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Supporting information for

# The coarse–grained model for water/oil/solid system: based on the correlation of water/air and water/oil contact angles

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## 1. Comparison of Bartell-Osterhof equation and the result of CGMD simulation

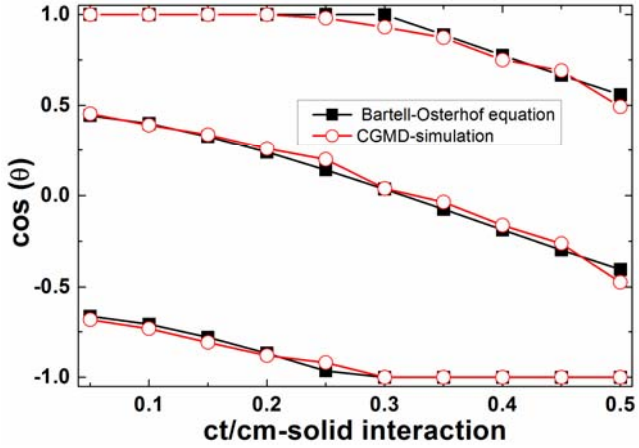


**Figure S1** Comparison of Bartell-Osterhof equation (squares) and the results of CG MD simulation (circles) for the different water/solid interaction( top, water/solid 0.75; Middle, water-solid 0.5; bottom, water/solid 0.15)

Figure S1 as obtained as following: Firstly, the water/solid interactions were set to 0.15, 0.5 and 0.75, respectively, and the $\cos(\theta_{wa})$ as obtained with the model (Figure 1 in the Manuscript). Secondly, the dodecane/solid interactions (in fact, the CT&CM-solid interactions) were set as 0.05 to 0.5, each per 0.05, respectively. The $\cos(\theta_{oa})$ were also calculated based on the model (Figure 1 in the Manuscript). Thirdly, the eq. 1 is translated to eq. 2, and a total of 30 $\cos(\theta_{ow})$s were obtained by combination of any water/solid and oil/solid parameters mentioned above. Finally, a total of 30 CG MD have been carried out under the different combinations of above parameters. Figure S1 was painted via comparing the CG MD results and the calculated data based on eq. S2.

$$\sigma_{\mathrm{wa}} \cos\theta_{\mathrm{wa}} = \sigma_{wo} \cos\theta_{wo} + \sigma_{\mathrm{oa}} \cos\theta_{\mathrm{oa}} \tag{S1}$$

$$\cos\theta_{wo} = \frac{\sigma_{\mathrm{wa}} \cos\theta_{\mathrm{wa}} - \sigma_{\mathrm{oa}} \cos\theta_{\mathrm{oa}}}{\sigma_{wo}} \tag{S2}$$

## 2. Water/alkanes system

### 2.1 Bonded interactions

Bonded interactions were calculated by a weak harmonic potential $U_{Bonded}$ (eq. 4). The bond interaction parameters and angle interaction parameters were adjusted by comparing corresponding bonds and angles, which were obtained from atomistic simulations respectively. Firstly, the Amorphous Cell tool of Materials Studio (AccelrysInc) was used to construct different systems (the hexane, nonane, dodecane and pentadecane systems, the number of moleculesare 100 composed by different molecular fragments, Equivalent to one CG interaction site). The periodic boundary condition was taken into account in all direction. Secondly AA MD was performed by discover tool of Materials Studio, the interatomic interactions were described by the forcefield of a condensed-phase optimized molecular potential for atomistic simulation studies (COMPASS)[1]. All the models were put into a npt ensemble, a fixed time sep of 1 fs was used and a 1 ns simulation time was performed, the last 0.5 ns was used for the data analysis. Thirdly, the bond stretch and angle bend distributions of the center of mass (COM) of different molecular fragments were generated. Gaussian distribution fitted these distributions and the fitting parameters were shown in Table S1 (Column AA MD). Finally, the Mesostructure model of Materials Studio (AccelrysInc) was used to construct different systems correspond to the AA MD systems, and these models were calculated with the time step of 10 fs in the npt ensemble. The bond stretch and angle bend distribution of CG MD were generated and the Gaussian distribution also fitted these distributions. The CG Bonded interactions were adjusted successively until the fitting parameters of CG is consistent with AA parameters. All spring constants and equilibrium constants were

shown in Table S1.

**Table S1** The Gaussian fitting parameters of AAMD、CGMD andthe final bonded parameters

| | AAMD | | Bonded parameters | | CGMD | |
|---|---|---|---|---|---|---|
| Bond | σ | μ | $K_{stertch}$ | $R_0$ | σ | μ |
| CT-CT | 0.24 | 3.74 | 10.89 | 3.77 | 0.24 | 3.73 |
| CT-CM | 0.23 | 3.69 | 11.54 | 3.73 | 0.23 | 3.69 |
| CM-CM | 0.23 | 3.66 | 11.35 | 3.71 | 0.23 | 3.67 |
| Angle | σ | μ | $K_{bend}$ | $\theta_0$ | σ | μ |
| CT-CM-CT | 19.02 | 149.42 | 4.20 | 158 | 19.00 | 148.51 |
| CT-CM-CM | 19.00 | 149.09 | 4.20 | 158 | 19.00 | 148.51 |
| CM-CM-CM | 19.30 | 148.83 | 4.22 | 157 | 19.13 | 148.23 |

2.2 Non-bonded interactions

We introduced models and aprocedure to obtain the intra water, the intra alkanes and the inter alkanes/water interaction parameters respectively.

Model and Setup

The non-bond interactions were fitted through experimental density and surface/interfacial tension data. The simulation boxes with size 4.0*4.0*4.0 $nm^3$ are filled with liquid, which was carried out in npt ensemble to generate the density data. And the simulation boxes, 8.0*4.0*4.0 $nm^3$ with a 4.0 nm-long liquid slab, were carried out in nvt ensemble, which are used to generate the surface tension data. The simulation boxes, 8.0*4.0*4.0$nm^3$ with a water slab and an alkane slab were carried out in npt ensemble, which are used to generate the water/alkanes interfacial tension data. The temperature was maintained at 298 K and the time step

is 10 fs for all the simulation. The surface tension is computed by equation S3[2].

$$\sigma = \frac{L_z}{2}[P_{zz} - \frac{P_{xx} + p_{yy}}{2}], \quad \text{(S3)}$$

Water

The water/water interaction parameters were determined by experimental surface tension and density data. The CG water/water interaction parameter was adjusted successively until that the density and surface tension of CG is consistent with AA parameters (Table S2).

**Table S2** Surface tension and density of the CG model for water

| LJ | $D_0$（Kcal/mol) | R(Å) | density（g/cm$^3$） | | Surface tension(N/m) | |
|---|---|---|---|---|---|---|
| | | | EXP[a] | CGMD | EXP[a] | CGMD |
| 12-4 | 0.86 | 5.01 | 0.997 | 1.000 | 71.99 | 71.06 |

[a]Experimental data are taken from Ref 3

Alkanes

There are two interaction sites, CT ($CH_3$-$CH_2$-$CH_2$-), CM (-$CH_2$-$CH_2$-$CH_2$-), and by combination of them a variety of alkanes could be built. The LJ9-6 was used as non-bonded interaction form. The CT-CT parameters were obtained using hexane. CM-CM parameters were determined through nonane, and CT-CM interaction parameters were calculated by Lorentz combining rules. Based on above parameters, surface tension and density of dodecane and pentadecane were calculated. Results of different alkanes by CG MD method were compared with experimental data and these values revealed a good agreementas shown in Table S3.

**Table S3** Surface tension and density of the CG model for pure alkanes

| System | Interaction sites | Density(g/cm$^3$) | | Surface tension(N/m) | |
|---|---|---|---|---|---|
| | | EXP[a] | CGMD | EXP[a] | CGMD |
| Hexane | Ct-ct | 0.656 | 0.657 | 17.98 | 18.20 |
| Nonane | Ct-cm-ct | 0.715 | 0.715 | 22.43 | 22.12 |
| Dodecane | Ct-cm-cm-ct | 0.745 | 0.745 | 24.94 | 24.27 |
| pentadecane | Ct-cm-cm-cm-ct | 0.765 | 0.763 | 26.68 | 25.89 |

[a]Experimental data are taken from Ref[3]

Alkanes/Water interaction

Parameters for the interactions between alkanes interaction sites and water are actually the interaction of CT-W, CM-W. The LJ12-4 was used as non-bonded interaction form, and the interfacial tension of water/alkaneswas used to fit the non-bonded interaction parameters (CT-W, CM-W). Both CGMD and experimental results are shown in Table S4. All interaction parameters are shown in Table S5.

**Table S4** Interfacial tension of the CG model for water/alkanes interface

| System | Interaction sites | Interface tension(N/m) | |
|---|---|---|---|
| | | EXP[a] | CGMD |
| Hexane-Water | CT-W | 50.38 | 50.57 |
| Nonane-Water | CT-W, CM-W | 51.63 | 51.85 |
| Dodecane-Water | CT-W, CM-W | 52.55 | 52.43 |
| Pentadecane-Water | CT-W, CM-W | 53.29(fitting） | 52.96 |

[a]Experimental data are taken from Ref[4]

**Table S5** Non-bonded and Bonded interaction parameters

| Non-bond interactions | | | | | | |
|---|---|---|---|---|---|---|
| Bead-1 | Bead-2 | LJ-Form | $D_0$(Kcal/mol) | R(Å) | $R_{cut}$(Å) | T(K) |
| W | W | 12-4 | 0.860 | 5.010 | 12.5 | 298 |
| W | CT | 12-4 | 0.345 | 5.115 | 12.5 | 298 |
| W | CM | 12-4 | 0.335 | 5.055 | 12.5 | 298 |
| CT | CT | 9-6 | 0.470 | 5.220 | 12.5 | 298 |
| CT | CM | 9-6 | 0.450 | 5.160 | 12.5 | 298 |
| CM | CM | 9-6 | 0.430 | 5.100 | 12.5 | 298 |
| Bond interaction | | | | | | |
| Bead-1 | Bead-2 | Functional form | $K_{stertch}$ (Kcal/mol/ Å$^2$) | $R_0$(Å) | $R_{cut}$(Å) | T(K) |
| CT | CT | Harmonic | 10.89 | 3.77 | 12.5 | 298 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| CT | CM | Harmonic | 11.54 | 3.73 | 12.5 | | 298 |
| CM | CM | Harmonic | 11.35 | 3.71 | 12.5 | | 298 |

Anlge interaction

| Bead-1 | Bead-2 | Bead-3 | Functional form | $K_{bend}$ (Kcal/mol/rad$^2$) | $R_0$($^O$) | $R_{cut}$(Å) | T(K) |
|---|---|---|---|---|---|---|---|
| CT | CM | CT | Harmonic | 4.17 | 157 | 12.5 | 298 |
| CT | CM | CM | Harmonic | 4.17 | 158 | 12.5 | 298 |
| CM | CM | CM | Harmonic | 4.60 | 155 | 12.5 | 298 |

## References


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