Electronic Supplementary Material (ESI) for RSC Advances.
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1. Morphology

The morphology of PVDF/1Gra and PVDF/1IL/1Gra was investigated by field emission scanning electron microscopy (FESEM, Hitachi-SU8020). The samples were brittle fractured in liquid nitrogen, and all the fracture surfaces were sputter-coated with gold layer before examination. As shown in Fig. S1, no pores nor voids can be observed in our composites samples, indicating that the solvent used to prepare the composites and air were fully removed from the samples. The white wrinkled topography in Fig. S1 represents the ends of Gra stretched out of the PVDF matrix. We can see that the PVDF/1Gra shows large Gra agglomerations indicating a poor dispersion of Gra in PVDF, and that the distance between Gra decreases as IL are added.[1] These observations suggest that IL can improve the compatibility between Gra and PVDF matrix, resulting in a uniform dispersion of Gra in PVDF matrix because the lubrication of IL becomes apparent when this rigid particles wears a piece of “oil film”.

2. Isothermal Crystallization Kinetics

Fig. S2 records the process of isothermal crystallization of samples at the different predetermined crystallization temperature (T_c). With T_c increasing in the test of sample the crystallization exothermic time becomes longer and the peak of crystallization exothermic becomes gentler. Compared with neat PVDF, the exothermic time becomes shorter and the peak becomes sharper at T_c of 150 °C. It indicates the additive can make the process of isothermal crystallization easy, especially the samples when with 1 *wt* % Gra. Fig. S3 displays the relative crystallinity versus time during isothermal crystallization of samples at different T_c. The relative crystallinity (X_t), a function of crystallization temperature T, can be formulated as:

$$X_t = \int_{T_0}^{T} \left(\frac{dH_c}{dT}\right) dT \Big/ \int_{T_0}^{T_\infty} \left(\frac{dH_c}{dT}\right) dT \qquad (1)$$

3. Non-isothermal Crystallization Kinetics.

Compared with isothermal crystallization behavior, the non-isothermal crystallization behavior gets more attention because of its practicability. In study of non-isothermal crystallization kinetics, data for the crystallization exothermic as a function of temperature were obtained at different cooling temperature rates for samples is manifested in Fig. S4. First, all samples confirm to classical theory of crystallization, there is enough time for polymer chain to move and fold at higher temperature when the cooling rate is slow, but it is difficult for them to crystal at high cooling rate. Then, the slope of the dotted line of PVDF/IL, PVDF/Gra and PVDF/IL/Gra is smaller than it of neat PVDF, this indicates the additive hinder the crystallization, so they need a high degree of undercooling.[2] The X_t, a function of crystallization temperature T, can be formulated by (1).

Fig. S5 shows the X_t at different crystallization temperature in the process of non-isothermal crystallization for samples. The crystallization time can be calculated by the eq. (2):

$$t = \left| T_0 - T \right| / \Phi \qquad (2)$$

Where T is the temperature at time t, T_0 is temperature at initial crystallization time and Φ is the cooling rate. Fig. S6 expresses the relationship between of X_t and different crystallization time. From these curves, the half-time of crystallization ($t_{1/2}$) can be picked out easily.

References

1 G. X. Chen, S. C. Zhang, Z. Zhou and Q. F. Li, *Polym. Compos.*, 2015, **36**, 94-101.

2 H. M. Chen, W. B. Zhang, X. C. Du, J. H. Yang, N. Zhang, T. Huang and Y. Wang, *Thermochim. Acta*, 2013, **566**, 57-70.

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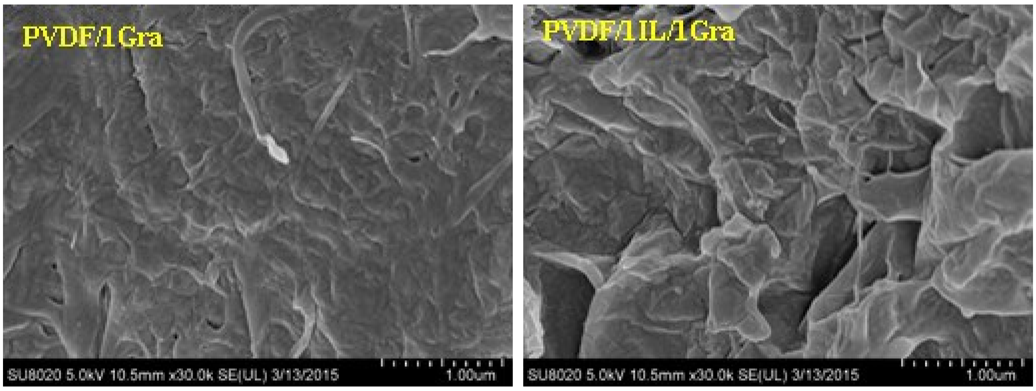
Fig. S3 Relative crystallinity versus time during isothermal crystallization of samples at different crystallization temperatures by DSC.

Fig. S4 Heat flow versus temperature during non-isothermal crystallization of samples at different cooling rates by DSC.

Fig. S5 Relative crystallinity (X_t) at different crystallization temperatures in the process of non-isothermal crystallization for samples.

Fig. S6 Relative crystallinity (X_t) at different crystallization time in the process of non-isothermal crystallization for samples.



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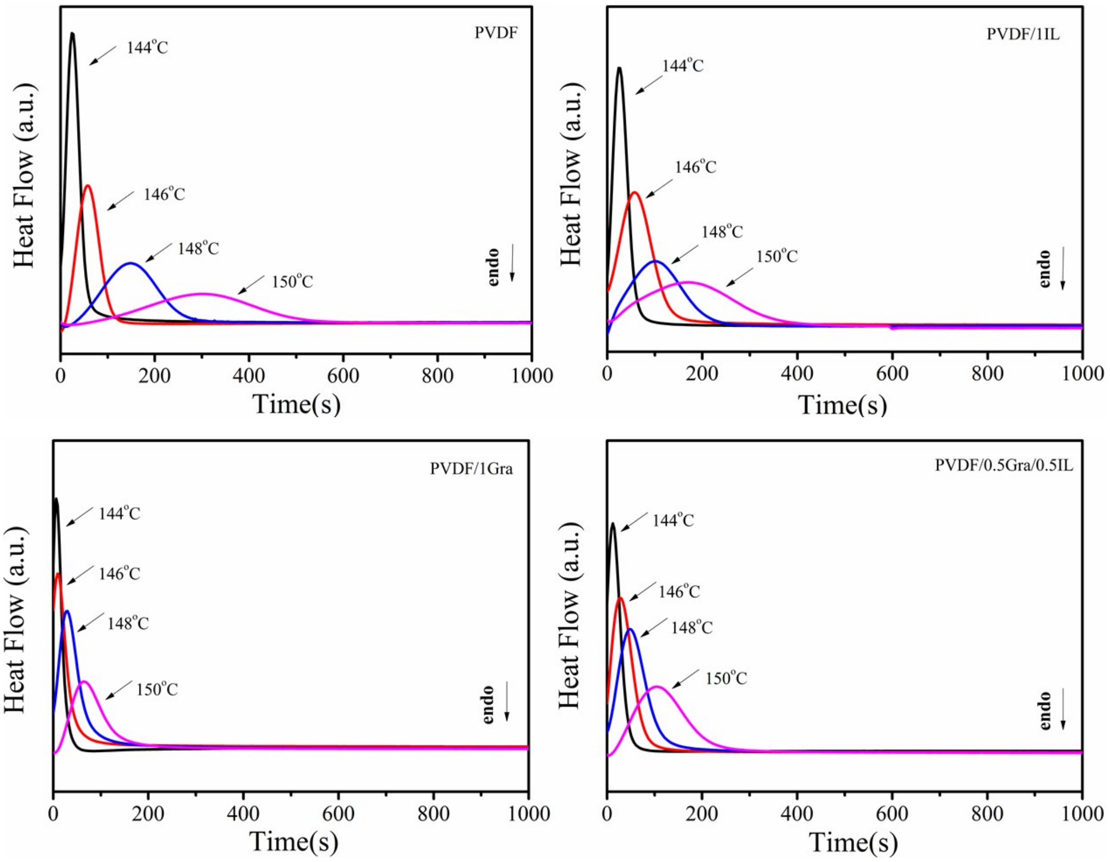

Fig. S2 Heat flow versus time during isothermal crystallization of samples at different crystallization temperatures by DSC.

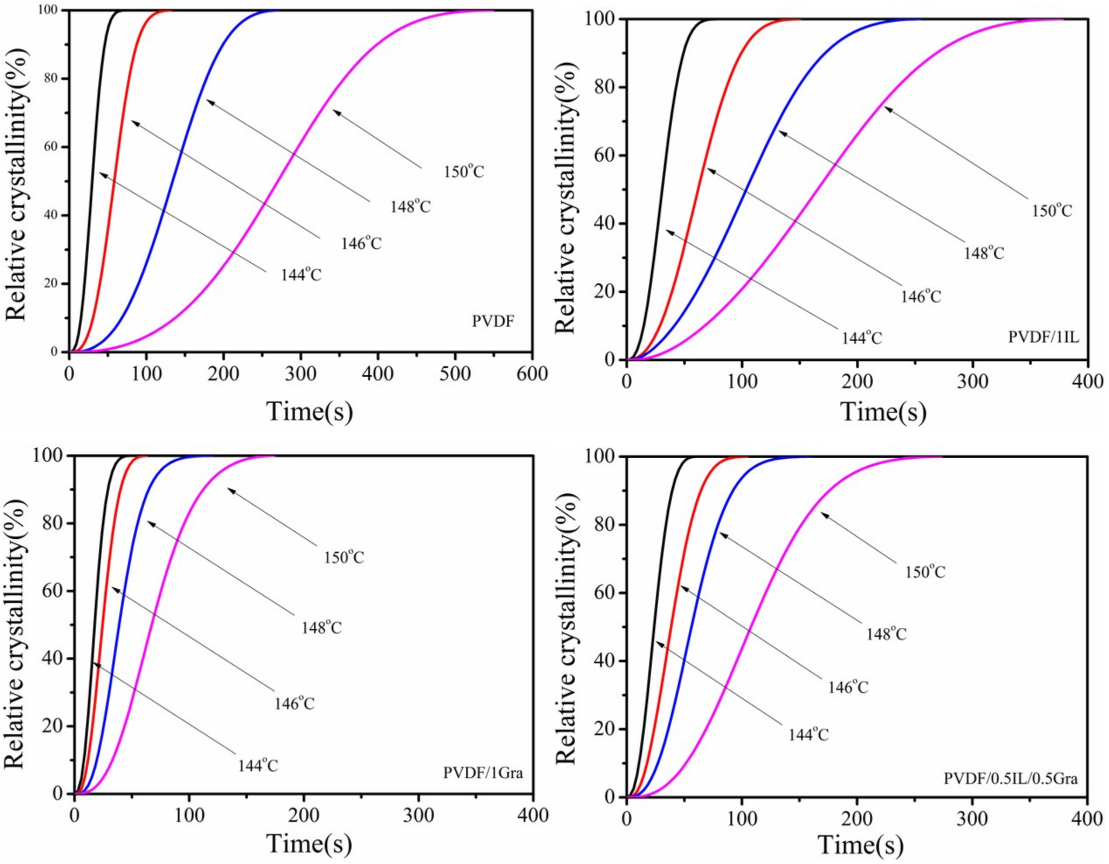

Fig. S3 Relative crystallinity versus time during isothermal crystallization of samples at different crystallization temperatures by DSC.

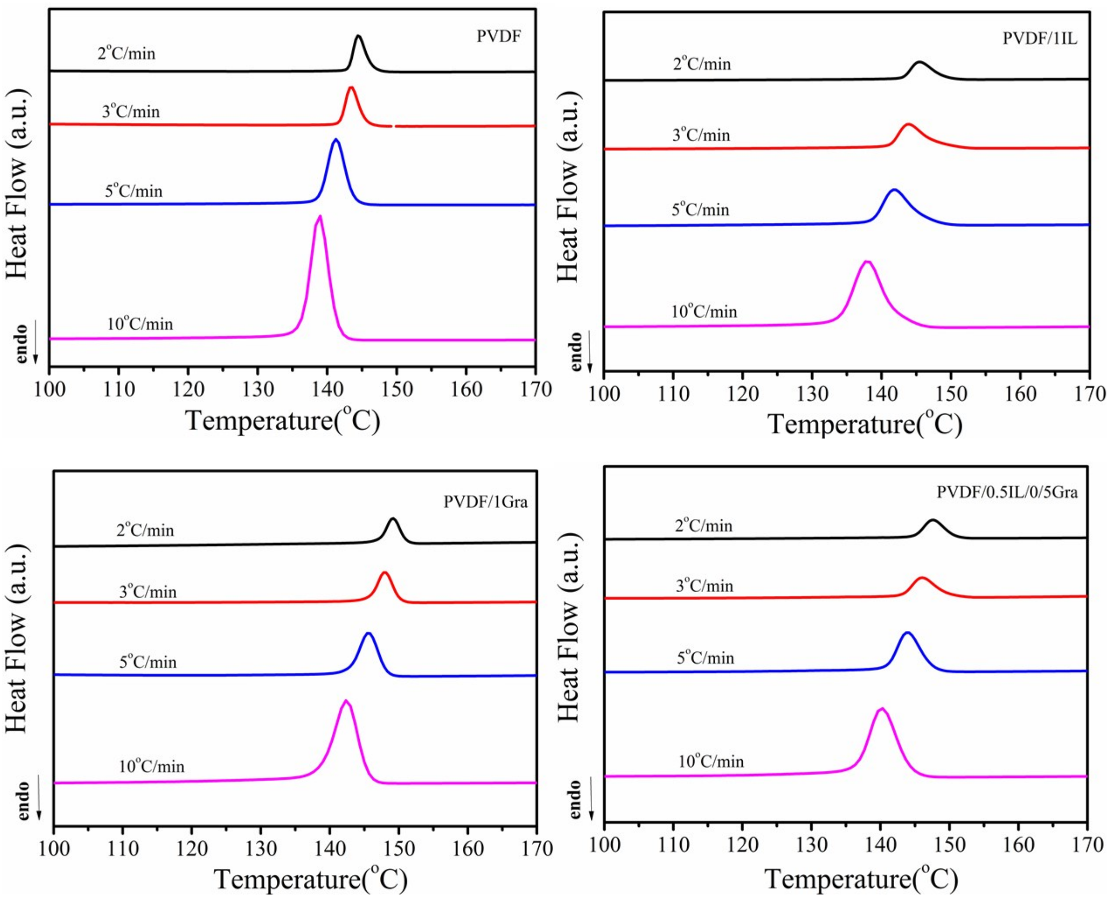

Fig. S4 Heat flow versus temperature during non-isothermal crystallization of samples at different cooling rates by DSC.

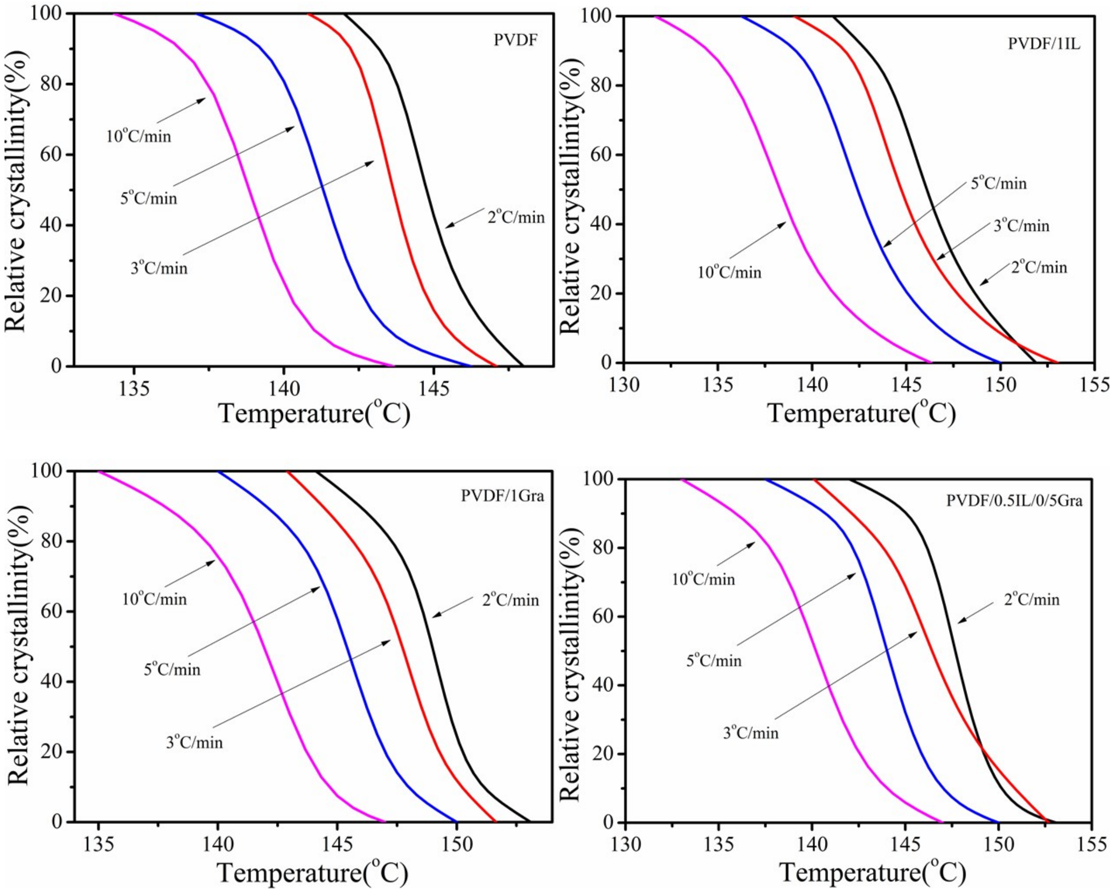

Fig. S5 Relative crystallinity (X_t) at different crystallization temperatures in the process of non-isothermal crystallization for samples.

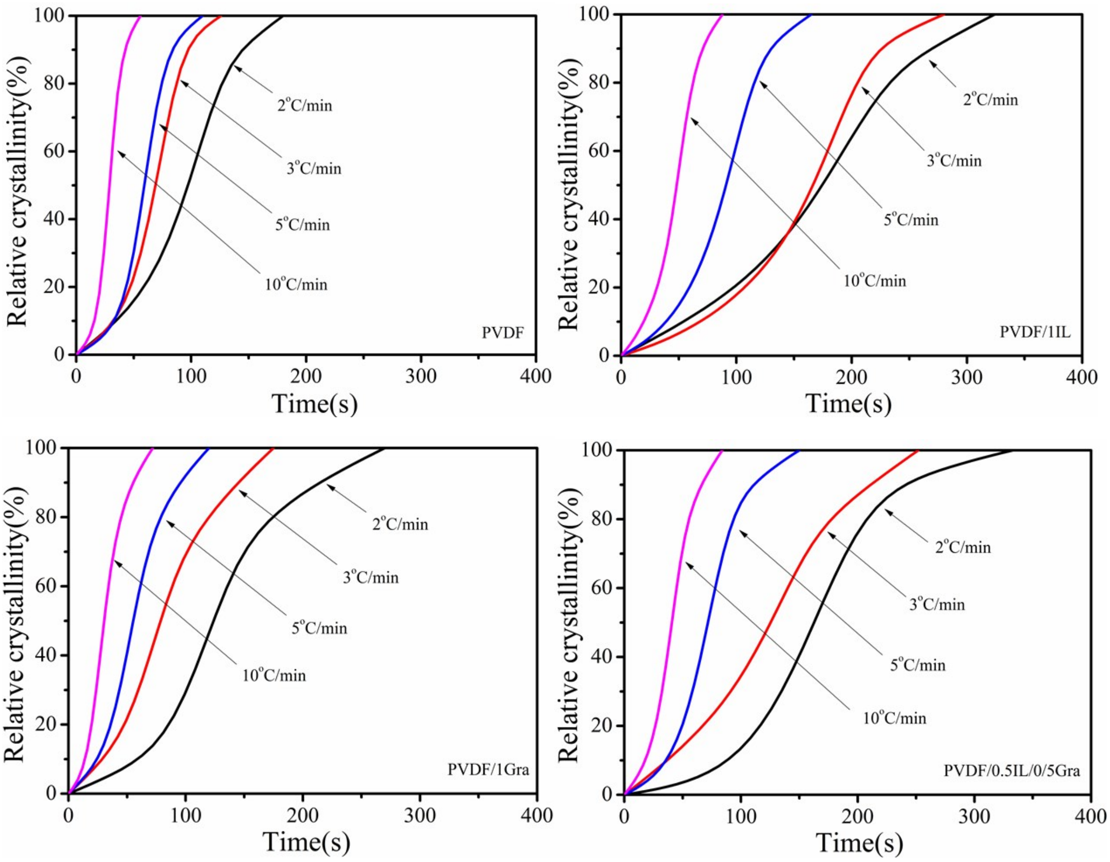

Fig. S6 Relative crystallinity (X_t) at different crystallization time in the process of non-isothermal crystallization for samples.