Supporting Information

Atomistic simulations

Force-field parameters

The OPLS-AA[?] force-field has been used to perform the atomistic simulations of AN and PN dimers. 1 shows the chemical structure of both dimers, which have been terminated by methyl groups. The figure also shows the numbers *n* associated to the OPLS-AA types chosen to describe each atom (types are named opls_*n* in OPLS-AA topologies). In a few cases, in order to preserve the neutrality of the whole molecule, partial charges have been modified. The opls_139 type in both dimers has been attributed a charge 0.05, and in the PN dimer the opls_145 type has been given no charge.

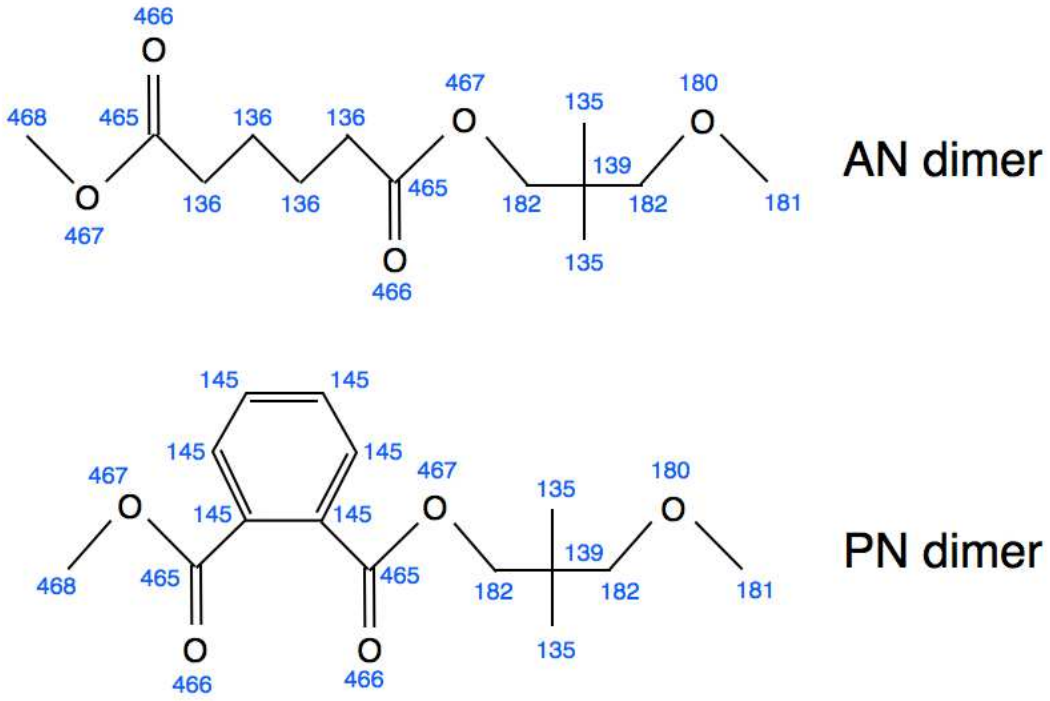

Figure 1 OPLS-AA atom types used in atomistic simulations.

Molecular Dynamics set-up

A cut-off of 1.3 nm was used for the calculation of the non-bonded interactions, without switch function nor long-range dispersion correction. Simulations were performed with a time step of 2 fs, with neighbour updates every 10 steps. Simulations were carried out in the NpT ensemble using the Nosé-Hoover thermostat ($\tau_t = 1$ ps) and Parrinello-Rahman barostat ($\tau_p = 4$ ps).

Coarse-grained simulations

Molecular Dynamics set-up

In the CG simulations, a cutoff of 1.2 nm was used in the calculation of non-bonded interactions, with a shift function starting from 0.9 nm for dispersion interactions. A time step of 20 fs was used, and the neighbour list for non-bonded interaction was updated every 200 fs. These conditions correspond to the standard ones used in the parameterization of MARTINI. Simulations in the NpT ensemble were carried out with the Nosé-Hoover thermostat (τ_T = 1 ps) and the Parrinello-Rahman barostat (τ_P = 4 ps).

Thermodynamic integration

The free energy of transferring an CG HMMM molecule from one solvent (water) to the other (octanol) was calculated using the thermodynamic integration formula[?] :

$$\Delta G = \int_0^1 \langle \frac{\partial H}{\partial \lambda} \partial \lambda \rangle_\lambda \tag{1}$$

where λ represents the degree of coupling between the HMMM molecule and the solvent, H is the total Hamiltonian of the system, and G the Gibbs free energy.