Electronic Supplementary Information

Strain-Induced Crystal Growth and Molecular Orientation of Poly(isobutylene-isoprene) Rubber at Low Temperatures

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1. Differential Scanning Calorimetry (DSC) Results

The thermal properties of samples were investigated in a nitrogen atmosphere in the temperature range from 25 to -100 °C with the cooling and heating rates of 10 °C / min using a TA Q2000 series DSC instrument. As shown in Fig. S1(a), the value of T_g is -66 °C. Concerns related to the temperature-induced crystallization is removed through annealing IIR rubbers at different temperatures as shown in Fig. S1(b). No exothermic peak is observed during heating, which indicates IIR does not crystallize at different temperatures under quiescent conditions.

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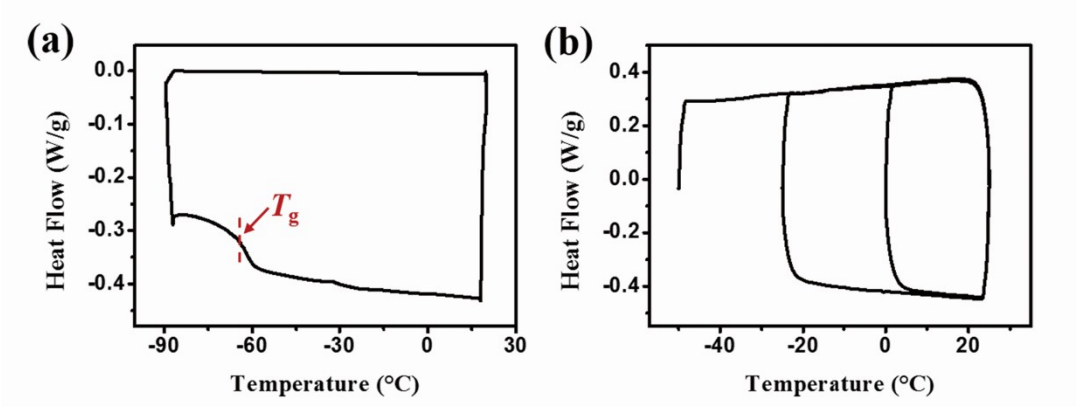

Fig. S1 DSC curves of IIR sample acquired at a heating rate of 10 °C/min. (a) The measurement of T_g; (b) Isothermal measurement at different temperatures.

2. Low temperature extensional Rheometer

This low-temperature extensional rheometer drew lessons from the commercial Sentmanat extensional rheometer (SER) with two drums rotating oppositely developed by Sentmanat. And it will not be bounded by strain scale in that only the sample between two drums could be stretched. Thus, all the strain in the manuscript represents the Hencky strain. It has less restriction on the sample in the direction perpendicular to the tensile direction. Thus, the value of Hencky strain may bigger than the tensile ratio λ after conversion.

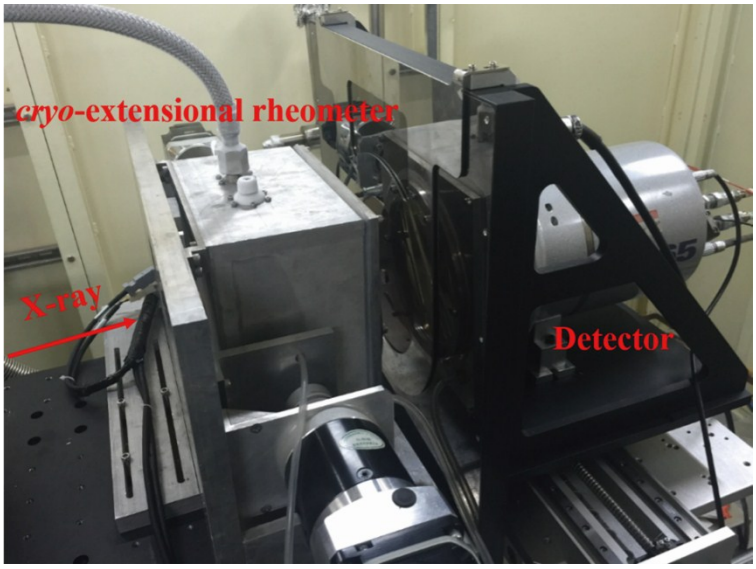

Fig. S2 Low-temperature extensional rheometer mounted at BL16B, SSRF.

Table S1 The original data of ε_{onset} at various temperatures.

Temperature / °C	ε_{onset}
0	2.40
-10	1.85
-20	1.65
-30	1.55
-40	1.70
-50	2.00
-60	2.55

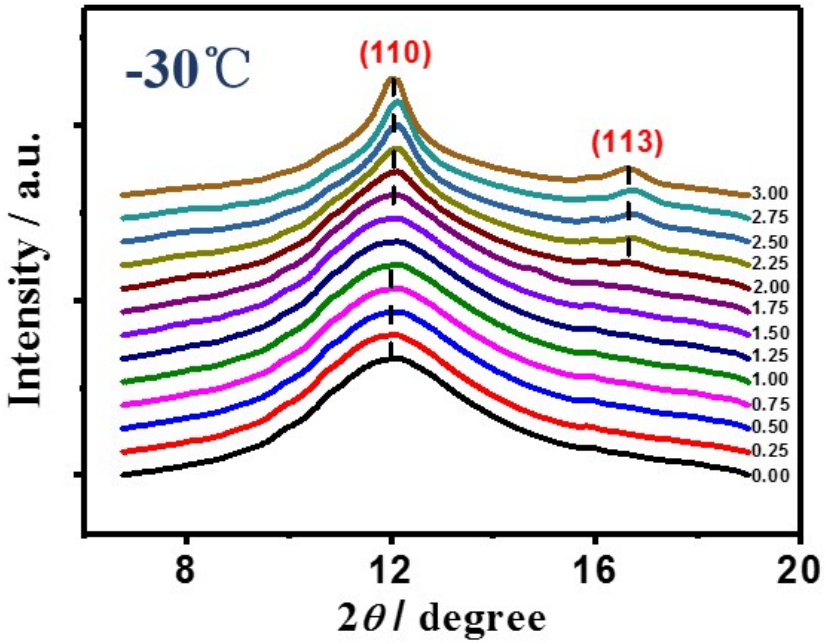

Fig. S3 Sequential evolution of 2D SR-WAXD patterns during uniaxial stretching at different temperatures. Magnified **Fig. 3b**.

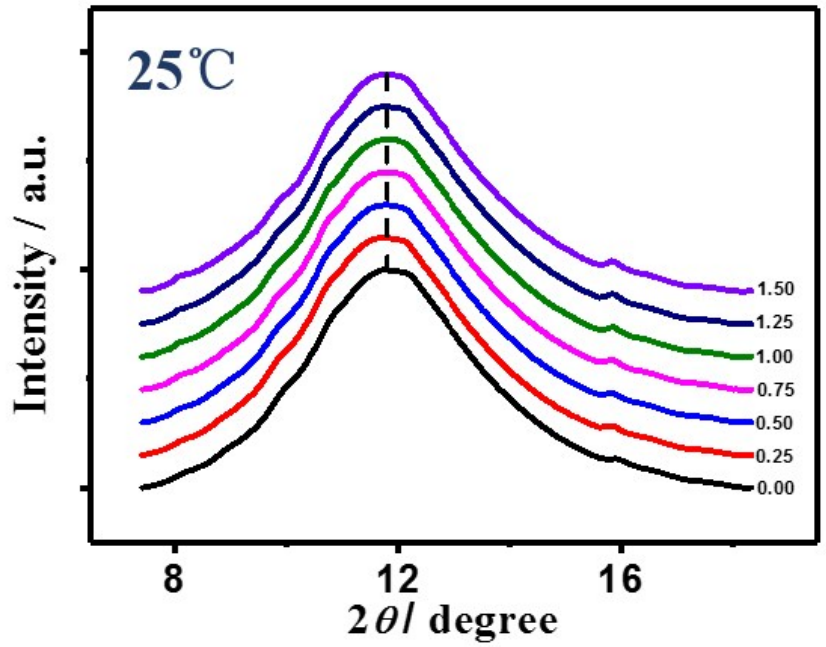

Fig. S4 Sequential evolution of 2D SR-WAXD patterns during uniaxial stretching at different temperatures. Magnified **Fig. 3c**.

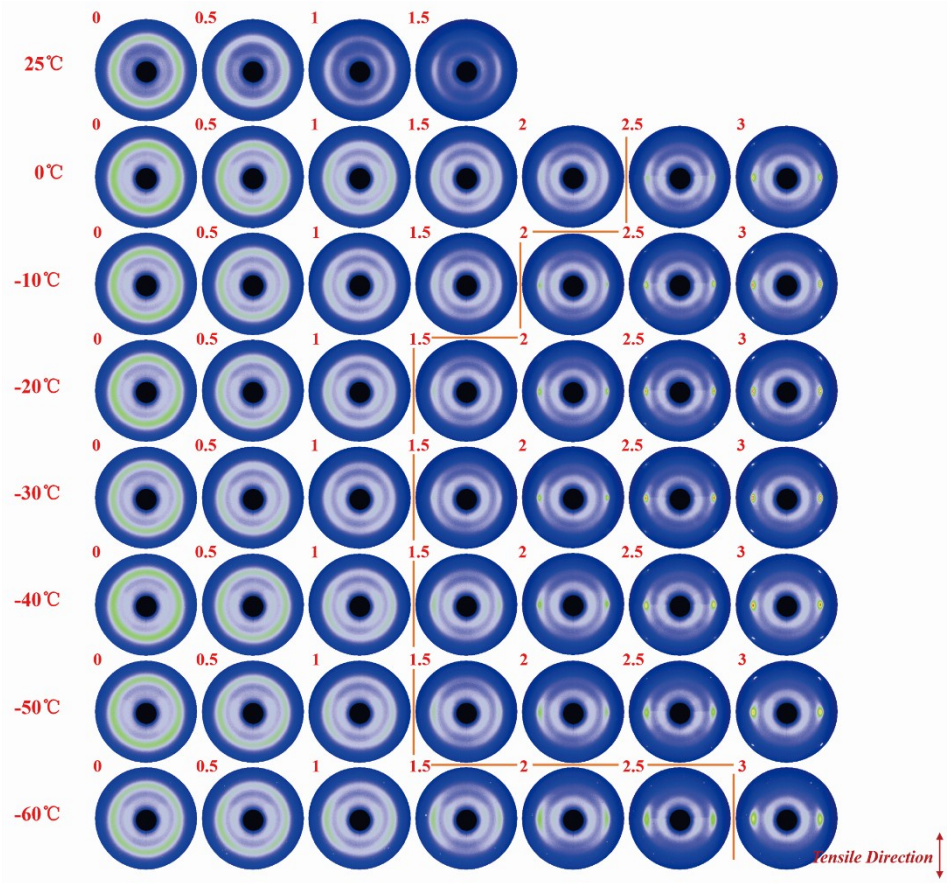

Fig. S5 Sequential evolution of 2D SR-WAXD patterns during uniaxial stretching at different temperatures. Magnified **Fig. 4**.