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Computer-aided Design of Bio-inspired Nanoporous Silica Materials

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1 Summary

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2   Acknowledgements

3   Introduction

Molecular Dynamics (MD) and Monte Carlos (MC) have been powerful tools to simulate molecular interactions in lipid in solvent systems, allowing a deeper understanding of self-assembly process of this sort of material (?, ?). Therefore, these structural conformations are specially useful to design bio-inspired silica materials (?, ?) since with the addition of silica they remain behaving as scaffolds to mesoporous or nanoporous structures that are maintained even after surfactant removal, silica oligomer polymerization and calcination (?, ?). A vast range of silica materials are examples of this phenomena, such as MCM-41 as reported by ? (?), HR (?, ?), MSU-V (?, ?) and many others, in which the self-assembly structure depend on the type of lipid and concentration of the substances involved. Moreover, most of the experimental methods used to obtain data are based on observation and interpretation of final silica structure by using X-ray diffraction (XRD) and transmission electron microscopy (TEM), thus initial lipidic self-assembled conformations are predicted as a reflex of the final results and little is known about the mechanistic of this process. However, by using MD simulations it is possible to observe and analyse these initial steps of self-assembly and further predict, with more accuracy, properties and framework provided by lipids (?, ?) to generate silica structures.

A major concern is that even though MD uses sophisticated software prepared to simulate enormous systems with thousands of atoms, using all capacities of robust hardware that are available, such as high-speed multi-core processors in conjunction with GPUs designed specifically to process data from arrays (?, ?), they hardly can achieve long time horizons and are commonly limited to a few μs depending on the size of the system. For this reason, several techniques have been develop to optimize the performance of the simulations such as united-atoms and coarse-grain models (?, ?). The work presented on following experiments are an attempt to create a cross-link method to upscale silica crystal-liquid phase interactions (?, ?) from the atomistic model to a mesoscale model with the advent of coarse-grain technique. The methodology applied to reach the desired model is based on work previously done by ? (?) which introduced the MagiC software package (?, ?) that in conjunction MD simulation software, in this case GROMACS (?, ?), will provide a suitable approximation to self-assembly of amphiphilic molecules in presence of silica. Further explanations of the process are described on Experimental Methods section.

For the scope of this project, a bolaamphiphilic molecule 1,12-diaminododecane (DMDD) has been chosen as surfactant because that as seen in previous research by ? (?) it self-assembly in multilamelar vesicles that in presence of a silica precursor is capable of generating a mesoporous structure with remarkable properties. In order to validate this structure formation and further framework formation for silica oligomers, a coarse-grain approximation is suitable option since amphiphilic molecules interaction with solvents can be described efficiently with Lennard-Jones potentials interactions, in such manner, that even implicit solvent interaction potentials generated from a bottom-up approach are applicable for a coarse-grain model (?, ?). It is supposed that thermodynamic changes on the system, originated from solvent interaction with amphiphilic molecules, are incorporated on the approximated potentials and therefore changes in parameters as concentration may not affect coarse-grained model performance on recreating self-assembly structures.

Moreover, previous works conducted by (?, ?) with a coarse-grained model of surfactant in presence of silica with explicit water were successful in describe rod-like structures detected on MCM-41 materials, demonstrating the capacities of up-scaling this type of systems. Hence, a implicit solvent scheme might exhibit similar behaviour since DMDD is to a certain extent a similar surfactant, although for this work variations on PH of solvent media are not considered. As a final objective at the end of this project, a implicit water coarse-grain model for DMDD will be generated and properly validated based on MD simulations and thermodynamic properties,in order to provide a good approximation to interactions with silica oligomers in a mesoscale model.

but now i want this instead .

4   Experimental Methods

About gromacs
–MD
–MC
–MPI and GPU
About MagiC
–General idea
–IBI + IMC

–MPI and sample size
–Convergence
Experiment 1
–Objective
–Model 1 vs Model 2
–gromacs Simulation Set-up
–MagiC CG M1 and M2 topology generation+Ref RDFs
–IMC process
–gromacs Set-up Reproduction Tests
Experiment 2
–Objective
–gromacs Set-up Up-scaling
–gromacs Set-up Self-assembly Model X(best)
—-AA Low, CG Low
——1k(?) up-scale @ low
——1k(?) up-scale @ high
—-AA Ideal, CG ideal
——1k(?) up-scale @ low
——1k(?) up-scale @ ideal
——1k(?) up-scale @ high
—-AA High, CG High
——1k(?) up-scale @ high
——1k(?) up-scale @ high
Experiment 3
–Silica addition
—-MagiC CG silica topology
—-gromacs Set-up Self-assembly Model X + Si
——1000, 10000 (Maybe more?)

5   Results

Experiment 1
–Model 1 vs Model 2
–gromacs Simulation
–IMC process Convergence
–gromacs Reproduction Tests RDFs and Properties
Experiment 2
–Self-assembly vs concentration Model X
—-Ideal con validation: Low, Ideal, High con comparison
——Ideal @ Low -> validate
——Ideal @ Ideal -> evaluate
——Ideal @ High -> validate
—-Extremes evaluation:
——Low @ Low vs High @ Low -> compare
——High @ High vs Low @ High -> compare
Experiment 3
–Silica addition
–CG silica Properties
–Self-assembly Model X + Si
——1000, 10000 (Maybe more?)

6   Discussion

Experiment 1
–Which is the best Model?
–Why Model X is better than Model Y?

–To what extent Model X represent well the AA model?
–What are the limitations and advantages of Model X?
Experiment 2
–What is the relation between the entropy "error" and concentration?
–Is the concentration really an problem?
–Do you need to run an AA for each con or the Ideal can represent any?
–Even extreme cases can be approximated? In this case how much is the "error"?
Experiment 3
–Is The silica CG model suitable?
–Can you the proprieties validate it?
–The self-assembly behaviour change with silica addition?

7   Conclusion

What did you really see from the results?
There was any bad assumption that is contestable?
What will be the next step to your research?

8   Nomenclature

H	Hamiltonian hdasdgfag (?, ?)a asdfasdfas dfadsfasdfasdfsdfa gsdfhg as djfg ajdf jasdfj adjhg asdjhgfajksdfaj,jahdskjfas
C_n	Molar concentration of species n (mM)

9   Appendix