Comment on "Anomalies in the Scaling of the Dielectric α-Relaxation"

Recently Schonhals *et al.* [1] reported dielectric measurements on four materials spanning over 15 decades in frequency. They wrote that "at temperatures which are sufficiently high ... the dependence of f_p [the peak frequency of $\epsilon''(f)$] upon temperature should be governed by an Arrhenius equation [2]." Their Fig. 2 shows f_p on an Arrhenius plot, where the high-temperature data are fitted by an Arrhenius equation $[f_p(T) = C\exp(B/T)]$ and the low-temperature data to a Vogel-Fulcher-Tammann (VFT) equation $(f_p = C\exp[B/(T - T_0)])$. Extrapolation of the two fits then identifies a crossover temperature T_A. This procedure has been described in previous publications where the crossover temperature T_A has been related to the physics of the liquid-glass transition [3,4].

Other authors, however, have found that Arrhenius behavior holds at *low* temperatures and VFT at *high* temperatures. Cohen and Grest [5] proposed an equation for the temperature dependence of the viscosity based on their free volume approach, which reduces to the VFT form at high T and becomes Arrhenius as $T \to T_g$. They showed that their equation fits published $\eta(T)$ data quite well for six different materials.

An unbiased approach to resolving the paradox implied by these two contradictory views of the temperature dependence of T_0 was suggested by Dixon [6] who fit dielectric $f_p(T)$ data for salol to a VFT function in a sliding three-decade frequency window. Dixon found that T_0 from these fits crosses over from $T_0 \sim 220$ K for high T to $T_C \sim 140$ K at low T but never becomes Arrhenius [7]. Dixon's result is clearly incompatible with the salol fits of Schonhals *et al.* [1], which imply that T_0 decreases toward zero with increasing T.

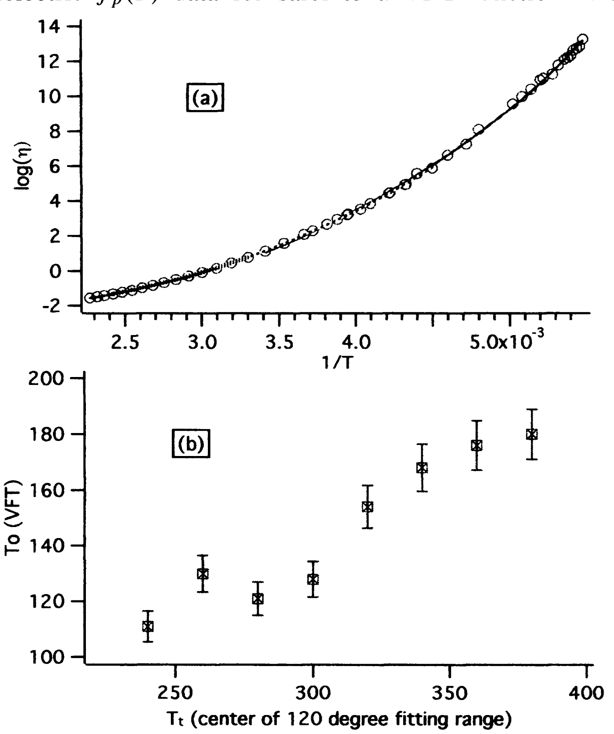

FIG. 1. (a) Points: composite viscosity data $\eta(T)$ [8]. Lines: VFT fits to sections of the data limited to $T_t \pm 60°$. (b) Values of T_0 found from the VFT fits to $\eta(T)$.

We have applied Dixon's method to published viscosity data for glycerol, one of the materials investigated by Schonhals *et al.* Viscosity data from several sources [8] spanning the range 182 to 440 K were combined with small adjustments in the regions of overlap. A single global VFT fit gave $T_0 = 121$ K. We then performed individual VFT fits to portions of the data covering 120 K intervals centered on temperatures T_t from 240 to 380 K in 20° steps. In Fig. 1(a) we show the $\eta(T)$ data together with the eight VFT fits. The T_0 values determined from these fits are plotted against T_t in Fig. 1(b). The value of T_0 resulting from these fits systematically *increases* with increasing temperature, contrary to the trend suggested by Schonhals *et al.*

It would be most interesting to see the $f_p(T)$ data of Schonhals *et al.* reanalyzed following the Dixon procedure, since their high-precision dielectric data should provide better evidence than the viscosity data we have examined for the direction of the trend of $T_0(T)$ with increasing temperature.

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