Polymer Physics
HW 13
April 15, 2022 (Due April 19)

Machine learning (ML) has been proposed as an advantageous tool for dealing with multidimensional space and large data sets. Shen Z-H, Bao Z-W, Cheng X-X, Li B-W, Liu H-X, Shen Y, Chen L-Q, Li X-G, Nan C-W *Designing polymer nanocomposites with high energy density using machine learning* Nature Computational Materials **7** 110 (2021) describes a method for the design of polymer nanocomposite capacitors.

a) What are the advantages of the ML method for Shen's study in terms of his conclusion in Figure 2 of the supplemental material. Consider that Osada M, Sasaki T *The rise of 2D dielectric/ferroelectrics* APL Materials **7** 120902 (2019) published a review article showing that Ca based perovskite crystals were useful dielectrics due to their sheet structures (Table 1). It is noted in the article by Shen that it was previously known that nanocomposites can overcome some problems with dielectric breakdown and dispersion at low concentrations of nanofiller and that it is known that the higher dielectric constant materials such as Ca perovskites will have better performance as capacitors.

b) Define some of the terms used by Shen on page 2: "inversely-design polymer nanocomposite", "scoring function", "back propagation neural network". Explain in detail the scoring function used by Shen and how it is used in the scheme of Figure 4 (center of figure).

c) Xu H, Sheridan RJ, Brinson LC, Chen W, Jiang B, Papakonstantopoulos G, Polinska P, Burkhart C *Chapter 11 Data-Driven Multiscale Science for Tire Compounding: Methods and Future Directions* 281-312 in "Theory and Modeling of Polymer Nanocomposites" Eds. Ginzburg VV, Hall LM, Springer Series in Materials Science **310** (2021) discusses the use of data analytics and machine learning in the design of tires focusing on the tan δ peak. Data analytics is used to construct representative volume elements (RVE) reflecting the microstructure and ML is used to reconstruct the interphase structure and the tan δ peak. Xu first discusses a simple model for the tan δ peak shown in figure 11.4. Explain how each of the prony series elements has an associated relaxation time, give the shape of G' and G" and tan δ in frequency at a fixed temperature from each of these elements and explain how a peak in tan δ could be shifted with increasing filler content from the overall model.

d) Xu then uses the method of Deng (Deng H, Liu Y, Gai D, Dikin DA, Putz KW, Chen W, Brison LC, Burkhart C, Poldneff M, Jiang B, Papakonstantopoulos GJ *Utilizing real and statistically reconstructed microstructures for the viscoelastic modeling of polymer nanocomposites* Comp. Sci. Tech. **72** 1725-1732 (2012)) to account for microstructure from micrographs and the interphase from shifts in the tan δ peak. Describe Deng's method and compare it with X-ray scattering measurements that can generate a correlation function over five orders of size from 1 Å to 5 µm.

e) Xu describes filler in terms of loading percent, "dispersion", and "geometry". Consider that carbon black filler is a hierarchical structure composed of primary particles, aggregates, agglomerates, and a carbon agglomerate network on the millimeter scale, and that "dispersion" has different meanings on all these size scales as does "geometry". Further, consider that the one parameter, tan δ at a single temperature or frequency, strain

amplitude and deformation geometry (shear strain) might not completely describe the behavior of a reinforced elastomer. For instance, how is tear resistance modeled with this approach, or the Mullins or Payne effects? Critique Xu's simplification. How realistic/feasible is the scheme shown in Figure 11.20 in this context?