



Article

Role of Ionic Strength in the Formation of Stable Supramolecular Nanoparticle–Protein Conjugates for Biosensing

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Supporting Information

S.1 Binding Energy decomposition

In this section we present an energy decomposition analysis performed with the *rerun gromacs tool*,¹ through which we determined that -30GFP molecules tether the AuArg and the +36GFP the AuCOO, mostly through electrostatic interactions. This result provides a solid basis for the rationalization of the effect of ionic strength on the binding mechanism and stability of AuArg. The results indicate that the stability of protein-AuNP conjugates can be maintained at comparable stability even at different ionic strengths due to the number of ions trapped within the monolayer at the different ionic strength, which alter the screening due only to the solution.

Figure S1. VdW and LJ interactions between AuArg and -30GFP molecules in the 4 conformations identified at each ionic strength.

¹ Lundborg, M., and Lindahl, E.: “Automatic GROMACS topology generation and comparisons of force fields for solvation free energy calculations”, The journal of physical chemistry. B, 2015, 119, (3), pp. 810-823

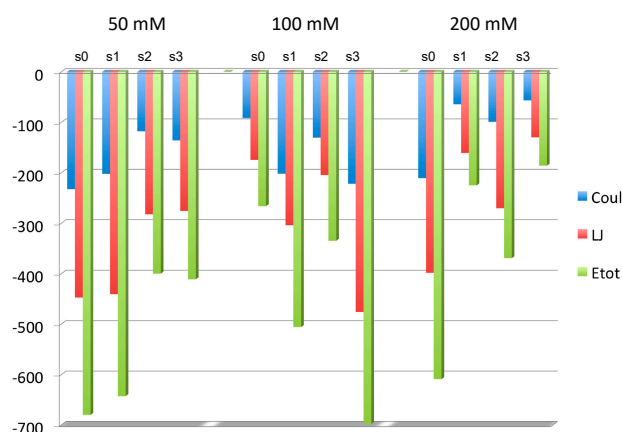
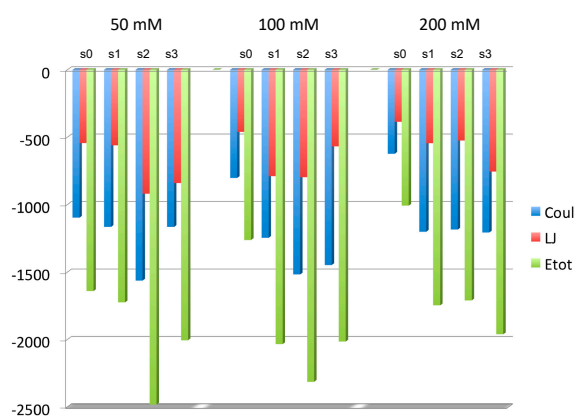


Figure S2. VdW and LJ interactions between AuCOO and +35GFP molecules in the 4 conformations identified at each ionic strength.



S.2. RESP charges parametrization

In this section we present the derivation of RESP charges from *ab initio* simulations.

Figure S3. RESP partial charges derivation by *ab initio* calculations for the surface atoms of the smallest interfacial repeating unit of AuArg, namely AUS-RS-AUL-SR-AUS. The SR=alkanethiol functional groups is connected to the core with a sulfur atom forming the so-called “staples” (dark and light yellow in the figure), where the sulfur atom is forming a covalent bond with one gold atom at the interface (AUL=gold ligand) and a gold atom at the surface (AUS = gold surface). Using R.E.D. server² the partial charges are evaluated from density functional calculations at b3lyp/6-31G*//b3lyp/6-31G* level and Connolly surface algorithm is used in MEP computation (2 stage RESP fit qwt=0.0005/0.001).

² Vanqualef, E., Simon, S., Marquant, G., Garcia, E., Klimerak, G., Delepine, J.C., Cieplak, P., and Dupradeau, F.Y.: ‘R.E.D. Server: a web service for deriving RESP and ESP charges and building force field libraries for new molecules and molecular fragments’, Nucleic acids research, 2011, 39, (Web Server issue), pp. W511-517

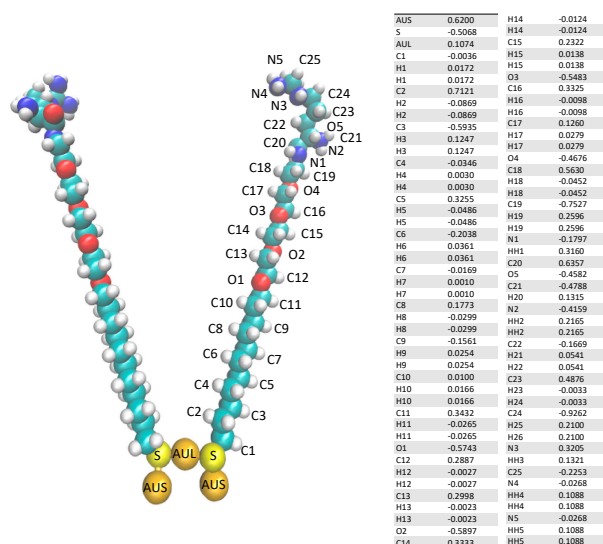
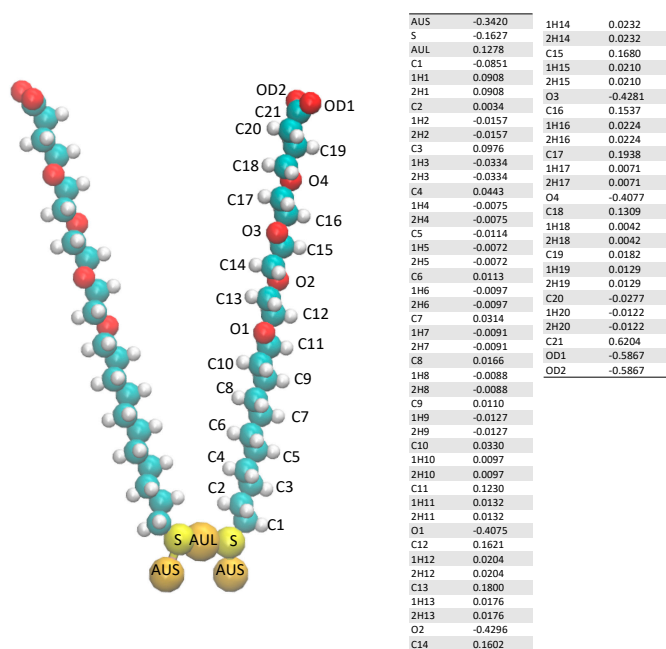


Figure S4. RESP partial charges derivation by *ab initio* calculations for the surface atoms of the smallest interfacial repeating unit of AuCOO, namely AUS-RS-AUL-SR-AUS. The SR=alkanethiol functional groups is connected to the core with a sulfur atom forming the so-called “staples” (dark and light yellow in the figure), where the sulfur atom is forming a covalent bond with one gold atom at the interface (AUL=gold ligand) and a gold atom at the surface (AUS = gold surface). Using R.E.D. server³ the partial charges are evaluated from density functional calculations at b3lyp/6-31G*/b3lyp/6-31G* level and Connolly surface algorithm is used in MEP computation (2 stage RESP fit qwt=0.0005/0.001).



³ Vanqualef, E., Simon, S., Marquant, G., Garcia, E., Klimerek, G., Delepine, J.C., Cieplak, P., and Dupradeau, F.Y.: ‘R.E.D. Server: a web service for deriving RESP and ESP charges and building force field libraries for new molecules and molecular fragments’, Nucleic acids research, 2011, 39, (Web Server issue), pp. W511-517