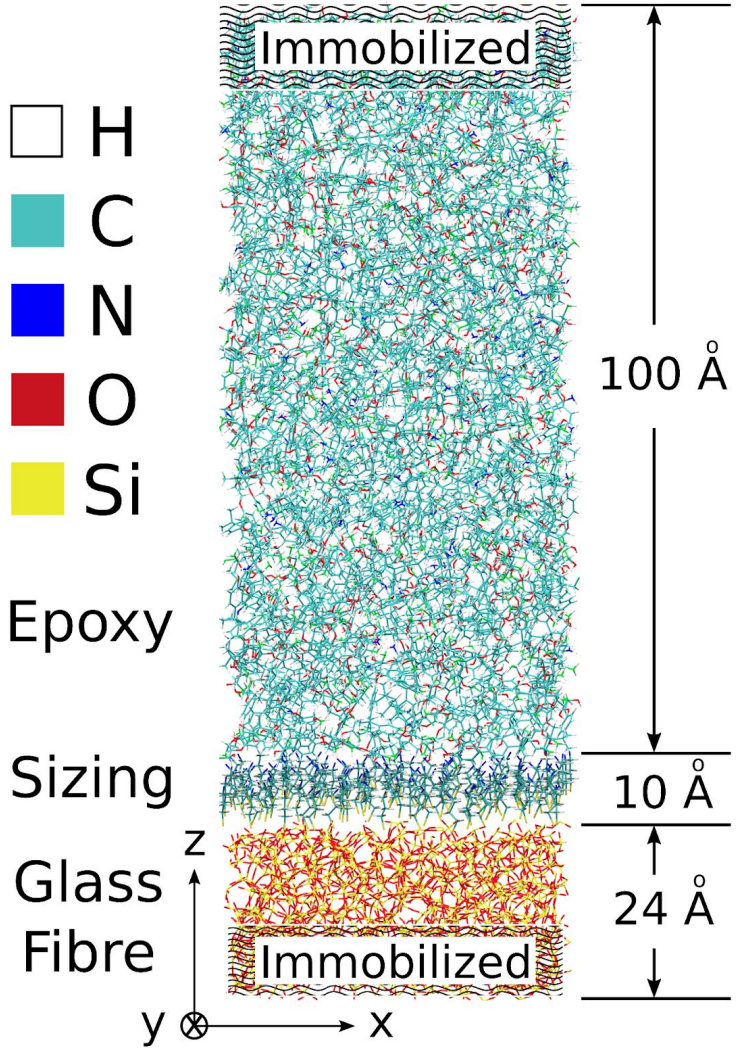
Modelling glass fiber reinforced epoxy composite using molecular dynamics

Composite System

3 Layers

- Crosslinked epoxy resin
- Sizing made of AMinoPropylTriEthoxySilane (AMPTES)
- Amorphous silica glass



Amorphous silica

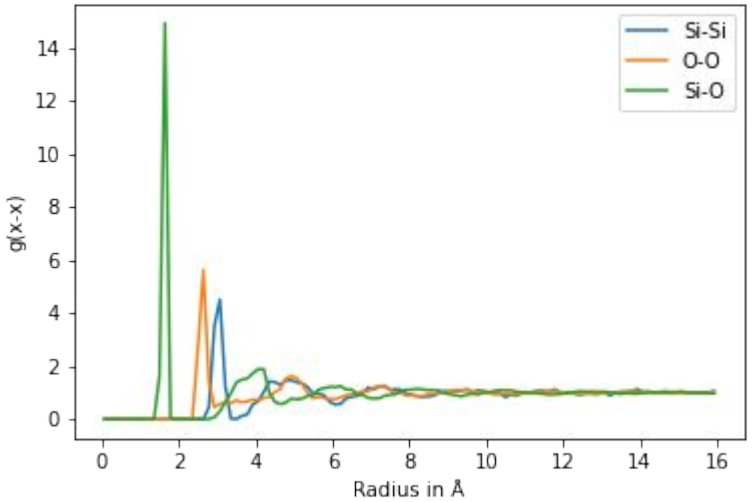
Amorphous silica structure generation:

Steps followed to generate amorphous silica in LAMMPs:

1. Ordered crystalline silica structure was created
2. Vashistha[1] potential was used to describe the interaction among the atoms.
3. NVT at 6000K for 5000 steps
4. NPT from 6000K to 4000K for 50000 steps, P=100atm
5. NPT from 4000K to 300K for 200000 steps, P=100 to 1 atm, over 200ps, cooling rate=18.5K/ps
6. NPT 300K 4000 steps, P=1atm
7. Resulting structure is amorphous silica structure

RDF of amorphous silica generated:

Partial rdfs of Amorphous SiO_2 from our simulation



Partial rdfs of Amorphous SiO_2 from literature[2]

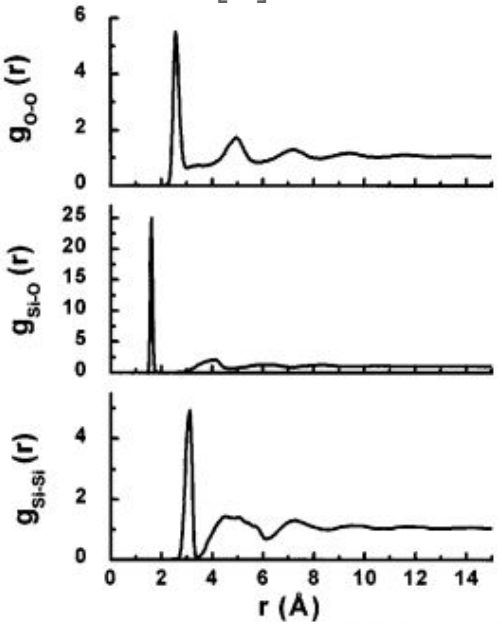
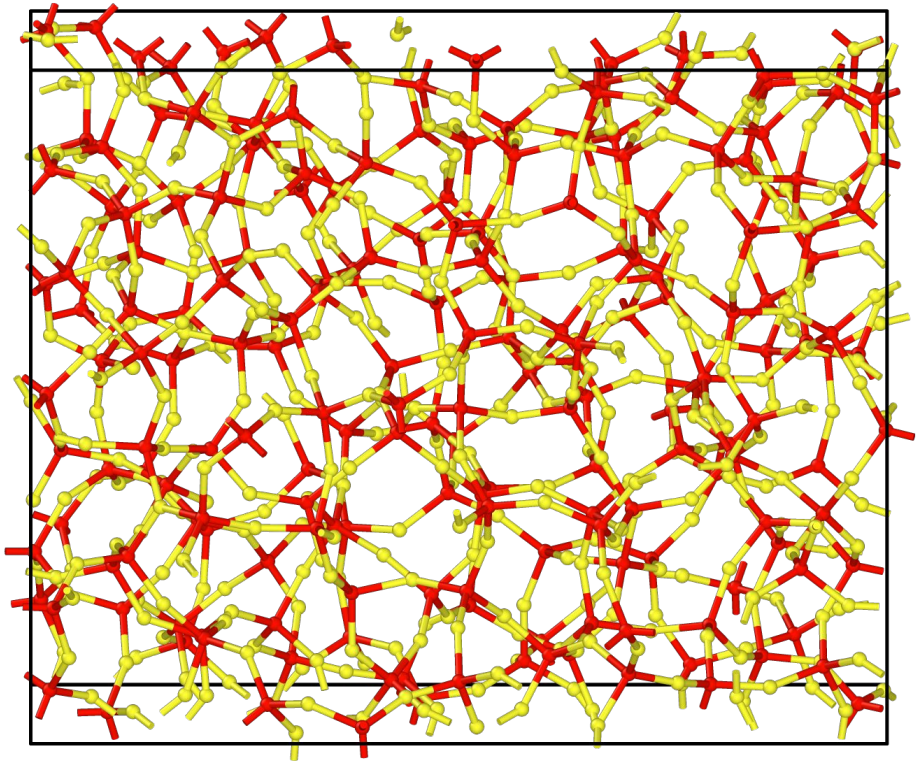


FIG. 1. Partial pair-distribution functions in amorphous silica at 300 K.

Amorphous Silica network structure generated:

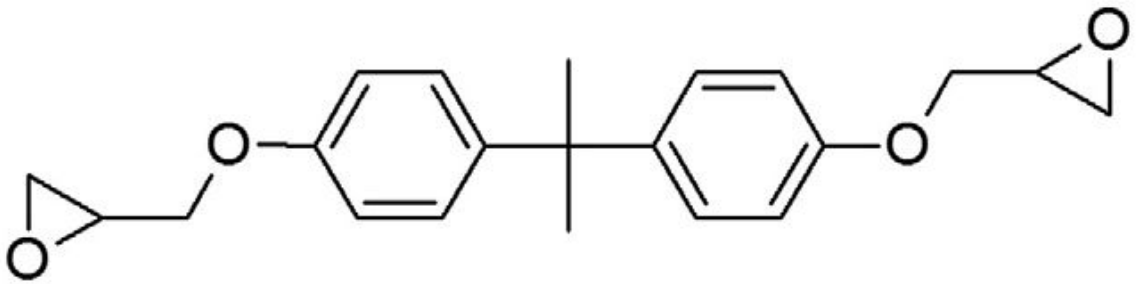


Resulting density-2.51 g/cm3

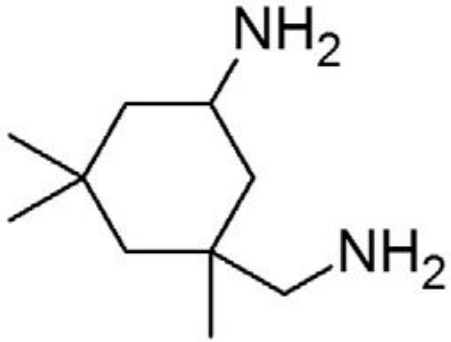
Current focus: Crosslinked Epoxy

Epoxy cross linking explained

Two monomers need to react:

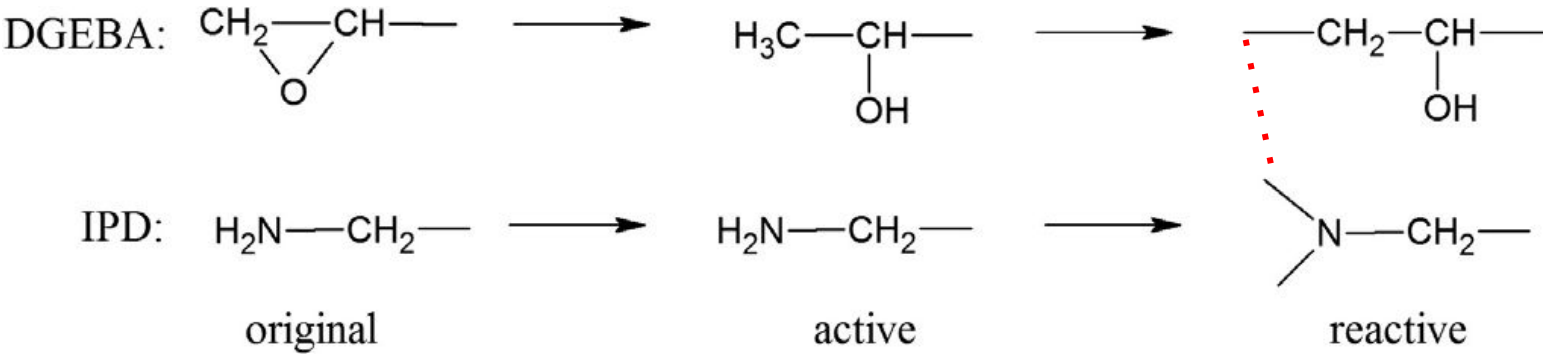


diglycidyl ether bisphenol A (DGEBA)



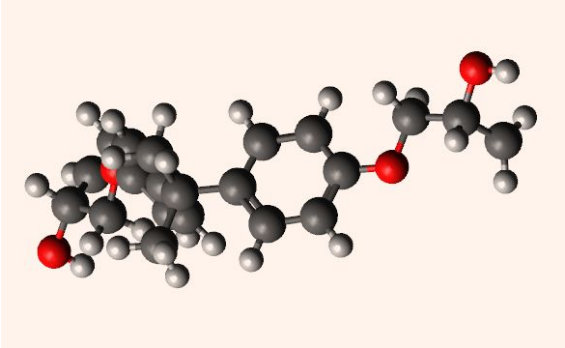
isophorone diamine (IPD)

Creation of reactive sites in the molecules:

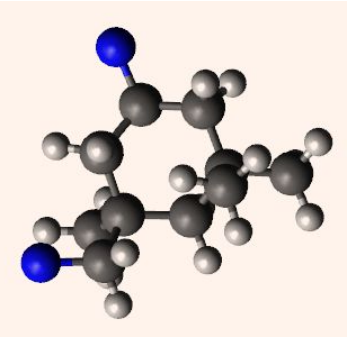


- Exact cross linking mechanism is a complex multistep process which can be difficult to model. So original molecules are modified to make the reaction simpler.
- Reactive sites are created by removing hydrogen atoms from the functional groups
- In the above image *reactive* nitrogen of IPD can link with the *reactive* carbon atom of DGEBA

Reactive molecules thus generated:



Reactive DGEBA



Reactive IPD

First Step:

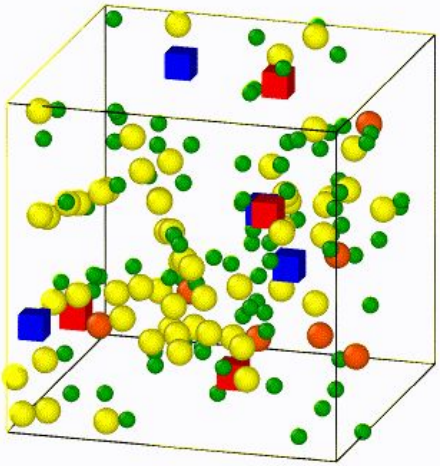
Simulation box was filled with reactive DGEBA and IPD molecules.

Polymer Consistent Force Field(PCFF) was used to define the interactions in the system. It takes into account the energy of each bond, angle, dihedral, the cross interactions among them and the intermolecular forces such as *van der waals* and electrostatic forces between the molecules.

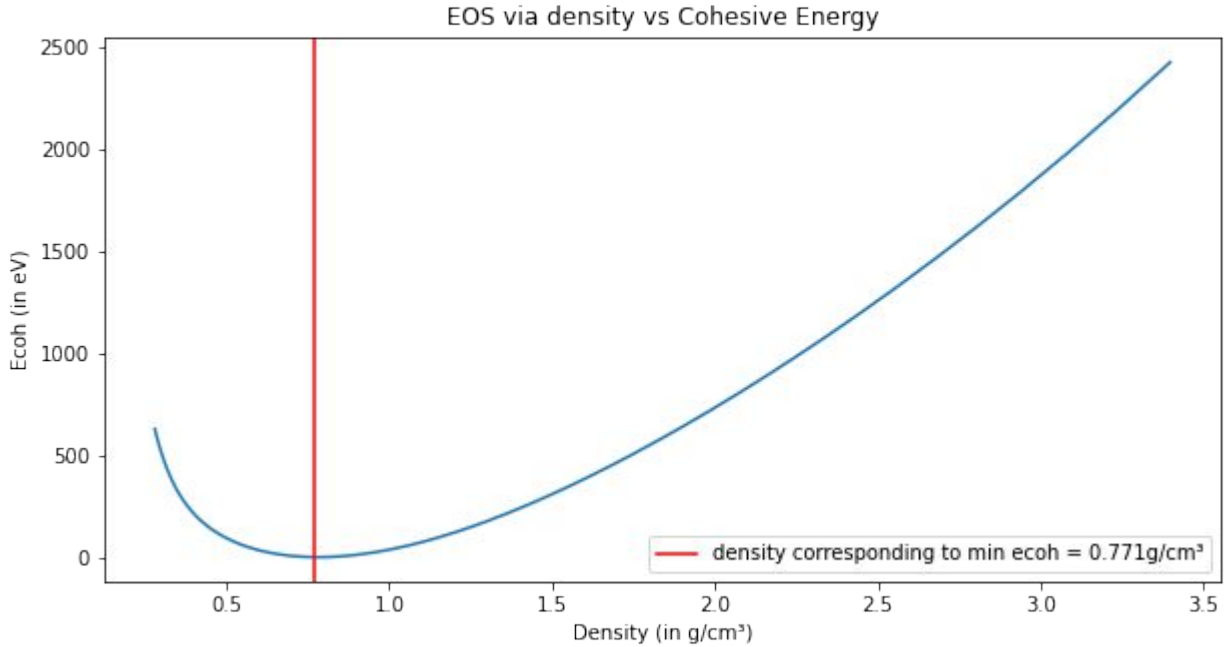
The topology file containing information about atomic coordinates, bonds, angles, dihedrals and the parameter files specifying potential of each bond, angle and dihedral via PCFF, were constructed using Enhanced Monte Carlo(EMC) tool developed by Pieter J. in 't Veld[3]

DGEBA + IPD system before cross linking:

- NVT run on a system containing two molecules each of reactive DGEBA and IPD, at 300K for 10000 fs
- Blue and Red **boxes** represent *reactive Carbon and Nitrogen* Sites.
- Yellow, green and orange dots represent non reacting carbon. hydrogen and oxygen atoms.



Equation of state for the system before cross linking:



Optimal density found to be 0.771 g/cm^3

Cross linking the reactive sites

- Reactive N and C atoms come closer than cutoff radius ➡ bond introduced
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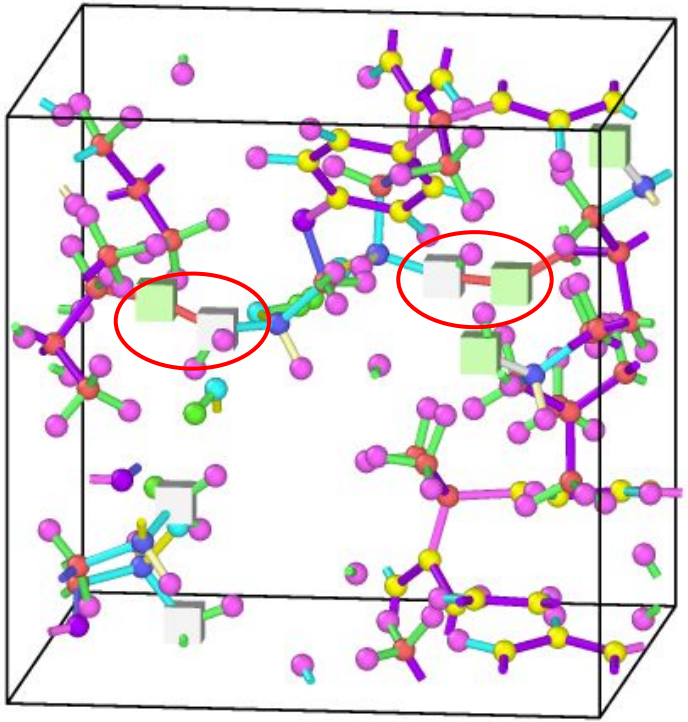

Current & future work:

- *fix bond/create command* in LAMMPs was used to
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References:

[1] Vashishta, P., Kalia, R. K., Rino, J. P., & Ebbsjö, I. (1990). Interaction potential forsio2: A molecular-dynamics study of structural correlations. *Physical Review B*, *41*(17), 12197–12209. https://doi.org/10.1103/physrevb.41.12197

[2] Du, M.-H., Kolchin, A., & Cheng, H.-P. (2004). Hydrolysis of a two-membered silica ring on the amorphous silica surface. *The Journal of Chemical Physics*, *120*(2), 1044–1054. https://doi.org/10.1063/1.1630026

[3] P.J. in 't Veld and G.C. Rutledge, *Macromolecules* 2003, 36, 7358

Supplemental Information

PCFF force field explained:

PCFF forcefield for polymers and other materials

Applicability

PCFF was developed based on CFF91 and is intended for application to polymers and organic materials.

It is useful for calculating mechanical properties, compressibilities, heat capacities, elastic constants.

$$E_{pot} = \sum_b \left[K_2(b-b_0)^2 + K_3(b-b_0)^3 + K_4(b-b_0)^4\right] \quad (1)$$

$$+ \sum_\theta H_2(\theta-\theta_0)^2 + H_3(\theta-\theta_0)^3 + H_4(\theta-\theta_0)^4 \quad (2)$$

$$+ \sum_\phi \left[V_1\left[1-\cos\left(\phi-\phi_1^0\right)\right] + V_2\left[1-\cos\left(2\phi-\phi_2^0\right)\right] + V_3\left[1-\cos\left(3\phi-\phi_3^0\right)\right]\right] \quad (3)$$

$$+ \sum_\chi K_\chi \chi^2 + \sum_b \sum_{b'} F_{bb'}(b-b_0)(b'-b'_0) + \sum_\theta \sum_{\theta'} F_{\theta\theta'}(\theta-\theta_0)(\theta'-\theta'_0)$$

(4) (5) (6)

$$+ \sum_b \sum_\theta F_{b\theta}(b-b_0)(\theta-\theta_0) + \sum_b \sum_\phi (b-b_0)\left[V_1\cos\phi + V_2\cos 2\phi + V_3\cos 3\phi\right]$$

(7) (8)

$$+ \sum_{b'} \sum_\phi (b'-b'_0)\left[V_1\cos\phi + V_2\cos 2\phi + V_3\cos 3\phi\right] \quad (9)$$

$$+ \sum_\theta \sum_\phi (\theta-\theta_0)\left[V_1\cos\phi + V_2\cos 2\phi + V_3\cos 3\phi\right] \quad (10)$$

$$+ \sum_\phi \sum_\theta \sum_{\theta'} K_{\phi\theta\theta'}\cos\phi(\theta-\theta_0)(\theta'-\theta'_0) + \sum_{i>j} \frac{q_i q_j}{\varepsilon r_{ij}} + \sum_{i>j}\left[\frac{A_{ij}}{r_{ij}^9} - \frac{B_{ij}}{r_{ij}^6}\right]$$

(11) (12) (13)