Molecular Effects of Encapsulation of Glucose Oxidase Dimer by Graphene

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Supplementary Information

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Figure S1 RMSD of GOx monomers in GOx dimer-graphene complexes after 100 ns of MD simulation in water environment. (a) GOx dimer interacting with unconnected graphene flakes. Monomer B of GOx (in green) stabilized at about 30 ns while monomer A (in blue), being in contact with graphene stabilizes at about 80 ns of MD simulation. (b) GOx dimer interacting with graphene in Box1 and Box4 models. All monomers stabilized after about 90 ns.
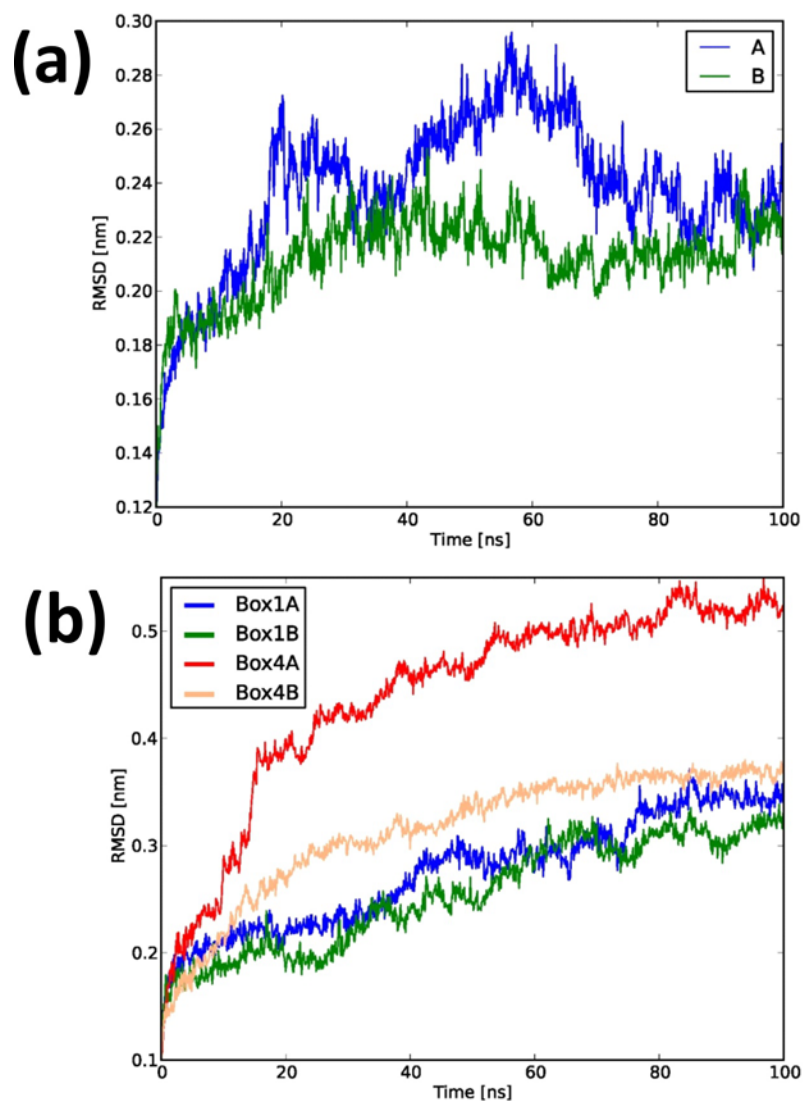
Figure S2 Distances in GOx dimer-graphene complexes during 100 ns MD simulation in water environment. (a) A separation between GOx monomers in Box1 and Box4 models. A separation in the crystal structure of GOx dimer is 3.85 nm. (b) The closest distance between cofactor FAD and graphene. In both cases it is about 1.85 nm although in Box4 it increases.
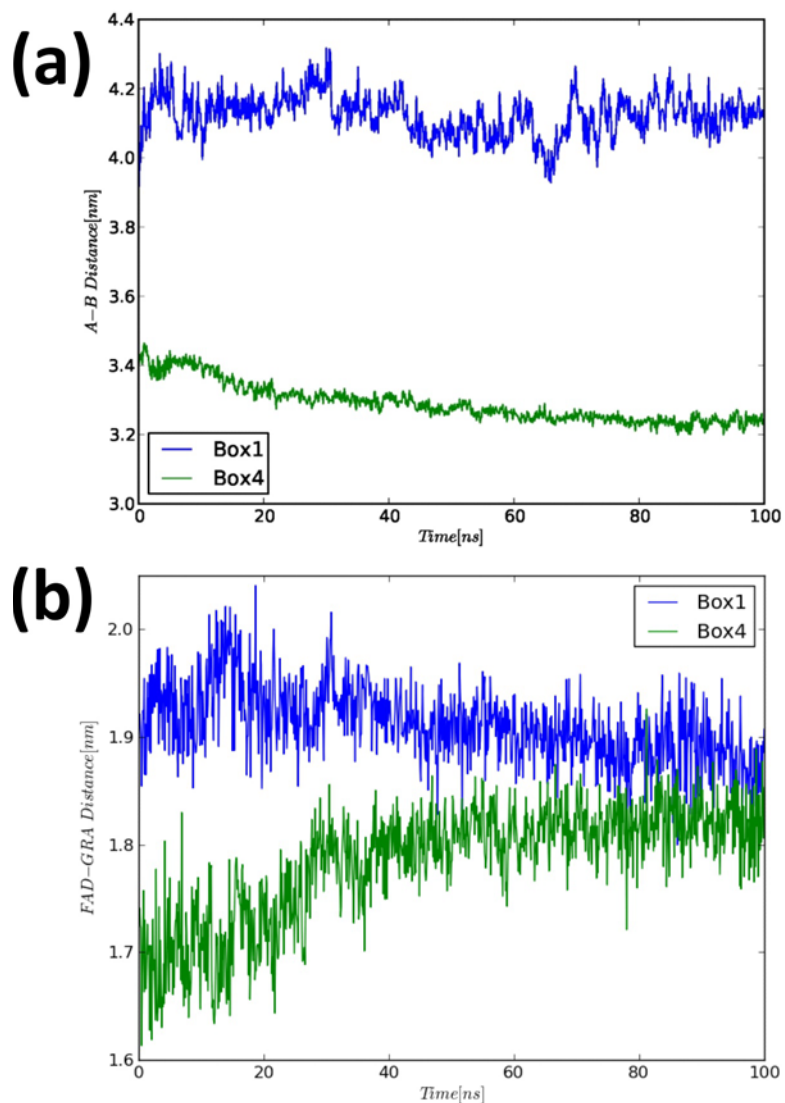
Figure S3 Volume of GOx dimer during 100 ns of MD simulation in Box1 and Box4. Although the final volumes are nearly the same in both cases there are much larger fluctuations of volume in Box4.
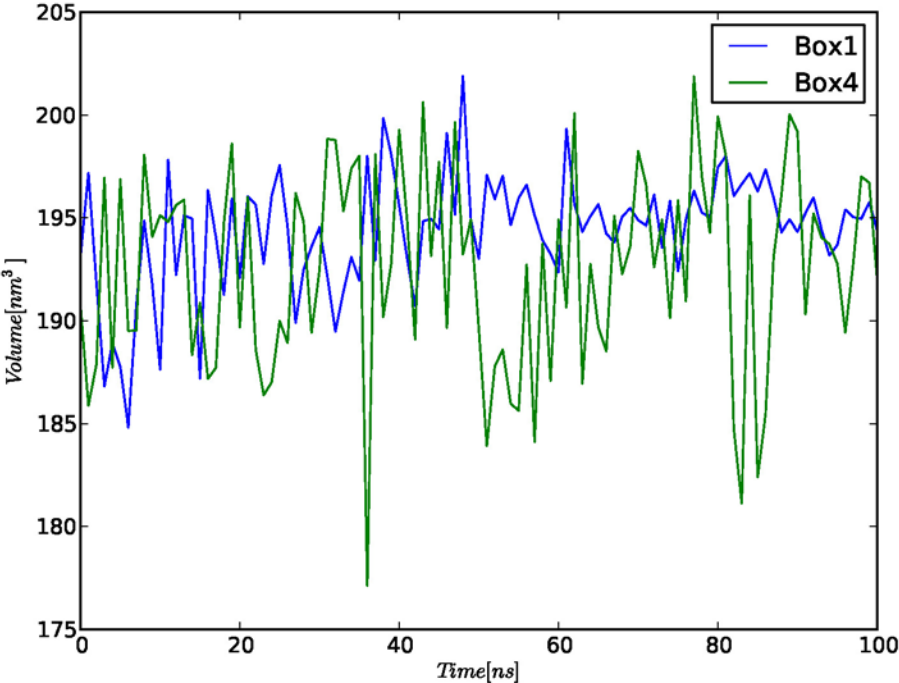
Figure S4 Root mean square fluctuations (RMSF) of amino acid residues in Box1 and Box4 model complexes of graphene with GOx dimer. Monomers are denoted A and B. The residues anchoring a cofactor FAD are colored in blue while the catalytic residues in red. Region N111-S144, binding to flavin group of FAD, is of slightly increased flexibility in Box4 model. Residues E55 and V254 bind to adenine part of FAD.
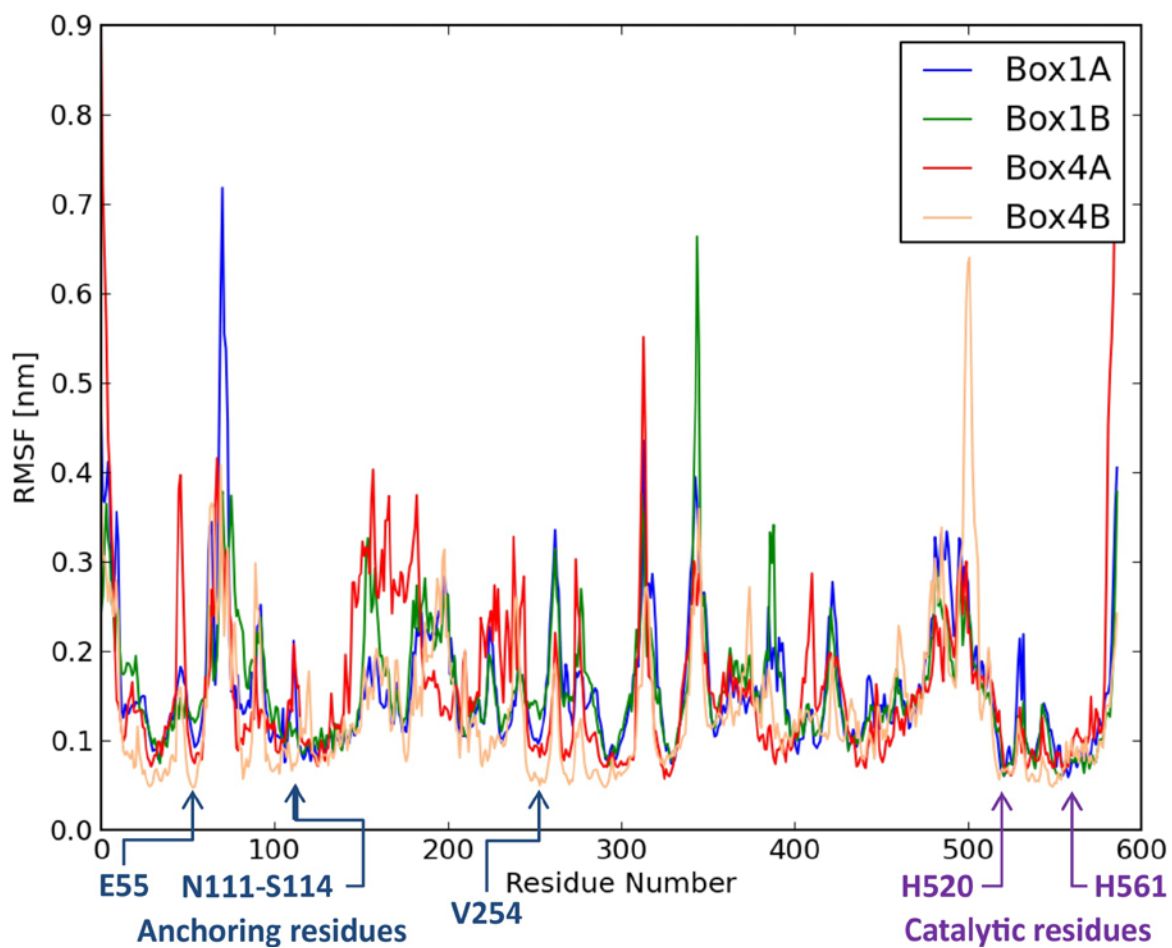
Figure S5 Changes of secondary structures of monomers A and B in GOx dimer-graphene complexes Box1 and Box4. In all cases a Loop (disordered structure) content increased while Helix content decreased. Helix and Strand contest is larger in Box1 compared to Box4.
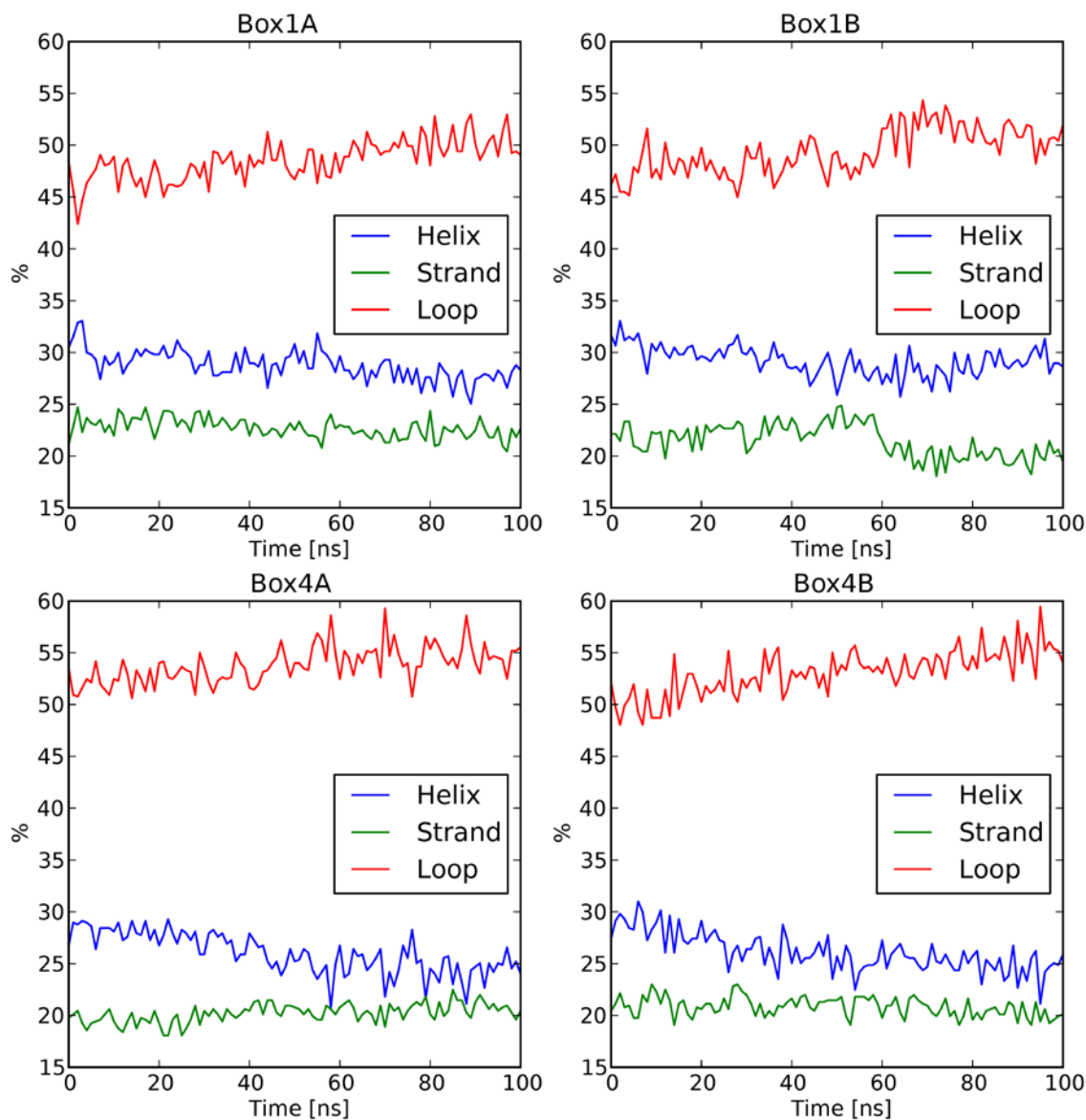
Figure S6 Interactions of individual amino acid types with graphene in Box1 and Box4 models. In all cases there is a stabilization of interactions. The amino acid type contributing most to the total interaction energy is Lys and Gln (blue and green in the first row) mostly because of large number of these residues on the surface of GOx dimer, 20 and 13, respectively. On the right half, the interactions of individual amino acid types normalized to single residue. Lys and Gln still contribute the most among other residues to the interaction energy in both Box1 and Box4.
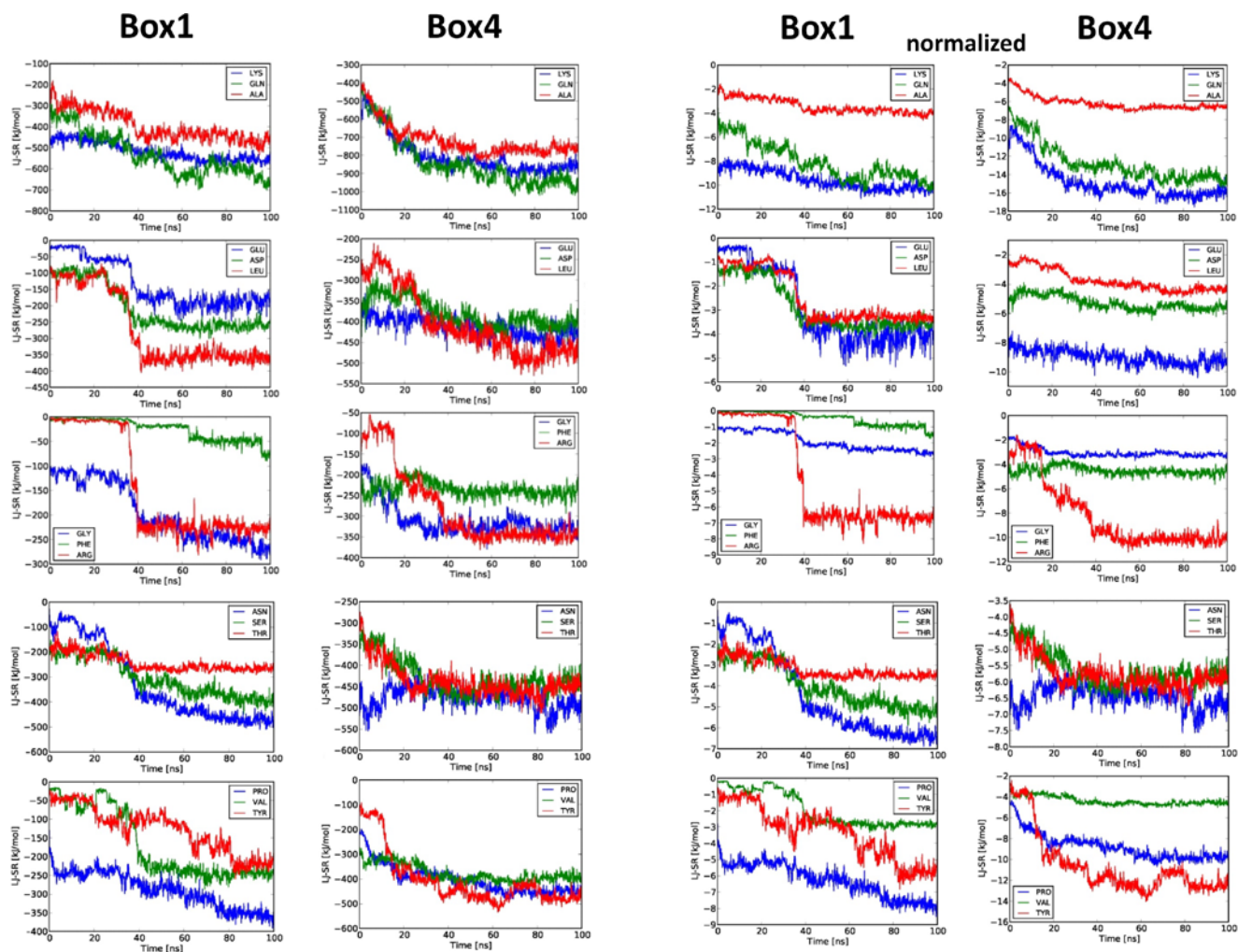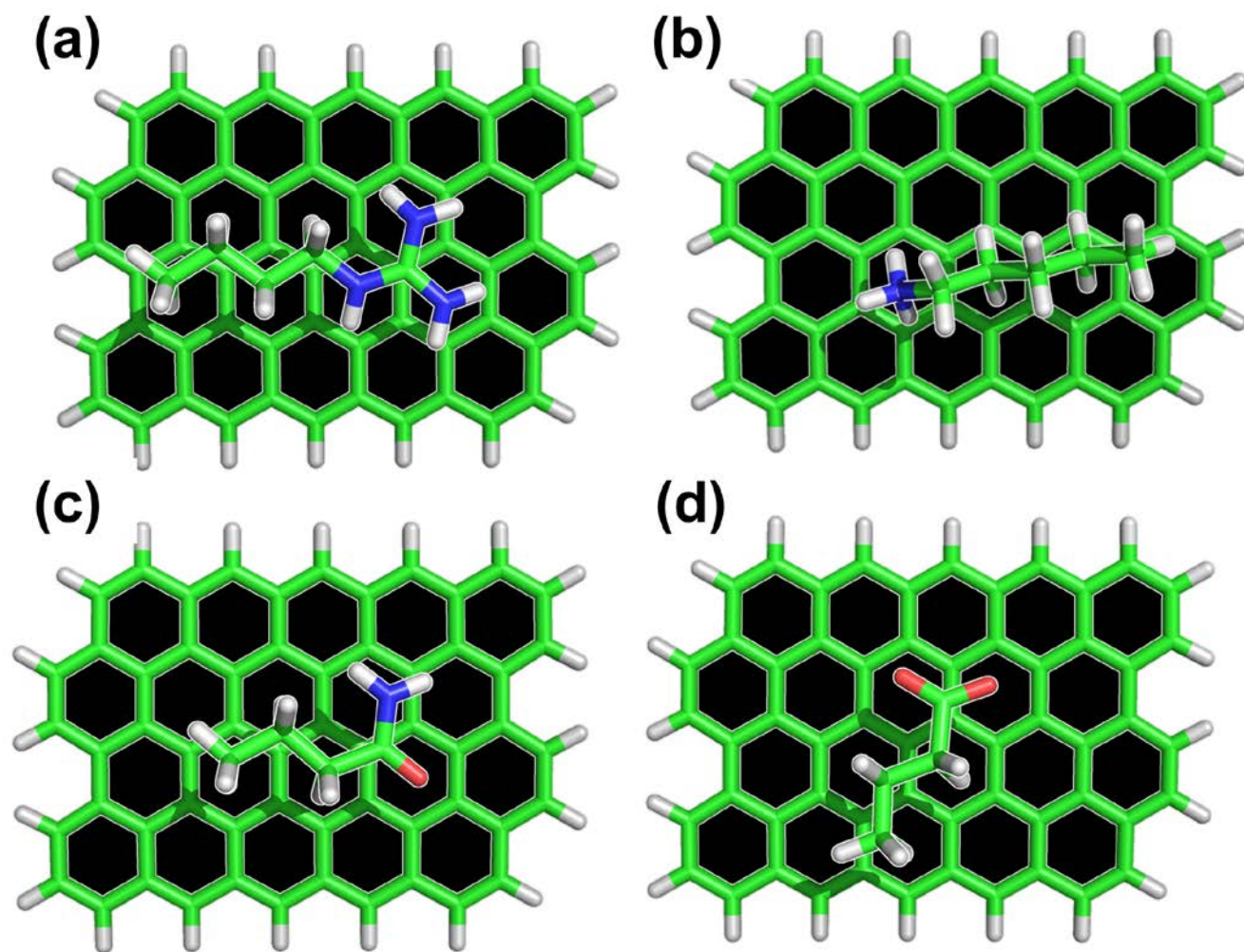
Figure S7 Complexes of graphene and side chains of amino acids optimized in DFT. Such structures were used for final energy calculations in DFT method M06-2X/6-31+G(d,p); (a) charged arginine; (b) charged lysine; (c) glutamine; (d) charged glutamate.

Movie M1 GOx dimer with unconnected graphene sheets. Fragment of 100 ns MD simulation in explicit water environment of GOx dimer and four unconnected graphene flakes manifesting a stacking effect.
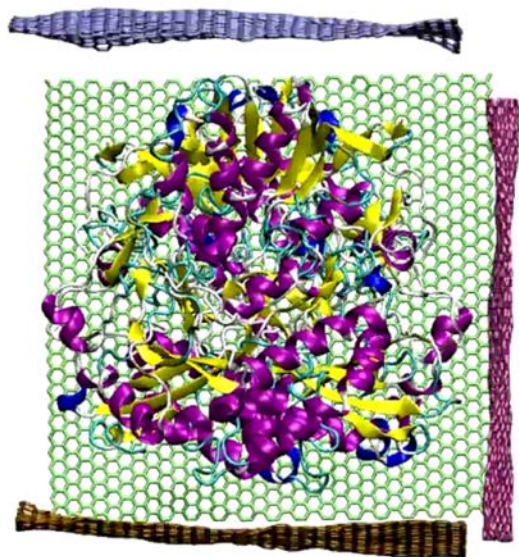
Movie M2 Encapsulation of GOx dimer by graphene sheets connected via narrow linkers. Part of stochastic dynamics (SD) simulation of GOx dimer and model Box1 (0.7 nm wide linkers) of graphene in vacuum. The initial simulation was followed by MD simulation in explicit water environment.
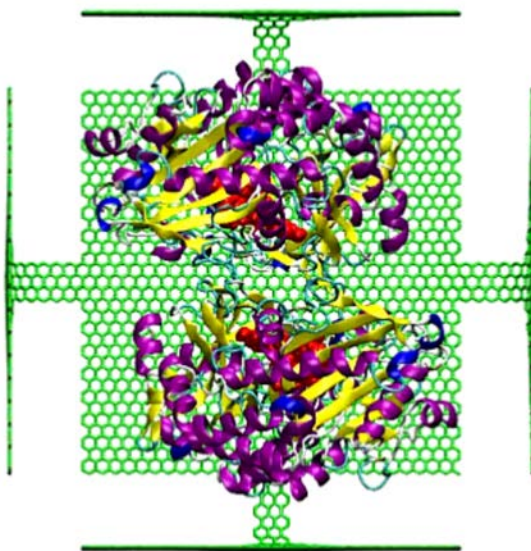
Movie M3 Encapsulation of GOx dimer by graphene sheets connected via wide linkers. Part of stochastic dynamics (SD) simulation of GOx dimer and model Box4 (7.0 nm wide linkers - equal to the size of the sheet) of graphene in vacuum. The initial simulation was followed by MD simulation in explicit water environment.