Designing Atomistic Simulation Workflows with MedeA® A Case Study: Viscoelasticity of Glycerol at Ultra-high Frequencies

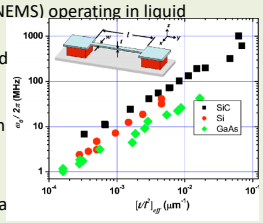
materials design

Naida Lacevic
Materials Design, Inc.,
materialsdesign.com, nlacevic@materialsdesign.com

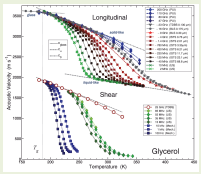
Atomistic simulations are increasingly used to complement or enhance experiments

Example: Nanoelectromechanical systems (NEMS) operating in liquid environments are of interest in biology[1].

- Resonance frequency increase into GHz and THz regime
- Same order of magnitude as atomistic/molecular relaxation processes in liquids such as water and glycerol
- Viscoelasticity becomes important

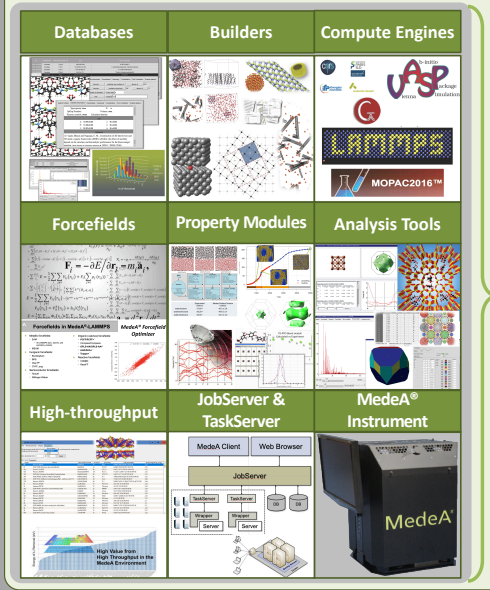


- It is challenging to obtain relevant data via experiments
  - E.g., DMA (dynamic mechanical analysis) cannot probe GHz regime
- Only one experimental study of G' and G'' of glycerol at 25GHz (shear) and 41.5GHz (long.) – Nelson et al., JCP[2]
  - Complex technique (TDBS)
  - Fluid structure not available
  - Difficult to gain insights into molecular motion
- Molecular dynamics simulations offer alternative
  - We examined glycerol, a model simple viscous liquid, under ultra-high frequency shear and longitudinal deformation.[3]
  - Many simulations needed to sweep temperature and frequency parameter space.



To obtain trends and compare them with experiments, access to tools that automate workflow increases productivity.

Tools in MedeA® automate complex workflows and ease mundane tasks of initial configuration construction

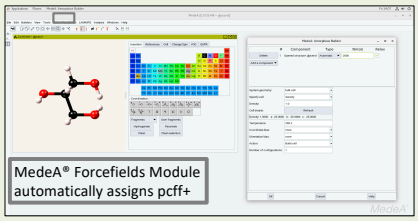


A Typical Workflow

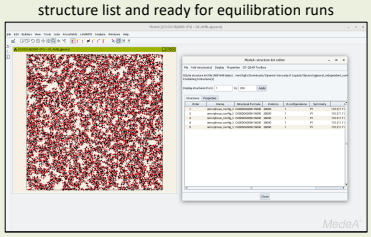
1. Retrieving, viewing, and editing structures
2. Selecting methods and programs
3. Setting up computational parameters
4. Performing calculations
5. Analyzing results
6. Storing results

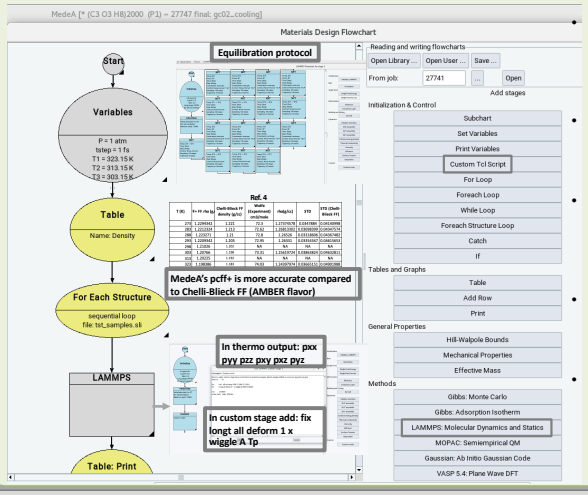
Model construction and equilibration/production run protocols

MedeA Molecular Builder and MedeA Amorphous Materials Builder are used for glycerol model constructions



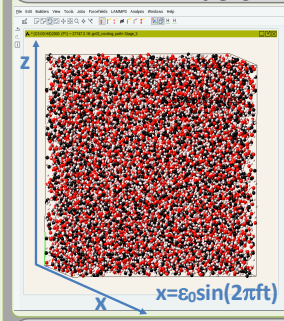
Independent configurations created with MedeA Amorphous Materials Builder are saved in structure list and ready for equilibration runs





- MedeA® environment provides graphical flowcharts to support the efficient construction of complex computational protocols (Ref. 5).
- E.g., the Foreach Structure Loop takes selected structures from a previous trajectory and performs the same flowchart on them.
- Custom-scripting steps can be added to flowcharts using the Tcl language, allowing almost unlimited extensions to be made to flowcharts.
- LAMMPS log file was processed via octave scripts, which can be called from Tcl custom stage, to obtain moduli.
- This computational procedure can be used to produce a range of input molecular systems, applying exactly that same computation protocol in every case.

Schematics of longitudinal and shear deformations



Impose strain amplitude ε_0 and f:

$$\varepsilon(t) = \varepsilon_0 \sin(2\pi f t + \delta_e)$$

Compute stress:

$$\sigma(t) = \sigma_0 \sin(2\pi f t + \delta_s)$$

σ_0 and δ_s are computed as a discrete Fourier transform (DFT) of stress magnitude and phase.

$$M' = \frac{\sigma_0}{\varepsilon_0}\cos\delta \quad M'' = \frac{\sigma_0}{\varepsilon_0}\sin\delta$$

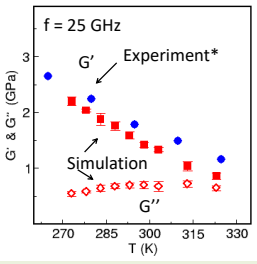
$$\delta = \delta_s - \delta_e$$

The shear modulus, G, is calculated in a similar manner

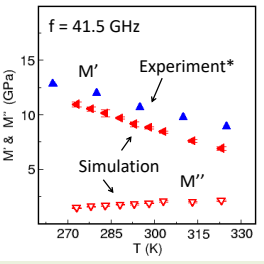
Results and comparison with experiments

The simulated temperature trends of G' and M' line up with experimental measurements

Shear Moduli



Longitudinal Moduli



Summary

- High-throughput and high-fidelity modeling enabled by MedeA® provides guidance to screen large numbers of design options for materials before committing to experiments.
- MD is an effective tool for predicting the viscoelastic properties of simple liquids at ultra-high frequencies.

References

(1) K. L. Ekinci and M. L. Roukes, Nanoelectromechanical systems, Review of Scientific Instruments, 2005. 76
(2) Klieber, C., et al., Mechanical spectra of glass-forming liquids. II. Gigahertz-frequency longitudinal and shear acoustic dynamics in glycerol and DC704 studied by time-domain Brillouin scattering. JCP, 2013. 138(12).
(3) N.M. Lacevic and J. E. Sader, Viscoelasticity of glycerol at ultra-high frequencies investigated via molecular dynamics simulations. JCP, 2016. 144(5)
(4) Wolfe, M. and J. Jonas, Reorientational Motions in Compressed Viscous Fluids - Selectively Deuterated Glycerol. JCP, 1979. 71(8).
(5) MedeA® version 3.0. MedeA® is a registered trademark of Materials Design, Inc., Angel Fire, New Mexico, USA.