

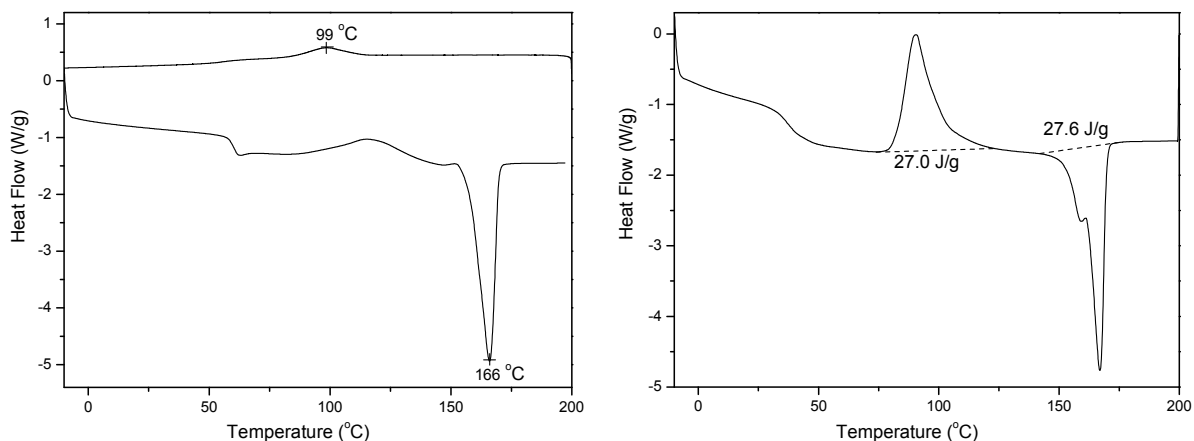
## Supplementary Information

### Spectroscopic Study of Water Diffusion in Poly(lactic acid) Film

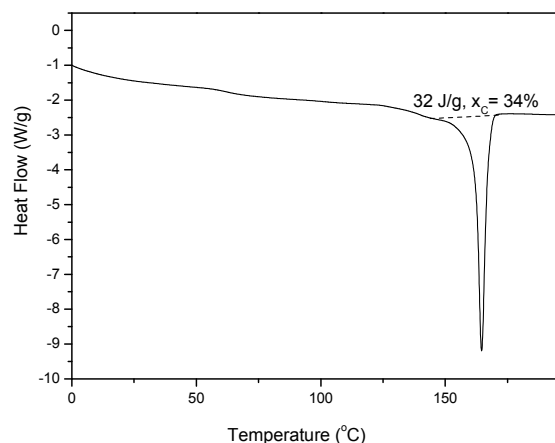
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**DSC.** A TA Instruments Q-100 calorimeter was employed to study the melting and crystallization temperatures of PLA. The film cast from solution was heated, cooled and then heated again at 10 °C/min in the range of -10 to 200 °C. Fig. S1 shows the cooling and heating traces. Then an isothermally crystallized PLA film was heated at 10 °C/min in the range of -10 to 200 °C for further structural analysis as shown in Fig. S2. In Fig. S1 (a), the DSC cooling and second heating runs exhibit the crystallization and melting temperatures of PLA. From the first heating trace, it can be found the enthalpy of cold crystallization is equal to that of melting, which indicates that the film cast from solution is amorphous. In Fig. S2, according to the melting enthalpy of PLA (93 J/g), it can be calculated that the degree of crystallinity of the PLA is ~34%.

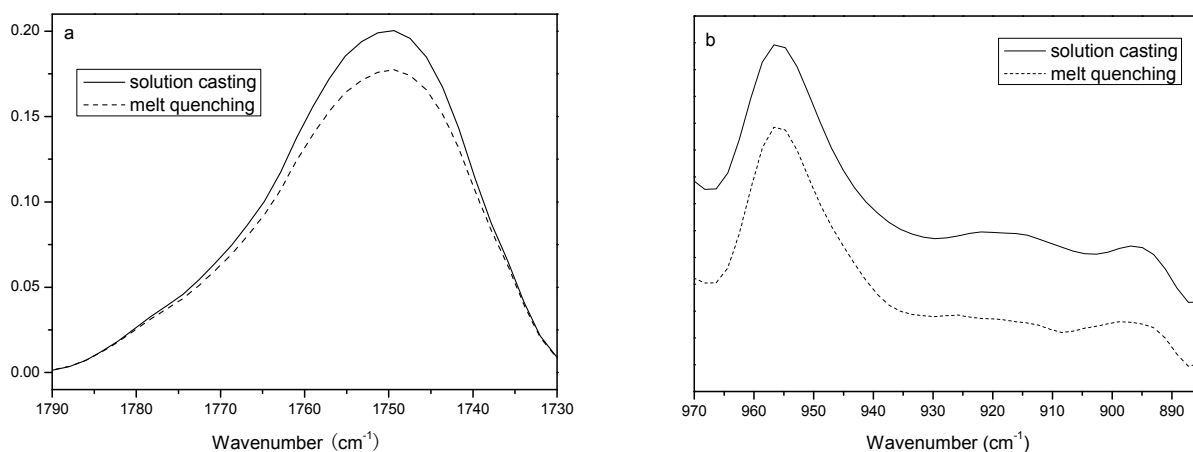


**Fig. S1.** DSC cooling and second heating traces (a), and first heating trace (b) of the PLA film from solution casting.

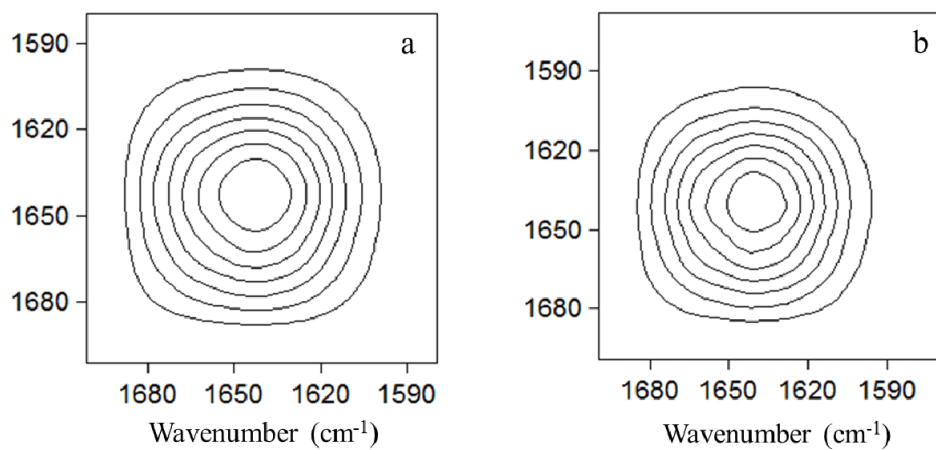


**Fig. S2.** DSC second heating trace of PLA after isothermal crystallization.

**FTIR.** FTIR was used to further verify the structure of the solution-cast PLA film. We compare the carbonyl stretching band (conformation sensitive) in the range of  $1790\text{-}1730 \text{ cm}^{-1}$  of the cast film with that of an amorphous film prepared by quenching from the melts. As shown in Fig. S3 (a), the band shapes for these two samples are identical, indicating that the cast film has the same conformation distribution as the amorphous film. In Fig. S4b, neither film exhibits the crystalline peak at  $921 \text{ cm}^{-1}$ , i.e. the PLA in the cast film is amorphous. Thus, we confirm that the solution-cast PLA film is amorphous.



**Fig. S3.** C=O stretching vibration (a) and PLA crystalline peak (b) of PLA films from solution casting and melt quenching.



**Fig. S4.** Synchronous 2D FTIR correlation spectra of water diffusion into (a) amorphous and (b) crystalline PLA films in the range of 1700-1580 cm<sup>-1</sup>.