)$	-610.44277	-610.44756	-610.44854	-610.44883	-610.44911
	$E(2H-AZ)$	-610.44592	-610.45216	-610.45346	-610.45384	-610.45422
	$E(4H-AZ)$	-610.43034	-610.43825	-610.43995	-610.44045	-610.44096
AAzTA	$E(1H-AZ)$	-461.26447	-461.26871	-461.26956	-461.26981	-461.27006
	$E(2H-AZ)$	-461.26483	-461.27103	-461.27233	-461.27271	-461.27309
	$E(4H-AZ)$	-461.24736	-461.25605	-461.25796	-461.25852	-461.25908
AzTA	$E(1H-AZ)$	-405.89620	-405.89959	-405.90027	-405.90047	-405.90067
	$E(2H-AZ)$	-405.89649	-405.90106	-405.90200	-405.90228	-405.90255
	$E(4H-AZ)$	-405.88180	-405.88938	-405.89102	-405.89150	-405.89198

Table 3s Cartesian coordinates (in Å) of 1*H*-, 2*H*-, and 4*H*- azide tautomers in gas phases obtained at different theoretical levels

(A) B3LYP/6-31G*

	1 <i>H</i> -NAzTA-AZ			2 <i>H</i> -NAzTA-AZ			4 <i>H</i> -NAzTA-AZ					
	X	Y	Z	X	Y	Z	X	Y	Z			
N	0.191652	-0.444792	0.000266	-0.236426	-0.449112	-0.000092	0.322326	-0.617830	-0.002457			
C	-1.014415	0.170436	-0.000121	0.964567	0.085568	-0.000025	1.656552	-0.280741	0.003659			
N	-1.020928	1.491215	-0.001197	0.909678	1.429432	0.000086	1.803758	1.021393	0.025416			
N	0.305925	1.751437	-0.001394	-0.375170	1.832854	-0.000330	0.535106	1.588244	0.008296			
C	1.011831	0.591969	-0.000299	-1.024086	0.662685	0.000123	-0.323011	0.606329	-0.008813			
N	2.396200	0.625182	-0.000105	-2.413767	0.658299	0.000011	-1.705565	0.800826	-0.002979			
N	2.962282	-0.488916	0.000625	-2.955521	-0.463494	0.000083	-2.429542	-0.204028	0.001751			
N	3.611099	-1.416813	0.001357	-3.592943	-1.402103	0.000079	-3.213413	-1.034777	0.006873			
N	-2.269748	-0.581093	0.000256	2.237362	-0.606434	0.000000	2.665123	-1.230827	-0.086750			
O	-3.306550	0.073852	0.003292	3.230454	0.129915	0.000050	-0.068319	-1.534409	-0.170295			
O	-2.176382	-1.802994	-0.002543	2.227681	-1.826410	-0.000003	2.655052	-1.915228	0.665266			
H	0.643592	2.705157	-0.002129	1.679545	2.086340	0.000177	3.567465	-0.764897	-0.115102			

<table border="0" style="width:100%; border:none;"> <tr> <td style="width:33%; text-align:center">1<i>H</i>-AAzTA-AZ</td> <td style="width:33%; text-align:center">2<i>H</i>-AAzTA-AZ</td> <td style="width:33%; text-align:center">4<i>H</i>-AAzTA-AZ</td> </tr> </table>										1 <i>H</i> -AAzTA-AZ	2 <i>H</i> -AAzTA-AZ	4 <i>H</i> -AAzTA-AZ
1 <i>H</i> -AAzTA-AZ	2 <i>H</i> -AAzTA-AZ	4 <i>H</i> -AAzTA-AZ										

	X	Y	Z	X	Y	Z	X	Y	Z
N	0.288552	-0.699277	-0.004544	0.254229	-0.675685	0.000075	0.322326	-0.617830	-0.002457
C	1.610905	-0.341715	-0.009979	1.547959	-0.392504	-0.005996	1.656552	-0.280741	0.003659
N	1.857625	0.965500	0.001371	1.728682	0.949538	0.004091	1.803758	1.021393	0.025416
N	0.583785	1.480086	0.002434	0.515460	1.598196	0.004303	0.535106	1.588244	0.008296
C	-0.313309	0.479134	0.002827	-0.312964	0.570043	-0.001264	-0.323011	0.606329	-0.008813
N	-1.675870	0.764036	0.010720	-1.692376	0.799096	0.004474	-1.705565	0.800826	-0.002979
N	-2.434414	-0.223424	0.001305	-2.412922	-0.211486	0.002974	-2.429542	-0.204028	0.001751
N	-3.242502	-1.020822	-0.005099	-3.199529	-1.033490	0.002524	-3.213413	-1.034777	0.006873
N	2.611750	-1.289300	-0.080673	2.574787	-1.311239	-0.080191	2.665123	-1.230827	-0.086750
H	2.350699	-2.181870	0.318245	2.578252	1.486840	-0.081906	-0.068319	-1.534409	-0.170295
H	3.512977	-0.957076	0.238866	2.242086	-2.255656	0.075697	2.655052	-1.915228	0.665266
H	0.428267	2.476838	0.007212	3.391369	-1.100929	0.482018	3.567465	-0.764897	-0.115102

	1H-AzTA-AZ			2H-AzTA-AZ			4H-AzTA-AZ		
	X	Y	Z	X	Y	Z	X	Y	Z
N	0.288552	-0.699277	-0.004544	0.254229	-0.675685	0.000075	0.322326	-0.617830	-0.002457
C	1.610905	-0.341715	-0.009979	1.547959	-0.392504	-0.005996	1.656552	-0.280741	0.003659
N	1.857625	0.965500	0.001371	1.728682	0.949538	0.004091	1.803758	1.021393	0.025416
N	0.583785	1.480086	0.002434	0.515460	1.598196	0.004303	0.535106	1.588244	0.008296
C	-0.313309	0.479134	0.002827	-0.312964	0.570043	-0.001264	-0.323011	0.606329	-0.008813
N	-1.675870	0.764036	0.010720	-1.692376	0.799096	0.004474	-1.705565	0.800826	-0.002979
N	-2.434414	-0.223424	0.001305	-2.412922	-0.211486	0.002974	-2.429542	-0.204028	0.001751
N	-3.242502	-1.020822	-0.005099	-3.199529	-1.033490	0.002524	-3.213413	-1.034777	0.006873
N	2.611750	-1.289300	-0.080673	2.574787	-1.311239	-0.080191	2.665123	-1.230827	-0.086750
H	2.350699	-2.181870	0.318245	2.578252	1.486840	-0.081906	-0.068319	-1.534409	-0.170295
H	3.512977	-0.957076	0.238866	2.242086	-2.255656	0.075697	2.655052	-1.915228	0.665266
H	0.428267	2.476838	0.007212	3.391369	-1.100929	0.482018	3.567465	-0.764897	-0.115102

(B) B3LYP/6-311++G**

	1H-AAzTA-AZ			2H-AAzTA-AZ			4H-AAzTA-AZ		
	X	Y	Z	X	Y	Z	X	Y	Z
N	0.193903	-0.437761	0.000836	0.234739	-0.441076	-0.000107	-0.159923	-0.349601	0.001039
C	-1.010817	0.175399	-0.000141	-0.961928	0.092979	-0.000003	1.066659	0.254258	-0.000430
N	-1.012432	1.490400	-0.001083	-0.905069	1.434659	0.000108	0.970106	1.551328	-0.000707
N	0.312095	1.753152	-0.001407	0.380371	1.830689	-0.000318	-0.361762	1.851727	-0.000195
C	1.015628	0.593195	-0.000504	1.028042	0.664319	0.000155	-1.024488	0.709417	0.000676
N	2.397481	0.617190	-0.000462	2.415095	0.650165	0.000018	-2.406988	0.659490	0.001835
N	2.957963	-0.495327	0.000613	2.951632	-0.469323	0.000039	-2.949231	-0.454938	-0.000289
N	3.594726	-1.421214	0.001603	3.578545	-1.404850	0.000081	-3.583862	-1.389184	-0.001970
N	-2.272760	-0.579543	0.000198	-2.238335	-0.606403	0.000033	2.277559	-0.536639	-0.000101
O	-3.305385	0.069594	0.003654	-3.231663	0.119696	0.000053	3.345855	0.038327	-0.001148
O	-2.179154	-1.795201	-0.003144	-2.224015	-1.819253	-0.000064	2.099864	-1.758209	0.001218
H	0.650611	2.705006	-0.002310	-1.670105	2.095640	0.000201	-0.320075	-1.348280	0.000677

	X	Y	Z	X	Y	Z	X	Y	Z
N	0.287640	-0.691332	-0.002681	0.255471	-0.670018	0.001259	0.324163	-0.617826	0.000417
C	1.610646	-0.340029	-0.007571	1.547081	-0.391205	-0.003945	1.657673	-0.276763	0.006321
N	1.851681	0.963228	0.000855	1.727615	0.949205	0.002822	1.798501	1.020875	0.024031
N	0.580535	1.482319	0.000964	0.513123	1.593958	0.003662	0.529377	1.584017	0.005978
C	-0.315283	0.482519	0.002553	-0.315859	0.571952	-0.000184	-0.324546	0.605238	-0.008866
N	-1.676992	0.759009	0.011071	-1.693952	0.793487	0.004164	-1.705728	0.792760	0.000382
N	-2.430823	-0.226794	0.001045	-2.411909	-0.213394	0.002392	-2.428469	-0.206075	0.001080
N	-3.227522	-1.023491	-0.005740	-3.190340	-1.031683	0.001521	-3.208039	-1.029859	0.003747
N	2.606493	-1.287251	-0.074093	2.566487	-1.311144	-0.074394	2.667807	-1.219223	-0.083792
H	2.347336	-2.193611	0.284901	2.576298	1.486515	-0.071022	-0.062223	-1.535206	-0.162745
H	3.518431	-0.968572	0.217823	2.254052	-2.258673	0.084548	2.645071	-1.937980	0.629670
H	0.424969	2.477430	0.007443	3.416863	-1.085200	0.421264	3.575106	-0.770352	-0.114559

	1H-AzTA-AZ			2H-AzTA-AZ			4H-AzTA-AZ		
	X	Y	Z	X	Y	Z	X	Y	Z
N	0.451065	1.025958	-0.000151	0.415312	1.001767	-0.000097	-0.525390	0.942891	0.000403
C	1.815826	0.990546	0.000244	1.733934	1.029829	-0.000036	-1.899220	0.916353	-0.000028
N	2.350268	-0.215759	-0.000190	2.217850	-0.227382	0.000128	-2.330124	-0.313543	-0.000559
N	1.248767	-1.015449	-0.000010	1.204059	-1.130589	-0.000225	-1.229585	-1.148036	-0.000279
C	0.135284	-0.253971	0.000095	0.146461	-0.335645	0.000083	-0.169981	-0.384981	0.000291
N	-1.120283	-0.848528	0.000125	-1.135848	-0.883693	0.000006	1.128472	-0.890486	0.000829
N	-2.087883	-0.070234	-0.000017	-2.075537	-0.078037	0.000036	2.064263	-0.085925	-0.000081
N	-3.051188	0.512625	-0.000064	-3.027391	0.528181	0.000081	3.014500	0.530941	-0.000751
H	1.332614	-2.020268	-0.000045	3.173432	-0.549167	0.000238	0.071321	1.756318	0.001606
H	2.425502	1.880536	0.000158	2.355079	1.912339	-0.000024	-2.511065	1.804546	-0.000118

(C) B3LYP/AUG-cc-PVDZ

	1H-AAzTA-AZ			2H-AAzTA-AZ			4H-AAzTA-AZ		
	X	Y	Z	X	Y	Z	X	Y	Z
N	0.194699	-0.441223	0.000573	-0.237054	-0.445302	-0.000144	0.160750	-0.345962	-0.000230
C	-1.014536	0.171977	-0.000133	0.966020	0.090215	-0.000084	-1.069828	0.252782	0.000241
N	-1.017062	1.493330	-0.001044	0.909747	1.434131	0.000137	-0.975183	1.557948	0.000295
N	0.309124	1.756255	-0.001278	-0.375725	1.835147	-0.000452	0.357564	1.860796	-0.000074
C	1.016598	0.596102	-0.000424	-1.028502	0.664677	0.000180	1.025102	0.712828	-0.000134
N	2.402857	0.625792	-0.000340	-2.419514	0.658908	0.000031	2.411675	0.667754	-0.000467
N	2.960992	-0.495061	0.000556	-2.956566	-0.467821	0.000113	2.949927	-0.456159	0.000005
N	3.603433	-1.426061	0.001402	-3.590395	-1.407484	0.000113	3.583857	-1.399532	0.000482
N	-2.273275	-0.580741	0.000195	2.239388	-0.606680	-0.000101	-2.274682	-0.540011	0.000033
O	-3.309914	0.070330	0.003017	3.236569	0.121022	0.000137	-3.349993	0.032681	0.000181
O	-2.180645	-1.800954	-0.002394	2.226676	-1.824486	0.000023	-2.094064	-1.765376	-0.000345
H	0.646730	2.710481	-0.002091	1.679776	2.092060	0.000266	0.323458	-1.345944	0.000355

	X	Y	Z	X	Y	Z	X	Y	Z
N	0.286614	-0.694828	-0.004077	0.254411	-0.673553	0.000674	0.322329	-0.615358	-0.001747
C	1.612586	-0.342839	-0.007003	1.550666	-0.392270	-0.004429	1.657976	-0.281251	0.004281
N	1.856123	0.965374	0.002211	1.731067	0.950080	0.000644	1.803993	1.022389	0.019167
N	0.583715	1.484954	0.001560	0.517059	1.597908	0.003793	0.535080	1.590295	0.004549
C	-0.315980	0.485215	0.001852	-0.316535	0.571966	-0.000160	-0.325248	0.607840	-0.004864
N	-1.680494	0.767347	0.009369	-1.696498	0.800804	0.005203	-1.708855	0.801997	0.007663
N	-2.433120	-0.227583	0.001327	-2.415502	-0.213766	0.002644	-2.430630	-0.205859	0.000661
N	-3.236236	-1.028351	-0.004301	-3.201344	-1.034897	0.001194	-3.212020	-1.037830	-0.002930
N	2.611565	-1.291272	-0.075662	2.573145	-1.314504	-0.076456	2.669174	-1.228790	-0.085587
H	2.352359	-2.197153	0.290265	2.583924	1.485977	-0.062154	-0.072933	-1.537183	-0.124833
H	3.522975	-0.968681	0.220575	2.256462	-2.261545	0.093240	2.646767	-1.937552	0.641904
H	0.427863	2.482088	0.007077	3.418467	-1.087108	0.432573	3.576293	-0.772715	-0.106017
	<i>1H-AzTA-AZ</i>			<i>2H-AzTA-AZ</i>			<i>4H-AzTA-AZ</i>		
	X	Y	Z	X	Y	Z	X	Y	Z
N	0.449818	1.029562	-0.000172	0.414078	1.005200	-0.000038	-0.522413	0.941972	0.000232
C	1.817118	0.995145	0.000255	1.737787	1.032491	-0.000040	-1.897298	0.924036	-0.000140
N	2.355418	-0.215485	-0.000177	2.222128	-0.226860	0.000089	-2.336147	-0.309953	-0.000181
N	1.253258	-1.016519	-0.000025	1.209370	-1.133268	-0.000107	-1.237916	-1.150512	-0.000130
C	0.135780	-0.256596	0.000101	0.146481	-0.335556	0.000013	-0.170132	-0.386800	0.000122
N	-1.121085	-0.857327	0.000133	-1.136266	-0.890850	-0.000014	1.128788	-0.900784	0.000366
N	-2.089628	-0.069783	-0.000014	-2.078449	-0.077502	0.000012	2.066207	-0.087660	-0.000052
N	-3.060151	0.515641	-0.000063	-3.038002	0.529976	0.000061	3.020364	0.536239	-0.000362
H	1.338337	-2.023291	-0.000073	3.180483	-0.546875	0.000169	0.081612	1.752379	0.001270
H	2.430865	1.889371	0.000162	2.363893	1.918395	-0.000030	-2.509214	1.819096	-0.000276