Supplementary Information (SI) for Journal of Materials Chemistry C.

Supporting information for

Physical aging of glasses of an organic semiconductor

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This supporting information file includes:

1. Temperature profiles for aging experiments

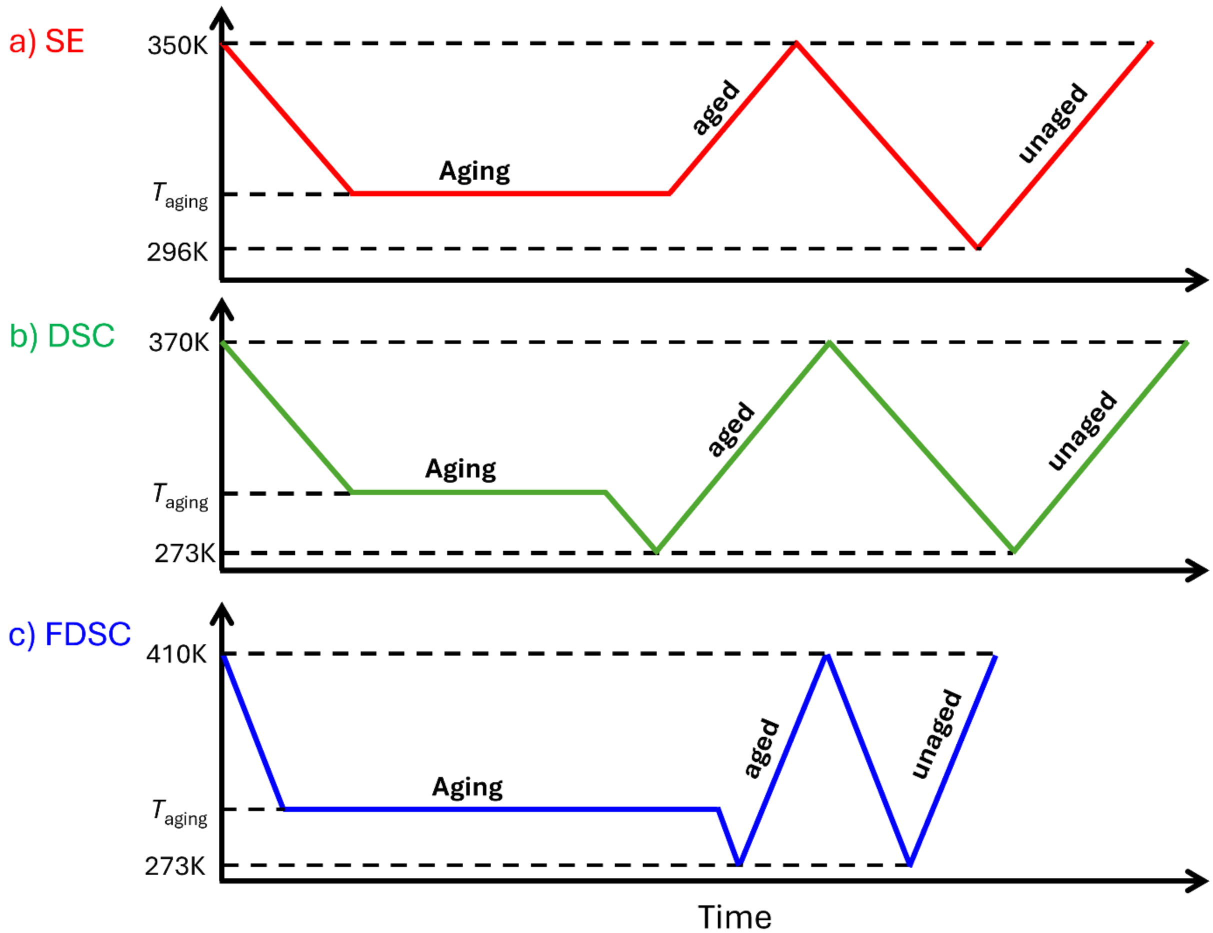

Fig. S1: The temperature profiles of aging experiments on liquid-cooled TPD glasses for a) SE, b) DSC, and c) FDSC measurements.

2. Representative fitting parameters of the isotropic Cauchy model for the liquid-cooled TPD film

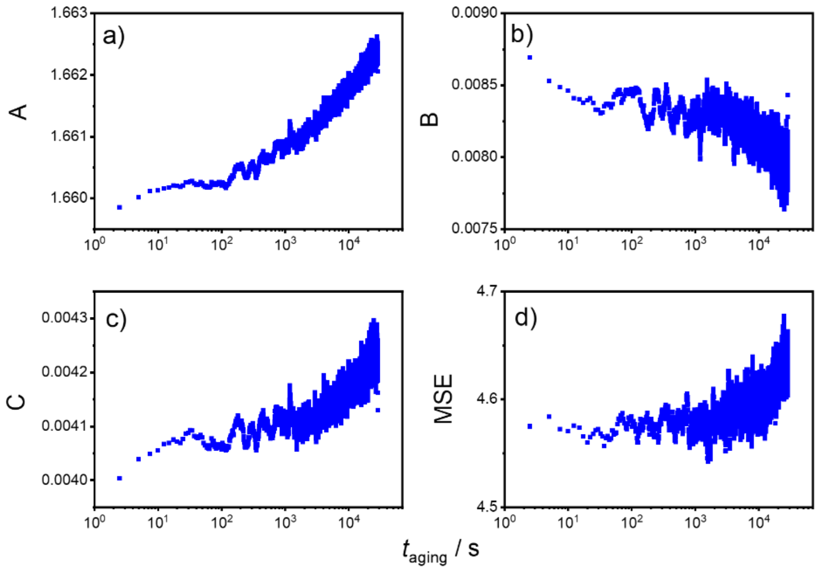

Fig. S2: Representative isotropic Cauchy model parameters for a 400 nm liquid-cooled TPD film as a function of aging time at T_{aging}=318K. **d)** The obtained Mean Squared Error (MSE) between the model and experimental data.

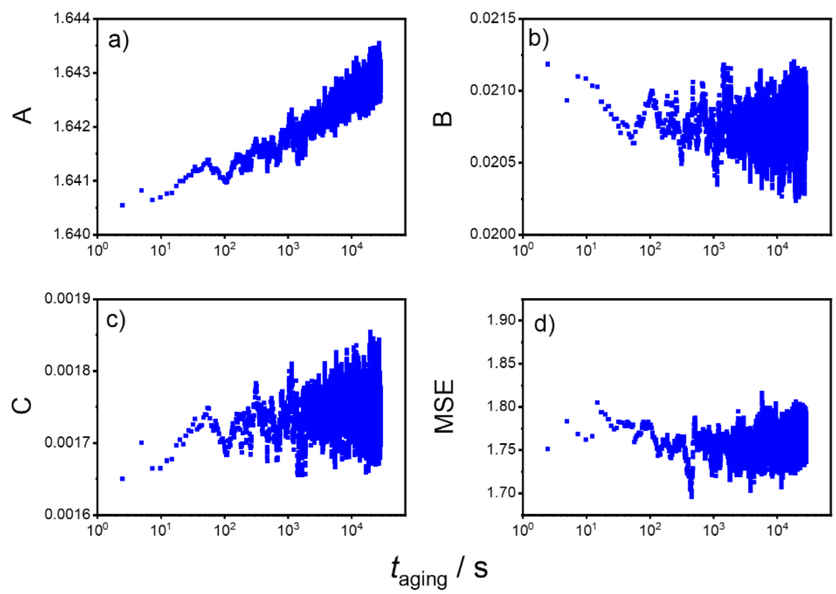

Fig. S3: Representative isotropic Cauchy model parameters for a 100 nm liquid-cooled TPD film as a function of aging time at T_{aging}=318K. **d)** The obtained Mean Squared Error (MSE) between the model and experimental data

3. Representative fitting parameters of the anisotropic Cauchy model for vapor-deposited TPD film

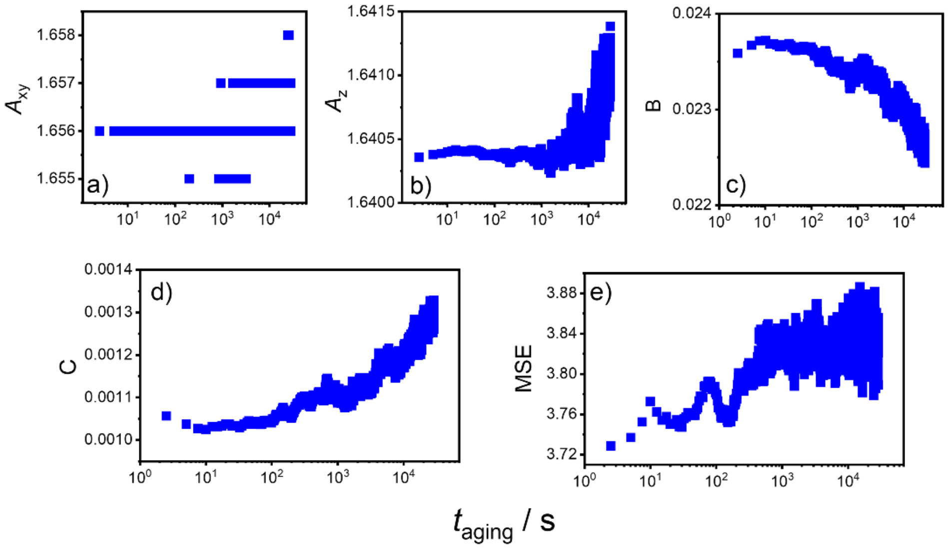

Fig. S4: Representative anisotropic Cauchy model parameters for a 400 nm vapor-deposited TPD film as a function of aging time at T_{aging}=318K. **d)** The obtained Mean Squared Error (MSE) between the model and experimental data.

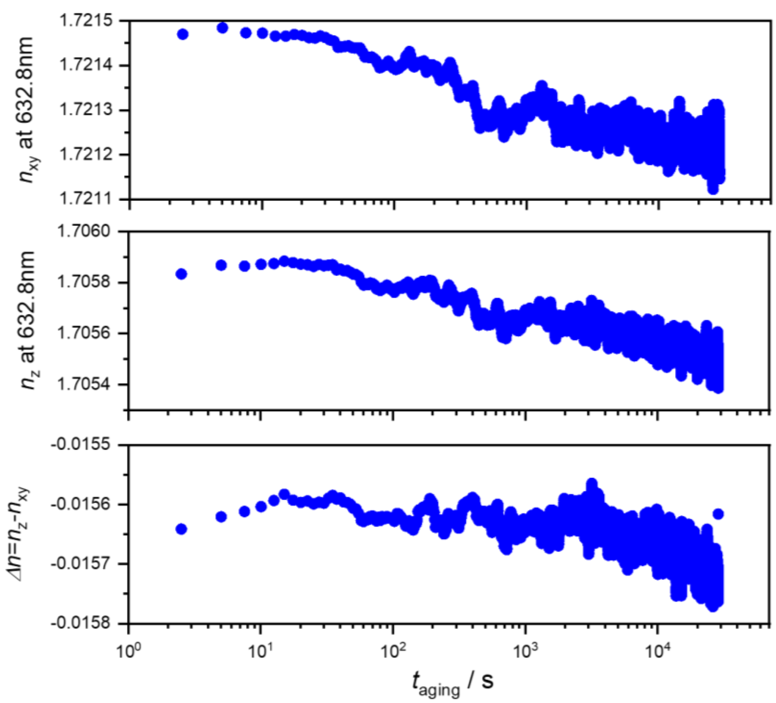

Fig. S5: The evolution of refractive indices **a)** n_{xy} and **b)** n_z at 632.8 nm with aging time for a 400 nm vapor-deposited TPD film at T_{aging}=318K. **c)** The determined birefringence, $\Delta n = n_z - n_{xy}$, at 632.8 nm as a function of aging time for vapor-deposited TPD film at T_{aging}=318K.

4. Refractive index for TPD films in glassy and supercooled liquid states

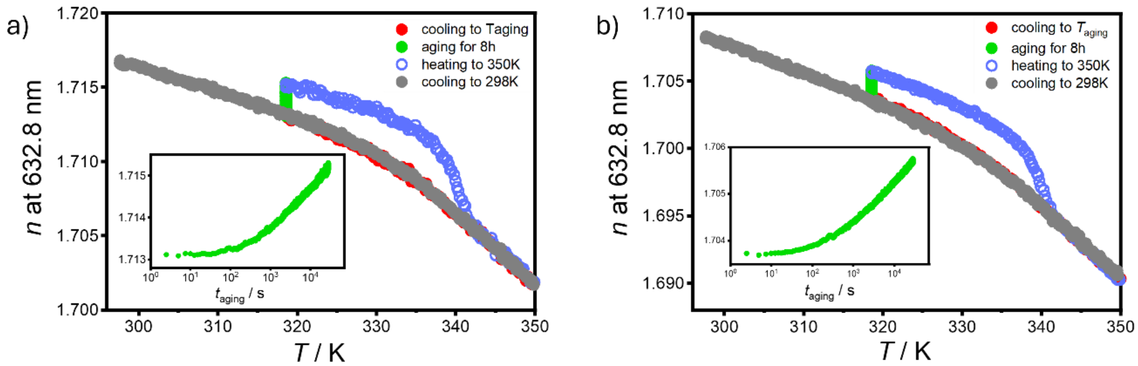

Fig. S6: The refractive index n at 632.8 nm as a function of temperature during an aging experiment at T_{aging}=318K for **a)** 400 nm and **b)** 100 nm TPD glasses and supercooled liquid.

5. Determination of T_f of TPD films from SE measurements

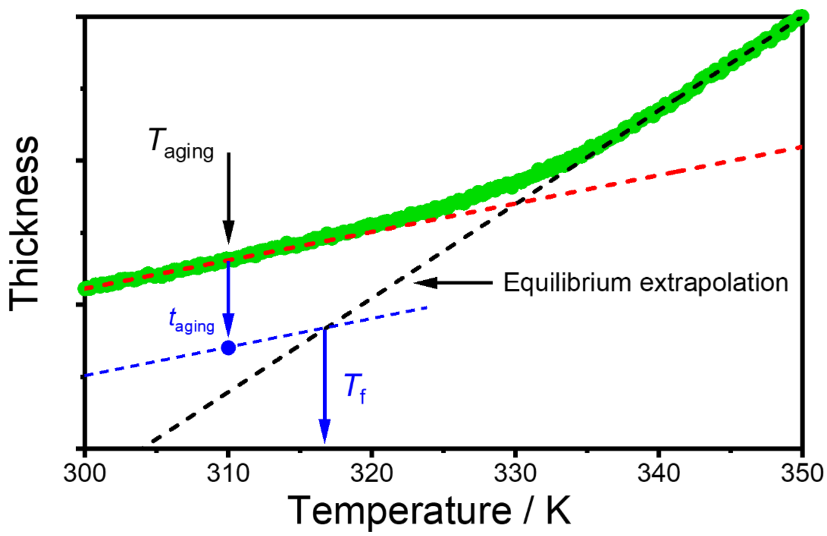

Fig. S7: Illustration of the method used to calculate T_f of aged TPD glasses at a given aging temperature T_{aging}. The blue dot represents the glass film thickness at T_{aging} after aging for t_{aging}. The dashed blue line indicates the predicted evolution of the film thickness with temperature of the aged glass, characterized by the expansion coefficient G determined from Eq. 1. The function of this blue line can be described as h-h_{aging}=G×(T-T_{aging}), where h_{aging} is the glass film thickness after aging for t_{aging} at T_{aging}. The black line represents the extrapolated thickness of the supercooled liquid with expansion coefficient M determined from Eq. 1. According to the definition of T_f, its value corresponds to the intersection of the blue and black lines. Therefore, h_{aging} as a function of t_{aging} at a specific T_{aging} determined from SE measurements is converted into T_f as a function of t_{aging}.

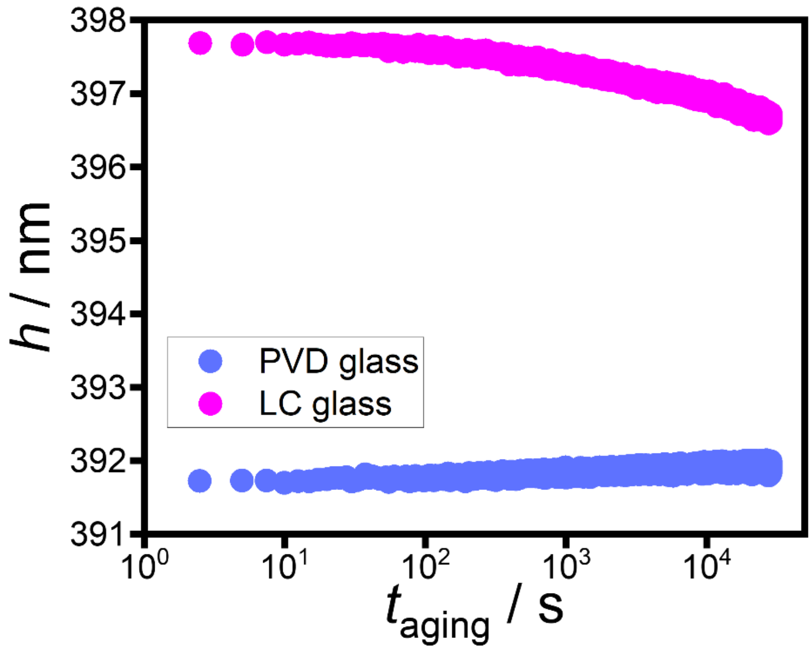

Fig. S8: The thickness as a function of aging time for vapor-deposited (blue) and liquid-cooled (pink) TPD thin films at T_{aging}=318 K.

6. Physical aging rate of liquid-cooled TPD films

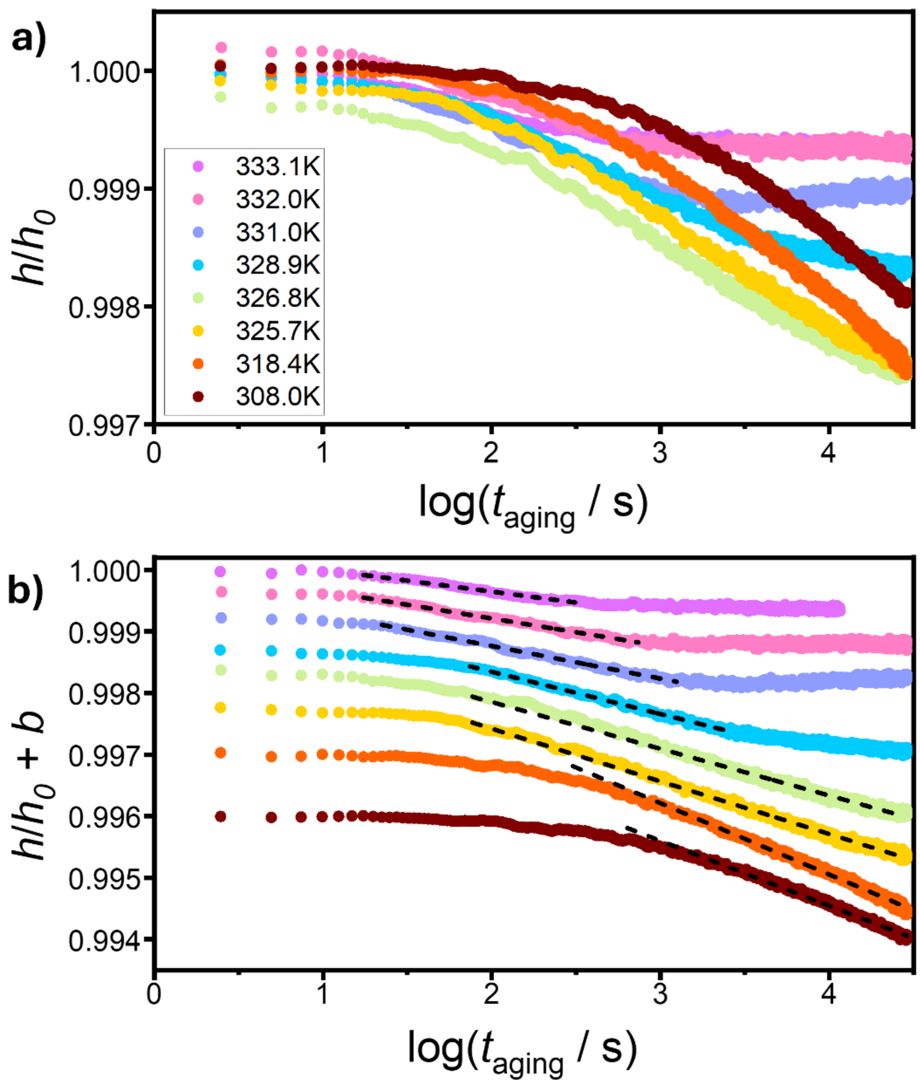

Fig. S9: a) Normalized thickness is plotted as a function of log(t_{aging}) of liquid-cooled TPD films at various aging temperatures. h_0 is the film thickness at t_{aging}=0 s; **b)** Illustration of the linear fits (dashed lines) to obtain physical aging rates of liquid-cooled TPD films at various aging temperatures. The *Y* axis is shifted by an amplitude of *b* for the presentation purposes.

7. DSC and FDSC thermograms of bulk TPD glasses

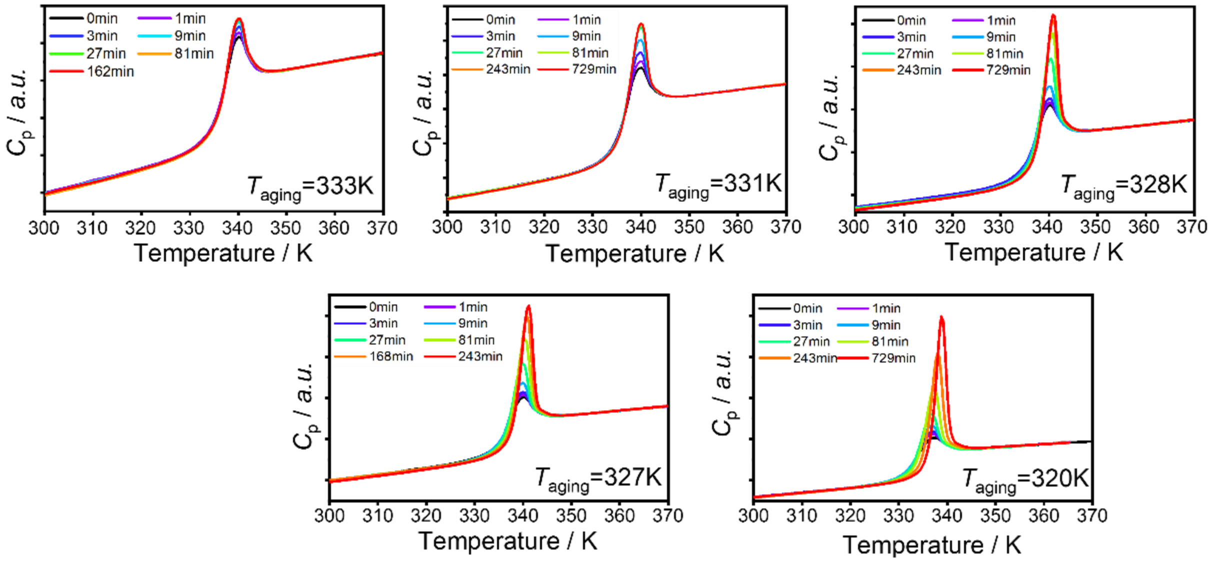

Fig. S10: DSC thermograms of bulk TPD glasses after aging at 333 K, 331K, 328K, 327K, and 320K, for the indicated aging times.

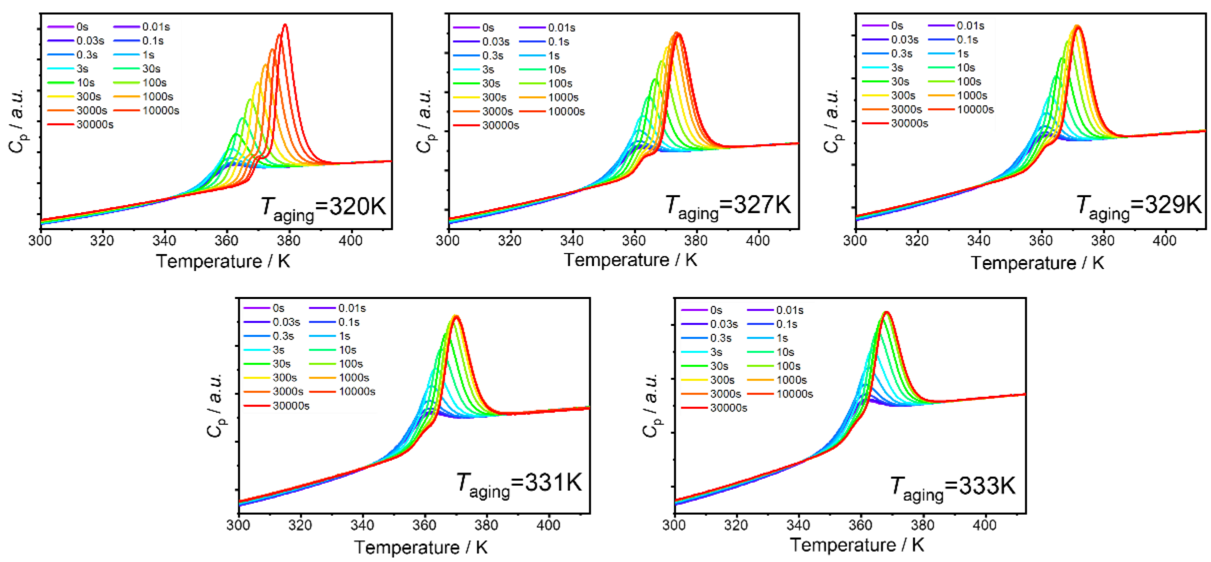

Fig. S11: FDSC thermograms of bulk TPD glasses after aging at 320 K, 327K, 329K, 331K, and 333K for the indicated aging times.

8. Examples for determining T_f of bulk TPD glasses from DSC and FDSC measurements

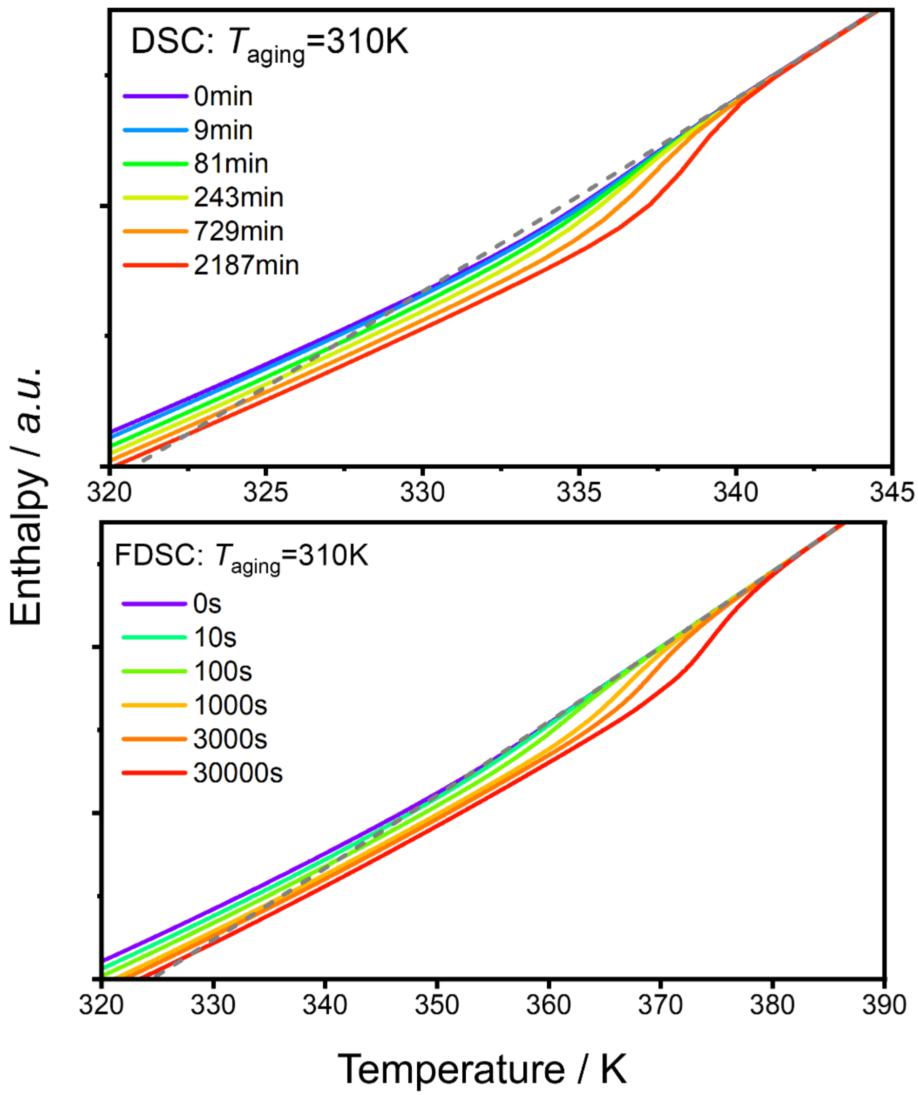

Fig. S12: Enthalpy as a function of temperature for bulk TPD glasses aged at 310K for various aging times. The dashed lines present the extrapolated enthalpy of supercooled liquid of TPD using a second-order polynomial function: ΔH=a+b×T+c×T^2. The T_f values are determined as the intersections of the dashed gray and the solid lines, which present the temperature-dependent enthalpy of the TPD glasses.