**Jumping In and Out of the Phase Diagram Using Electric Fields:**
**Timescale for Remixing of Polystyrene / Poly(vinyl methyl ether) Blends**

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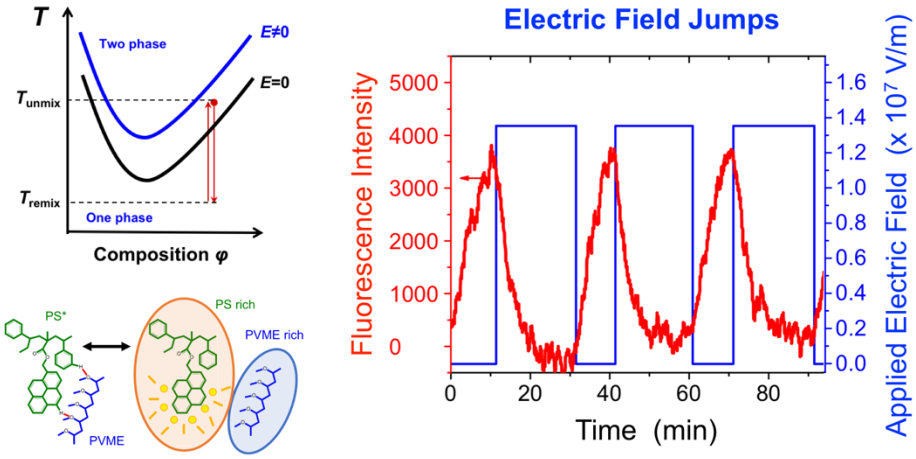
**ABSTRACT:** We demonstrate it is possible to repeatedly jump polystyrene (PS) / poly(vinyl methyl ether) (PVME) blends from the one-phase to two-phase region by simply turning on and off an electric field at a fixed temperature near the phase boundary. This builds on our previous work that established electric fields enhance the miscibility of PS/PVME blends by shifting the phase separation temperature $T_S(E)$ of 50/50 blends up by 13.5 ± 1.4 K when field strengths of $E = 1.7 \times 10^7$ V/m are applied [*J. Chem. Phys.* **2014**, *141*, 134908]. Monitoring the early stages of phase separation and remixing by fluorescence, we measure the remixing timescale $\tau(T)$ with and without electric fields, finding $\tau(T)$ is unchanged by the presence of the field and well fit by a Vogel-Fulcher-Tammann expression. These observations are consistent with a mobility-limited process several degrees from the phase boundary where electric fields have shifted the miscibility transition.

**TOC Graphic:**



Phase manipulation of multicomponent systems by applying electric fields is a promising route to forming idealized morphologies for advanced technologies such as photovoltaics, batteries, and sensors where maximizing conducting pathways is desirable and electrodes are already present in the device.[1-5] One method of obtaining morphologies with small domain sizes is to limit phase separation to the early stages, which requires understanding the dynamics near the phase boundary. Electric fields are known to align domains in the direction of the field, but there is still little understanding of how the presence of electric fields alters the miscibility and shifts the phase boundary.[5-8] Following our recent work demonstrating that electric fields strongly enhance the miscibility of polystyrene (PS) / poly(vinyl methyl ether) (PVME) blends where the phase separation temperature $T_S(E)$ was shifted up by 13.5 ± 1.4 K in a 50/50 blend for $E = 1.7 \times 10^7$ V/m,[9] we explore here the dynamics of phase separation and remixing near the phase boundary with and without electric fields. In a true test that electric fields alter the miscibility, we demonstrate that PS/PVME blends can be jumped in and out of the phase diagram showing alternating phase separation and remixing at a fixed temperature simply by turning the electric field off and on.

The investigation of how electric fields affect phase behavior date back to the 1960s with Debye.[7,10] Most of the work in the field has been done on mixtures of small molecules where the shifts in $T_S(E)$ have been tiny, of order tens of mK.[10-14] Only a few studies have been done on polymers systems[9,14-17] with often larger effects shown, of order a few K for polymer solutions[15,18] and of order ten K for polymer blends.[9,16,17] The magnitudes of the shifts in $T_S(E)$ are larger for polymers because a greater field strength can be applied without causing conduction and Joule heating. The relative shift $\Delta T_S(E)/E^2$ tends to be comparably ~$10^{-14}$–$10^{-13}$ $Km^2/V^2$ for all systems.[9] Experimentally, the presence of electric fields appears to enhance miscibility regardless of whether the mixture forms an upper (UCST) or lower (LCST) critical solution temperature type phase diagram.[14] The exceptions being blends containing the piezoelectric polymer poly(vinylidene fluoride) (PVDF),[16,17] and an early study by Reich and Gordon[19] on PS/PVME that we concluded[9] was likely erroneous. Theoretically, the standard prediction of adding an electrostatic free energy term to the free energy of mixing is too small and generally opposite to the observed shifts.[7,9] However, recent work by Orzechowski et al.[20] has argued the dominant term at high field strengths may be the dielectric contrast between the two components suppressing concentration fluctuations parallel to the field direction[21,22] and

adding an energy penalty for creating interfaces during phase separation, in essence the same mechanism responsible for domain alignment along the field direction.[5,23] This mechanism has similarly been used to drive orientation-dependent order-disorder transitions between microphase-separated morphologies.[3,5] Our 2014 study on PS/PVME blends supported this argument that the dominant effect of how electric fields enhance miscibility is likely this extra energy penalty associated with the formation of interfaces between domains with different dielectric constants, but efforts to quantify the effect did not match our observations.[9] Thus, there is still much left to uncover about how electric fields alter miscibility, especially under strong electric fields where the dielectric response may be nonlinear and field-dependent.[24]

In the present work we are interested in exploring how electric fields can be used to manipulate the phase behavior of a blend. In particular, instead of using the common method of temperature jumps to switch from the one-phase to two-phase region, can we switch between phase separation and remixing by simply turning on and off an applied voltage (electric field)? We use fluorescence to monitor the phase separation and remixing process by attaching pyrene dye to the PS component,[25] a method we developed building on the work by Halary et al.[26-28] who used anthracene dyes. When PS and PVME are intermixed in the one-phase region, the more polar PVME component quenches some of the fluorescence of the aromatic dye, which means a large increase in fluorescence intensity can be observed when phase separation occurs and the dyes become segregated in the less polar PS domains.[26] Fluorescence quenching is a very local process (~few nm)[29] making this method sensitive to the early stages of phase separation and remixing, shown to be comparable to studies my small angle neutron scattering (SANS).[27] Figure 1 illustrates our experimental protocol for the present study. We start with the 50/50 PS/PVME blend in equilibrium in the one-phase region a few degrees below the LCST phase boundary, with zero applied field. A temperature jump is then made into the two-phase region and the blend is allowed to phase separate for a few minutes. The electric field is then applied and a temperature jump is made back into the one-phase region while the fluorescence intensity is monitored to measure the remixing process, observed as an exponential decay of the fluorescence intensity from which we quantity a remixing timescale $\tau$. We have measured the temperature dependence $\tau(T)$ with and without the presence of electric fields, and demonstrate that remixing can occur at a fixed temperature above the zero-field phase separation temperature $T_S(E = 0)$ simply by applying an electric field that according to our previous work[9] shifts the

phase separation temperature $T_S(E)$ up by an amount that transitions the blend back into the one-phase region.

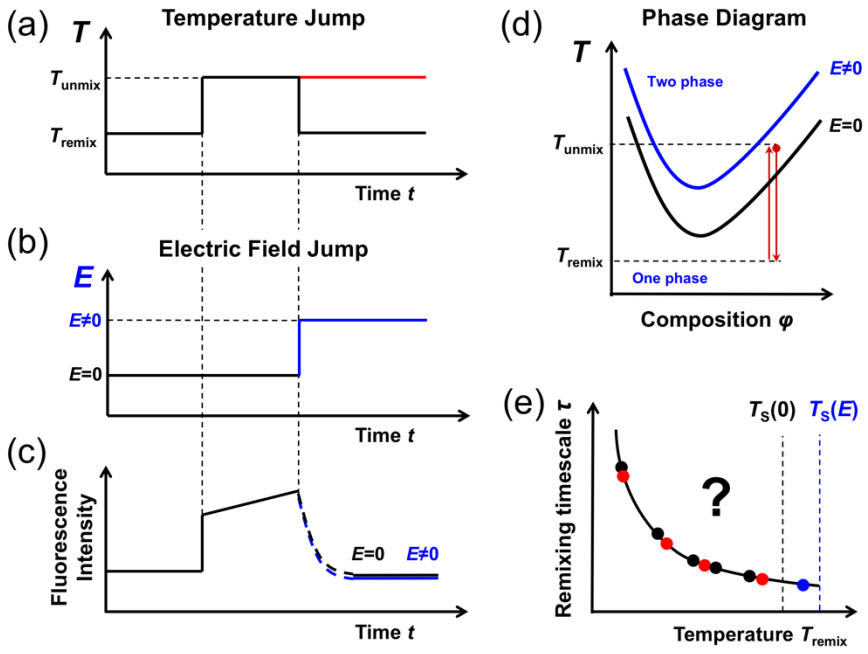



**Figure 1.** Schematic illustrating temperature jump protocol for measuring the remixing timescale $\tau(T)$ with and without the presence of electric fields using fluorescence. Profiles depicting the temperature (a), electric field (b), and fluorescence intensity (c) during the course of an experiment to generate jumps in and out of the phase diagram (d) and measure the remixing timescale (e) are illustrated. (See text for details.)

The sample geometry for this study was described in detail in our previous work.[9] Briefly, PS ($M_w$ = 101.3 kg/mol, $M_w/M_n$ = 1.04) and PVME ($M_w$ = 80 kg/mol, $M_w/M_n$ = 2.5) are mixed with a pyrene-labeled polystyrene (PS*) ($M_w$ = 86.8 kg/mol, $M_w/M_n$ = 1.65, 1.93 mol% dye content) to create (40% PS + 10% PS*)/50% PVME blends with <0.2 mol% total fluorescent dye content. Samples 24 ± 2 μm thick are cast from toluene solutions to make PS/PVME disks (5/8" diam.) sandwiched between transparent, conducting indium tin oxide (ITO) coated quartz substrates (1"×1" in size), where a Kapton sheet with an opening for the sample is used as a spacer to define the electrode separation and help prevent dielectric breakdown. Electrical wires are glued to the corners of the ITO contacts with silver paste and connected to a DC Agilent Technologies N5752A high-voltage power supply. All samples were extensively vacuum

annealed to remove residual solvent and stored under vacuum at room temperature until measured to avoid moisture uptake.[9] Fluorescence measurements (Photon Technology International QuantaMaster fluorimeter) were done by exciting pyrene at 324 nm (4.00 nm bandpass) and monitoring the intensity at 379 nm (4.25 nm bandpass) during a 3 s time window every 15 or 30 s.[9] Sample temperature was controlled to an accuracy of ± 0.3 K using an Instec HCS402 heat stage with liquid-nitrogen cooling capability, where the phase separation temperature $T_S$ was measured on heating at 1 °C/min[9] and temperature jumps were done at ≈30 °C/min. Further information detailing sample preparation, data collection, and background subtraction are provided in Supporting Information.

As outlined in Fig. 1, temperature-jump experiments were started from equilibrium at 94 °C, followed by a jump to 104 °C to initiate phase separation for 5 min; $T_S(E=0) = 99.4 \pm 1.9$ °C, based on an average of five samples. An electric field strength of $E = 1.28 \times 10^7$ V/m was then applied and the temperature jumped back down to 94 °C (or alternatively a different remixing temperature $T_{remix}$), while the fluorescence intensity was collected. We observe that the intensity decays exponentially as the sample remixes, consistent with previous experiments,[26,30,31] and that the data for times where the temperature has stabilized at $T_{remix}$ are well fit to a simple-exponential decay $I(t) = I_0 \exp(-t/\tau)$ allowing us to characterize a remixing timescale $\tau$. The phase separation process unfortunately occurs too rapidly for us to adequately characterize. Figure 2 plots the natural logarithm of normalized fluorescence intensity ($I/I_0$) vs time $t$, where linear fits to the slope of the data are done to determine the remixing time $\tau$ ($t = 0$ has been defined as when the sample temperature stabilized at $T_{remix}$). The blend can then be completely remixed[9] by holding the sample for 2 hours at 94 °C (one-phase region) and the experiment repeated with and without applied electric field. Equivalent data have been collected on nominally identical samples. On average across multiple samples and runs, we measure $\tau = 360 \pm 60$ s for $E = 0$ and $\tau = 280 \pm 60$ s for $E = 1.28 \times 10^7$ V/m at a remixing temperature of 94 °C. Thus to within the experimental variability of 15-20%, we do not observe a change in the measured $\tau$ values with electric field. In our previous work,[9] we demonstrated that a 50/50 PS/PVME blend can be repeatedly phase separated and remixed enabling the reproducible measurement of $T_S$ with and without the presence of electric fields on the same sample and the shift $\Delta T_S(E) = T_S(E) - T_S(0)$ to be measured independent of sample-to-sample variability. We determined the magnitude of the shift relative to electric field strength to be $\Delta T_S(E)/E^2 = (4.8 \pm$

0.4) × $10^{-14}$ $Km^2/V^2$, such that a field strength of 1.28 × $10^7$ V/m corresponds to shift $\Delta T_S(E)$ = 7.9 ± 0.7 K.[9]

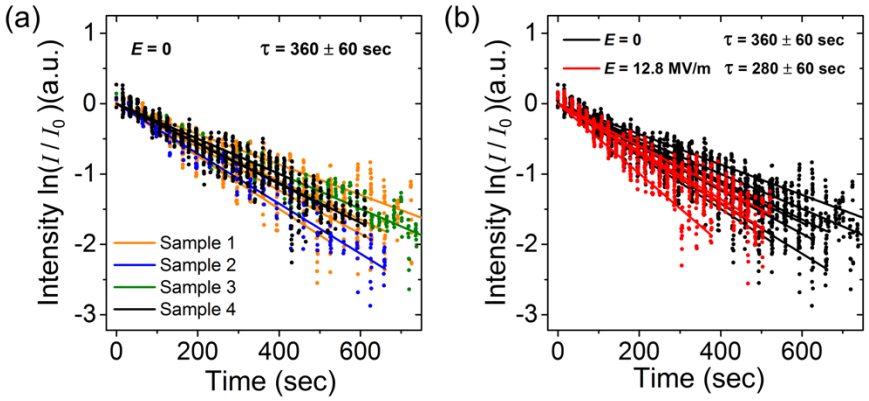



**Figure 2.** Plot of the natural logarithm of normalized fluorescence intensity ($I/I_0$) vs time $t$, where the lines represent linear fits to $\ln(I/I_0) = -t/\tau$ to determine the remixing time $\tau$. (a) Demonstrates reproducibility across eight different runs on four different samples (at zero applied field), and (b) compares data collected with (red) and without (black) the presence of an electric field of $E$ = 1.28 × $10^7$ V/m.

Figure 3 graphs the temperature dependence of the remixing timescale $\tau(T)$ measured across multiple samples comparing data collected at zero field with that under an applied field of $E$ = 1.28 × $10^7$ V/m and $E$ = 1.30 × $10^7$ V/m. The data are plotted on semilogarithmic axes relative to the zero field $T_S(E=0)$ values measured for each sample demonstrating that the measured $\tau(T)$ values are the same to within the experimental variability with or without applied E-field despite $T_S(E)$ being shifted up by ≈8 K. This observation suggests that remixing is limited by the mobility of the chains, consistent with expectations from previous studies on the dynamics of phase separation and remixing where the proximity of the phase boundary leading to deviations in $\tau(T)$ from a primarily mobility dominated process is only felt very close to the boundary.[31,32] It is possible that additional electric field effects may be felt in very close proximity to the spinodal boundary. In the present work, we find the $\tau(T)$ data are well fit by a Vogel-Fulcher-Tammann (VFT) temperature dependence, $\log(\tau) \sim \frac{B}{T-T_\infty}$, as would be expected for a viscosity-limited process. We find our fit parameters, $B$ = 5.8 ± 1.7 and $T_\infty$ = 353 ± 5 K, to be notably different than previous literature reports for the viscosity and diffusion of PS/PVME

blends.[33,34] However, these reports are for mixed blends at temperatures well above the blend's glass transition. Our $\tau(T)$ values agree well with interdiffusion studies by Jabbari and Peppas[35-37] who investigated the interdiffusion of PS and PVME at temperatures above and below the glass transition temperature of PS (100 °C). For PS ($M_w$ = 105 kg/mol) and PVME ($M_w$ = 99 kg/mol), comparable molecular weights to our own, they found $D = 1.1 \times 10^{-12}$ cm$^2$/s at 105 °C and $D = 4.2 \times 10^{-14}$ cm$^2$/s at 85 °C, observing a significant non-Fickian (case-II) diffusion contribution, especially at lower temperatures.[35] Treating these values as experimentally measured effective mutual diffusion coefficients, we can calculate roughly how long a blend would take to remix at 85 °C after 5 min of phase separation at 105 °C, where the growth and decay for any particular wavevector $q$ would be expected to go as $\sim e^{\pm Dq^2 t}$ during the early stages of phase separation and remixing based on Cahn-Hilliard treatment as a first-order estimate.[31,38] Such an estimate would predict $\tau \approx 1600$ s at 85 °C in good agreement with our measured values of $\tau$(85 °C) = 1500 ± 250 s. It is worth noting that Debye et al. measured the relaxation times of concentration fluctuations in a mixture of small molecules after short pulses of electric fields to be independent of field strength.[39] Figure 3 also includes remixing $\tau(T)$ data collected at a temperature 4 K above the zero field $T_S(E=0)$ under a field strength of $E = 1.30 \times 10^7$ V/m that shifts $T_S(E)$ up by 8.1 ± 0.7 K,[9] confirming that the application of electric fields can be used to shift the phase boundary transitioning the blend from the two-phase to one-phase region.

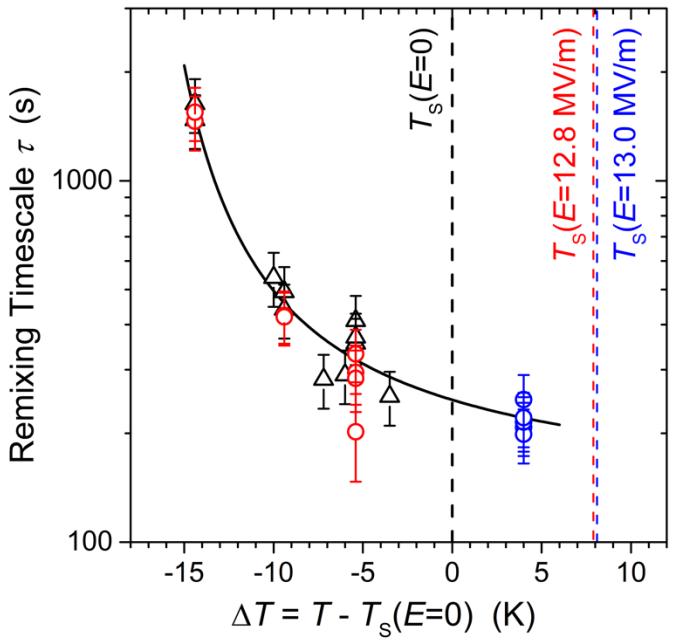


**Figure 3.** Graph of the measure remixing timescale $\tau(T)$ on semi-logarithmic axes as a function of temperature relative to the zero field $T_S(E=0) = 99.4$ °C. Black triangles collected under zero field, red circles under an applied electric field of $E = 1.28 \times 10^7$ V/m where $T_S(E) = T_S(E=0) + 7.9$ K, and blue circles under $E = 1.30 \times 10^7$ V/m where $T_S(E) = T_S(E=0) + 8.1$ K. Black curve is a VFT fit to all the data.

Figure 4 plots the measured fluorescence intensity and applied E-field as a function of time while the blend is held at a fixed temperature of $T_S(E=0) + 4$ K and the electric field is switched on and off to repeatedly transition the blend from the one-phase to two-phase region alternating between remixing and phase separation. For the first eight E-field jumps, an applied field of $E = 1.30 \times 10^7$ V/m is used to increase the phase separation temperature $T_S(E)$ up by 8.1 ± 0.7 K,[9] enough to transition the blend into the one-phase region ~4 K below the phase boundary when the electric field is on, while the blend returns to the two-phase region when the E-field is turned off. The remixing timescale $\tau(T)$ is determined from the first four E-field jumps and plotted in Fig. 3 showing good agreement with the $\tau(T)$ data collected at $E = 1.28 \times 10^7$ V/m below $T_S(E=0)$. The last three E-field jumps apply a field strength of $E = 1.60 \times 10^7$ V/m corresponding to a $T_S(E)$ increase of 12.3 ± 1.0 K. These observations confirm our previous work[9] that electric fields shift the phase boundary of PS/PVME blends towards enhanced miscibility. Such shifts in blend miscibility are reminiscent of shear flow induced mixing and phase separation that have been previously investigated in PS/PVME blends.[40-44] The advantage of E-field jumps over other methods such as temperature for switching a blend from the one-phase to two-phase region is that the electric field (applied voltage) can be turned on and off instantaneously, and the data in Fig. 4 demonstrate that the blend responds to this change immediately. It is worth noting that although these electric field strengths are large, with active layers in photovoltaic devices getting smaller ~100 nm, only 1 V would need to be applied to such a system to get comparable E-field strengths ~$10^7$ V/m. The results shown in Fig. 4 demonstrate that electric fields can be used to manipulate the phase behavior of PS/PVME blends enabling remixing to occur at temperatures that correspond to the two-phase region under zero field conditions.

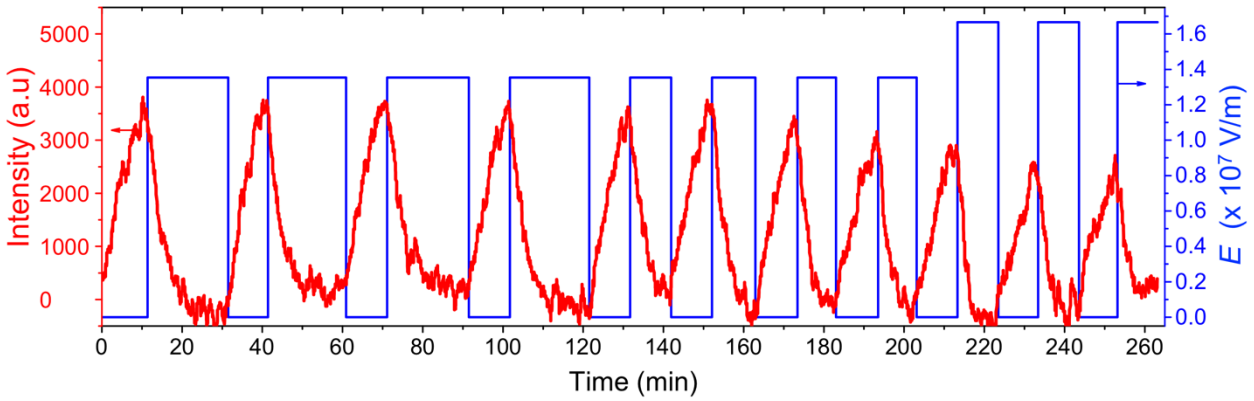



**Figure 4.** Plot of measured fluorescence intensity (red curve) and applied electric field $E$ (blue curve) as a function of time while sample is held at a fixed temperature of $T_S(E=0) + 4$ K. For the first eight E-field jumps, an applied field of $E = 1.30 \times 10^7$ V/m is turned on and off increasing $T_S(E)$ up by 8.1 K, repeatedly switching the blend from the one-phase to two-phase region, while for the last three E-field jumps, a field of $E = 1.60 \times 10^7$ V/m is used increasing $T_S(E)$ up by 12.3 K.

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**Supporting Information Available**: The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsmacrolett.XXXXXXX. Sample preparation details and experimental methods, along with details of the analysis to determine the remixing time.

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Supporting Information for:

# Jumping In and Out of the Phase Diagram Using Electric Fields: Timescale for Remixing of Polystyrene / Poly(vinyl methyl ether) Blends

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## Sample Preparation and Measurement Details

Polystyrene (PS) with molecular weight $M_w$ = 101.3 kg/mol and $M_w/M_n$ = 1.04 (Scientific Polymer Products) was used for the matrix (neat) PS. Pyrene-labeled polystyrene (designated PS*) with $M_w$ = 86.8 kg/mol and $M_w/M_n$ = 1.65, containing a label content of 1.93 mol%, was synthesized by free radical polymerization from styrene in the presence of trace levels of 1-pyrenylmethyl methacrylate monomer (purchased from Polysciences), as described in our previous work.[1,2] Fluorophore label content was measured using UV-visible absorbance spectroscopy in high-performance liquid chromatography (HPLC) grade tetrahydrofuran (THF). Poly(vinyl methyl ether) (PVME) was purchased from Scientific Polymer Products and washed prior to use by dissolving in toluene and reprecipitating into heptane, repeated nine times. The resulting PVME used had a molecular weight of $M_w$ = 80 kg/mol and $M_w/M_n$ = 2.5, as determined by gel permeation chromatography (GPC) measured with THF as eluent relative to PS standards, as reported in our previous work.[1] PVME molecular weight values were determined using universal calibration with Mark-Houwink parameters $a$ = 0.739 and $k$ = 13.5 × $10^{-3}$ mL/g.[3]

Polymer blend compositions of 50/50 PS/PVME were prepared by dissolving 40% PS, 10% PS*, and 50% PVME in toluene to make solutions containing 18 wt% total polymer content. These solutions were then cast on a glass dish with flat bottom and dried in a fume hood at room temperature for 24 h, followed by annealing in a vacuum oven at 80 °C for at least 24 h to remove residual solvent. Disks of the polymer blend where then cut and used to fill a 5/8" diameter hole within a 24 μm thick Kapton sheet used as a spacer to define the electrode spacing between two transparent, conducting indium tin oxide (ITO) coated quartz substrates (1"×1" in

size). Electrical wires were attached with silver paste to the corners of the ITO contacts to make connection to a DC Agilent Technologies N5752A high-voltage power supply. The assembled samples, containing a final dye content <0.2 mol%, were placed under vacuum at 80 °C and pressed together with a 2.2 kg weight for 3-5 days, until the polymer blend filled in the form of the Kapton spacer. The thickness of each sample was measured to within ± 2 μm using an optical microscope (Leica DMIRB inverted microscope) by focusing on the top and bottom of the optically transparent samples and recording the displacement of the micrometer scale. Electric field strengths up to $1.8 \times 10^7$ V/m where achieved by applying up to 440 $V_{DC}$. A schematic of our sample geometry was published in our previous work.[1]

Steady-state fluorescence measurements were carried out using a Photon Technology International QuantaMaster fluorimeter with the sample mounted in an Instec HCS402 heat stage with liquid-nitrogen cooling capability, where the sample temperature was controlled to within ± 0.3 K. Following our previous work,[2] measurements of the phase separation temperature $T_S$ were done by heating the sample at a rate of 1 °C/min and monitoring the fluorescence intensity for a large increase in intensity signifying a strong reduction in fluorescence quenching occurring when the pyrene dye covalently bonded to the PS component phase separated from the more polar PVME and segregated in predominantly non-polar PS domains. The pyrene dye was excited at a wavelength of 324 nm with 4.00 nm bandpass and the resulting fluorescence monitored for 3 s every 30 s at an emission wavelength of 379 nm with 4.25 nm bandpass. Temperature jumps up and down from the one-phase to two-phase regions were done at a rate of ≈30 °C/min while the fluorescence was monitored every 15 s. In our previous work, we demonstrated that PS/PVME blends can be phase separated and subsequently remixed over and over again, while repeatedly measuring the same phase separation temperature $T_S$ value.[1] Typically remixing is carried out by holding the blend at 94 °C for 2 hours. Using this method, we determined an average reproducible $T_S(E=0) = 99.4 \pm 1.9$ °C across five different samples. Slight differences in $T_S(E=0)$ from sample-to-sample variability are accommodated by plotting data from each sample relative to its measured $T_S(E=0)$. After finding reproducible $T_S(E=0)$ values for each sample, measurements were continued with temperature and electric field jumps.

Figure S1 demonstrates the data collection procedure to measure the remixing timescale $\tau$ from a temperature jump. For the sample shown, the blend starts in an equilibrium, well-mixed state within the one-phase region at an initial temperature of 94 °C, 5.8 °C below its $T_S(E=0)$ =

99.8 °C. A temperature jump is then made to 104 °C, 4.2 °C above the $T_S(E=0)$, into the two-phase region and allowed to phase separate for 5 min. After 5 min, the temperature is jumped back down to the desired remixing temperature, in this case 94 °C. The black curve shown in Fig. S1a corresponds to the temperature profile assigned to the temperature controller (MK1000 High Precision Temperature Controller), while the red curve shows the measured temperature of the sample chamber recorded using the associated WinTemp software. Fig. S1b graphs the measured response of the fluorescence intensity $I(t)$ measured as a function of time through the temperature jump profile. We frequently find there is a small linear background drift of the intensity with time that we subtract off by making a linear fit to the data before and after the temperature jump, shown as a blue fit line in Fig. S1b. This gives us the intensity profile $I^*(t)$ shown in Fig. S1c, where the small linear background has been subtracted, allowing us to focus on the fluorescence intensity changes associated with the temperature jump profile. During the up jump in temperature, we notice a brief downward spike in fluorescence intensity right after the temperature is raised from 94 °C to 104 °C (from A to B) associated with fluorescence intensity generally decreasing with increasing temperature, prior to the onset of phase separation marked by the large increase in fluorescence intensity.[2] Phase separation is allowed to proceed for 5 min at 104 °C before performing a second temperature jump back down to 94 °C. At long times the fluorescence intensity $I^*(t)$ in Fig. S1c is observed to return the same initial baseline value at 94 °C. We have quenched samples that have been phase separated for 5 min at 104 °C down to room temperature for inspection using optical microscopy finding uniformly sized domains typically smaller than ~1 μm, consistent with the reports by Halary et al. that fluorescence is sensitive to the early stages of phase separation.[4] The fluorescence method we employ using pyrene is equivalent to that developed by Halary et al. using anthracene.[5] Our previous work[2] established that pyrene gives equivalent $T_S$ values to that measured by anthracene, but has the added benefit of having a higher quantum yield. Previous effort by Halary et al. also demonstrated that this fluorescence method provides equivalent phase separation temperature values to that measured by small angle neutron scattering (SANS),[4] confirming that fluorescence is sensitive to the early stages of phase separation.

Following the temperature jump back down to 94 °C, the fluorescence intensity in Fig. S1c is observed to decay exponentially, consistent with previous reports demonstrating remixing in PS/PVME blends.[5-7] We fit this data to a single exponential decay by doing a linear fit to

$\ln(I^*) = -t/\tau + \ln(I_0)$ as depicted in Fig. 1Sd, where $t = 0$ is defined as when the measured sample temperature has stabilized at the remixing temperature (point D). Fits are extended until the data fall to within the noise level of the intensity data. This experiment was repeated on both nominally identical samples and on the same sample where complete remixing of the blend is done by holding the sample for 2 h at 94 °C, as demonstrated in our previous work.[1] Figure S2 graphs the fluorescence intensity decay $\ln(I^*)$ vs time for samples remixed at 90 °C and 85 °C, without (a) and with (b) an applied electric field of $E = 1.28 \times 10^7$ V/m. The measured remixing timescales for these samples are (a) $\tau$(90 °C) = 440 s and $\tau$(85 °C) = 1600 s under zero field, and (b) $\tau$(90 °C) = 420 s and $\tau$(85 °C) = 1550 s under an electric field of $E = 1.28 \times 10^7$ V/m, representing some of the $\tau(T)$ data plotted in Figure 3 of the main text.

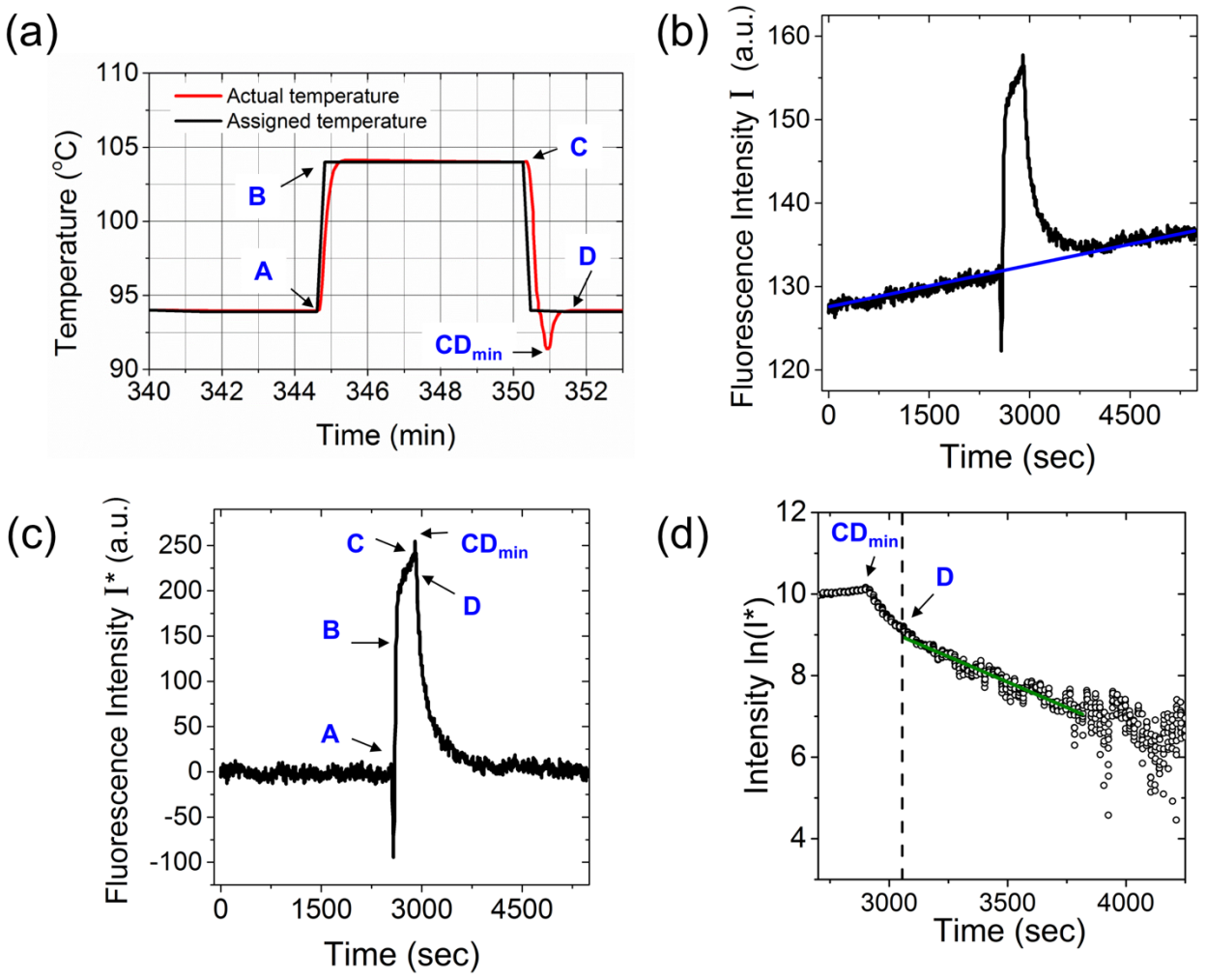


**Figure S1.** Fluorescence intensity data collection procedure during a temperature jump. (a) Temperature jump profile applied (black) and measured (red) to the sample starting in

equilibrium within the one-phase region at 94 °C, jumping up to 104 °C to allow the blend to phase separate for 5 min, followed by a temperature jump back down to 94 °C. (b) Raw measured fluorescence intensity during the temperature jump, along with a straight line fit (blue line) used to subtract the small linear background drift. (c) Time dependence of the fluorescence intensity profile $I^*(t)$, measured intensity with small linear background drift subtracted, demonstrating the sharp increase in fluorescence intensity during phase separation followed by the exponential decay during remixing. (d) Semi-log plot used to determine the remixing timescale $\tau$.

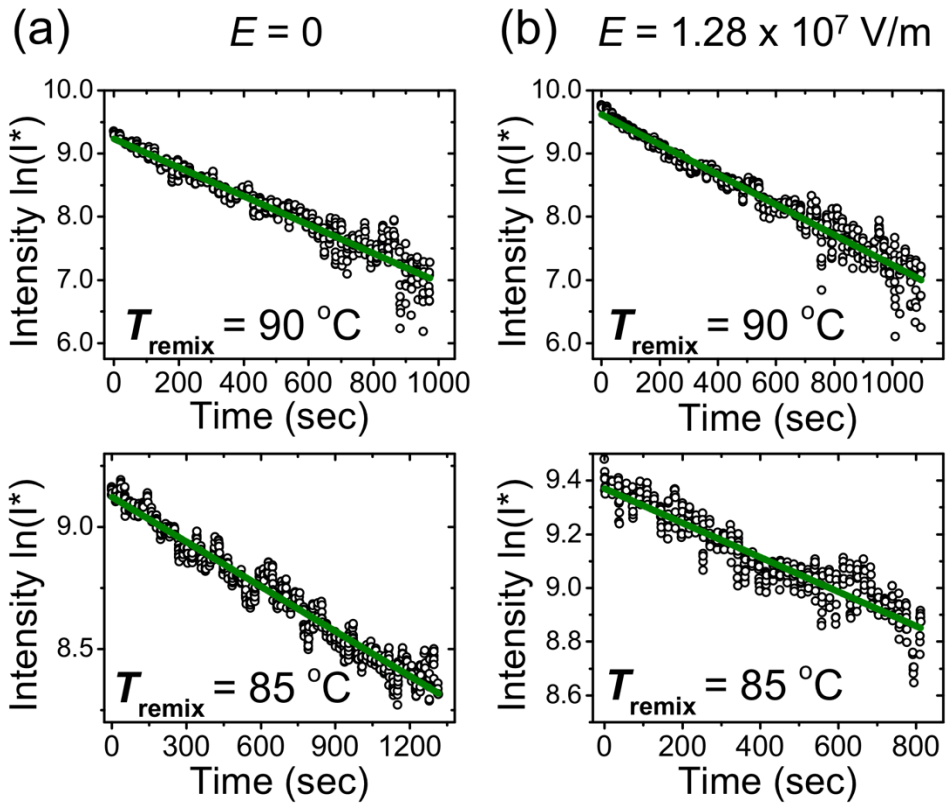


**Figure S2.** Time-dependence of the fluorescence intensity decay plotted as ln($I^*$) vs. time $t$ for 50/50 PS/PVME blends remixed at 90 °C and 85 °C without (a) and with (b) an applied electric field of $E = 1.28 \times 10^7$ V/m. Green lines show fits to a single exponential decay: $\ln(I^*) = -t/\tau + \ln(I_0)$.

**References**


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