LETTER

Reply to Elmatad: Supercooled viscous liquids display a fragile-to-strong dynamic crossover

Theory predicts a fragile-to-strong (FS) dynamic crossover temperature T_x in supercooled liquids but, contrary to what is reported in ref. 1, T_x must be $>T_g$ (2), where T_g is the glass transition temperature. Ref. 4 of ref. 1 hypothesizes that a parabolic form is valid in a range $T_o > T > T_x$, where T_o is defined as an onset temperature that marks the crossover from normal liquid behavior to supercooled liquid behavior. A second paper by the same authors (ref. 5 of ref. 1) proposes the range of the hypothesized parabolic behavior can be extended to cover $T < T_x$. Further, both refs. 4 and 5 of ref. 1 state that above T_o, the temperature dependence of transport coefficients is nearly temperature independent. This statement contradicts the experimental data—indeed, visual inspection of the figures in refs. 4 and 5 of ref. 1 indicates that, in very limited temperature intervals of $T > T_o$, changes of three to four orders of magnitude take place in transport parameter values. Moreover, glass transition theories such as mode coupling theory (2) consider these data to be extremely relevant.

Many experimental data are not consistent with the hypothesized parabolic fit. A particularly striking example is the failure to fit the extensive viscosity data on salol that exist from several independent laboratories (see ref. 4 and references therein). To complement the analysis of 84 different liquids reported in ref. 3 and to explicitly compare the parabolic hypothesis and our FS dynamic crossover hypothesis, Fig. 1 plots all available salol data for both relaxation time and viscosity. One sees that the parabolic fit is significantly less satisfactory than our fit to a FS dynamic crossover between non-Arrhenius behavior at high temperature and Arrhenius at low. In particular, we note our fit of Fig. 1 to all of the salol data below T_x uses two parameters, not three as claimed in ref. 1.

In summary, our result that is perhaps most relevant to the claims of ref. 1 is presented graphically in figure 3 of ref. 3: a log-log plot of the self-diffusion constant as a function of the shear viscosity demonstrated that the universality of the FS dynamic crossover emerges directly from the experimental data *independent* of any model or hypothesis. Rather than a comment on our work, ref. 1 appears to be more focused on defending the "parabolic form hypothesis," which recent literature (see, e.g., ref. 5 and literature cited therein) has demonstrated to be of limited validity compared with the equation we use to describe the non-Arrhenius dynamics of supercooled fluids.

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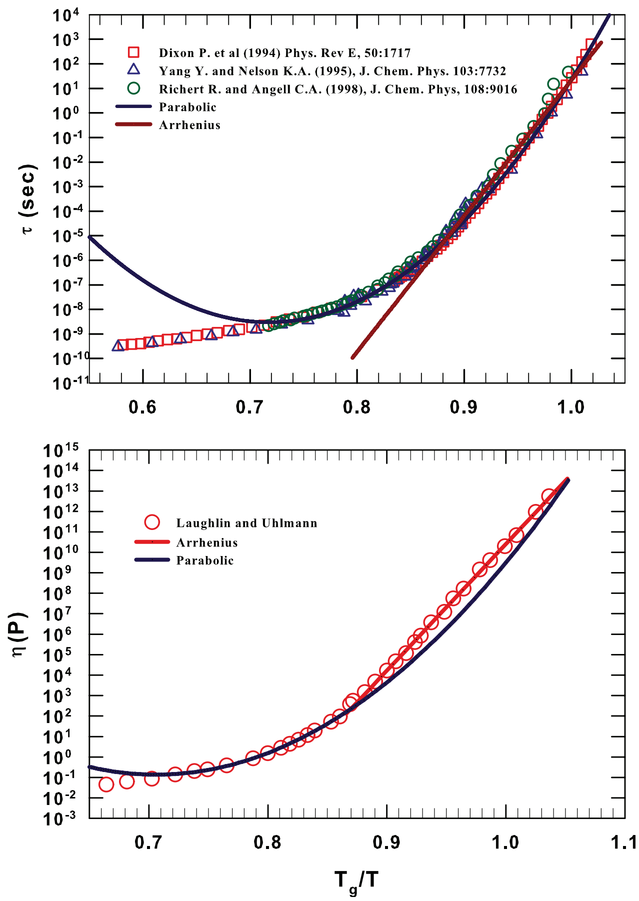

Fig. 1. Two salol transport quantities, the relaxation time $\tau(T)$ obtained from light scattering and dielectric relaxation data (*Upper*) and the shear viscosity $\eta(T)$ obtained from the data of ref. 4 (*Lower*). All of the data are fit using two mutually exclusive data fitting procedures: (*i*) the "parabolic hypothesis," using the same parameter values used in ref. 1, and (*ii*) the FS dynamic crossover approach of ref. 3, fitting the data for $T_g/T > 0.88$ ($T < T_x$).

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The authors declare no conflict of interest.

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