A Molecular Dynamics Study of Solubility Behavior of Different Glucans in Water

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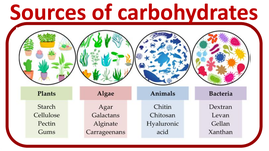
Motivation

- Features like biocompatibility, non-toxicity, biodegradability and wide availability has given rise to the use of polysaccharides in various applications.
- Elucidating the chemical structures can aid in understanding the solubility of these biomaterials, which in turn is useful as most polysaccharides function in aqueous solutions.

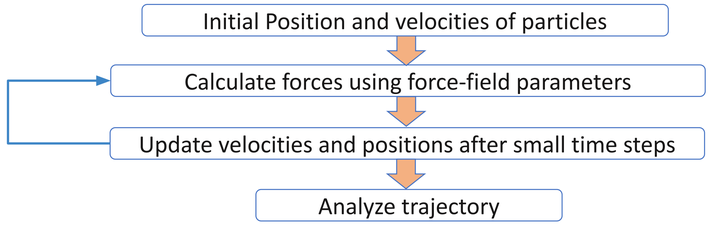
Sources of carbohydrates



Applications

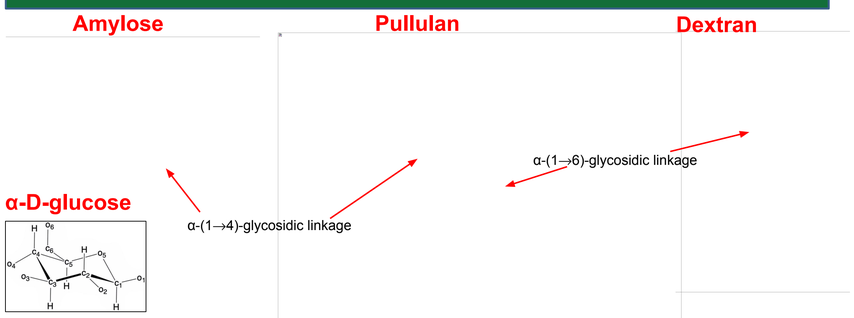
Molecular Dynamics (MD) simulations can be used to better understand polysaccharides at the molecular level

Molecular Dynamics flowsheet



Structure of α-Glucans

Amylose **Pullulan** **Dextran**



- α-glucans are polysaccharides of D-glucose linked with α -(1→X, where X=1, 2, …, 6)
- Pullulan and dextran dissolves readily in water, whereas amylose is poorly soluble in water

Force field

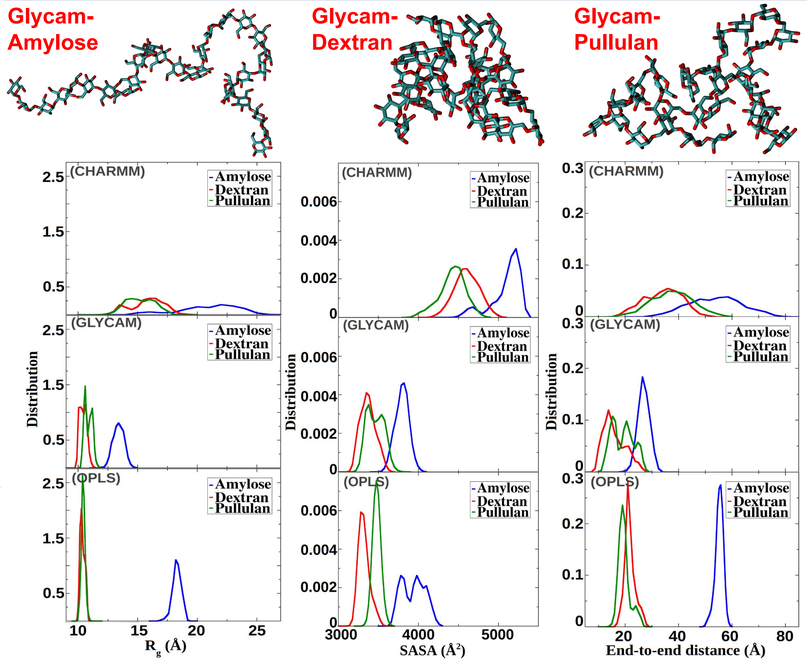
- Through a comparative study, we can evaluate if the FFs being used can properly reflect the system of interest
- We have used **CHARMM**, **GLYCAM_06j** and **OPLS-AA** force fields for MD simulations. **TIP3P** water model was used with CHARMM, GLYCAM FF, and **SPCE** was used with OPLS FF.

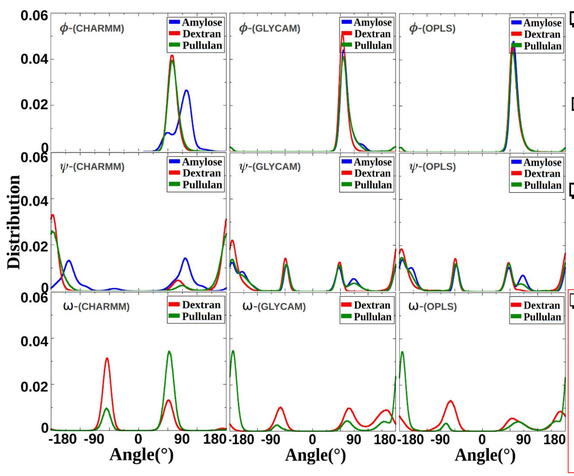
All atom (AA) MD simulation setup

- GROMACS 2020.4 program was used for simulating aqueous systems of single chain glucans (30-mer) at 298K and 1 bar for 100ns
- Initial structures and parameters were generated with the help of CHARMM-GUI and doGlycans tools

Results

Glycam-Amylose **Glycam-Dextran** **Glycam-Pullulan**

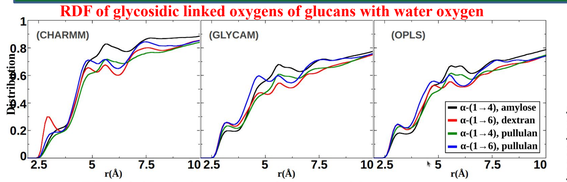




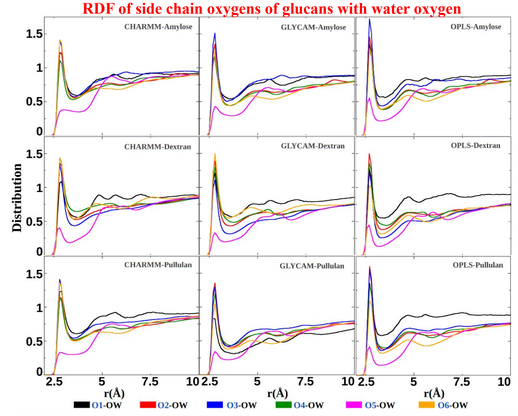
- ϕ(O5`-C1`-O6-C6 for dextran,O5`-C1`-O4-C4 for amylose)
- ψ(C1`-O6-C6-C5 for dextran,C1`-O4-C4-C5 for amylose)
- ω(O6-C6-C5-O5) torsion angle causes additional flexibility for (1→6) linkage
- Torsion angle ψ shows gauche (~-90° and ~+90°) for the glucans with GLYCAM and OPLS FF, which could cause the lower Rg and SASA

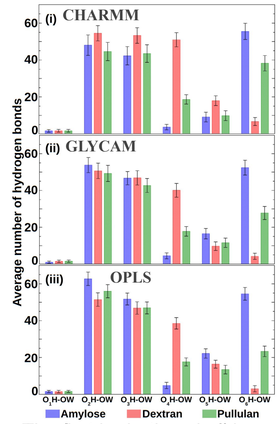
Role of -OH groups and glycosidic oxygen

RDF of glycosidic linked oxygens of glucans with water oxygen



RDF of side chain oxygens of glucans with water oxygen

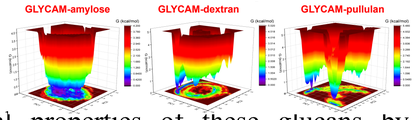




- The first hydration shell is within 2.8-3.2 Å
- Low distribution of water about O_5 and glycosidic oxygens ould be due to steric hindrance and low number of H-bonds formed

Conclusion and future work

- More flexibility in glucans is seen with CHARMM FF over the other two FFs. This could be due to more stretched conformation of torsion angle ψ for CHARMM. The higher intensities beyond first hydration shell for linked oxygen could also be due to ψ.
- We are currently doing ED analysis to find conformational space explored by each glucan



- We **aim to study** rheological properties of these glucans by **developing coarse-grained models**

Acknowledgement

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References

- Guvench, Olgun, et al. *Journal of chemical theory and computation* 5.9 (2009): 2353-2370
- Kirschner, Karl N., et al. *Journal of computational chemistry* 29.4 (2008): 622-655.
- Jorgensen, William L., and Julian Tirado-Rives. *Proceedings of the National Academy of Sciences* 102.19 (2005): 6665-6670.