Supporting Information

An Unusual Spherulite Morphology Induced by Nano-fillers from Concentrated Cellulose Ionic Liquid Solution

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Experimental Section

Materials

Microcrystalline cellulose (MCC) was Vivapur 101, purchased from the Sen-Jun Chemical Agents Accessories Co. Ltd, Shanghai, China. The degree of polymerization was about 220, measured by using the Ubbelohde viscometer. The ionic liquid, 1-allyl-3-methylimidazolium chloride (AmimCl), was synthesized and purified according to the literature.[16] The water content of AmimCl was 0.26 wt%, determined by Karl Fischer titration. Multi-walled carbon nanotube (MWCNTs) was provided by the Nano Port Co. Ltd, Shenzhen, China, with 40-60 nm in diameter, 0.5-50 mm in length. Nanoclay was provided by Shang-hai Grid Trading Co. Ltd. Graphene oxide (GO) was prepared by a modified Hummers method.[22]

Sample preparation

MCC/AmimCl solutions with concentrations of 16 and 20 wt% were prepared, then the MWCNTs, nanoclay, and GO were added, respectively, with contents of 0.1 wt %, 0.3 wt % and 0.5 wt % of cellulose. Then the MCC/AmimCl solution was cast on a glass slide, which was preheated to 100 ^{o}C on a hot stage and held at 100 ^{o}C for 10 min to eliminate the effects of thermal history before it was moved to another hot stage with a preset temperature (40 or 60 ^{o}C) in an environmental chamber. Its relative humidity was kept at about 25%.

Characterization

POM observation was carried out using a BX51 Olympus polarized optical microscope equipped with a Pixera Penguin 150CL CCD camera. AFM measurements were performed on Multi-mode 8 (Bruker Instruments Industry Co., Ltd.), and all images were taken with the tapping mode at ambient temperature. The micro-Raman instrument was DXRxi Raman Microscopes spectrometer from Thermo Fisher Scientific.

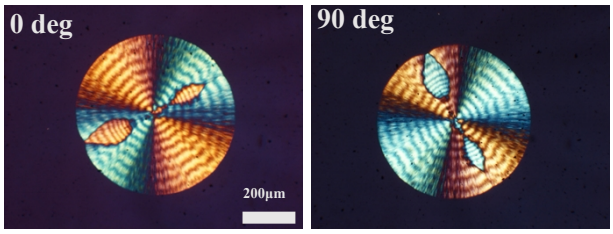

Figure S1. POM images of cellulose spherulites crystallized from 20 wt% MCC/AmimCl solution at 60 °C with 0.5 wt% nanoclay and by rotating the sample clockwise.

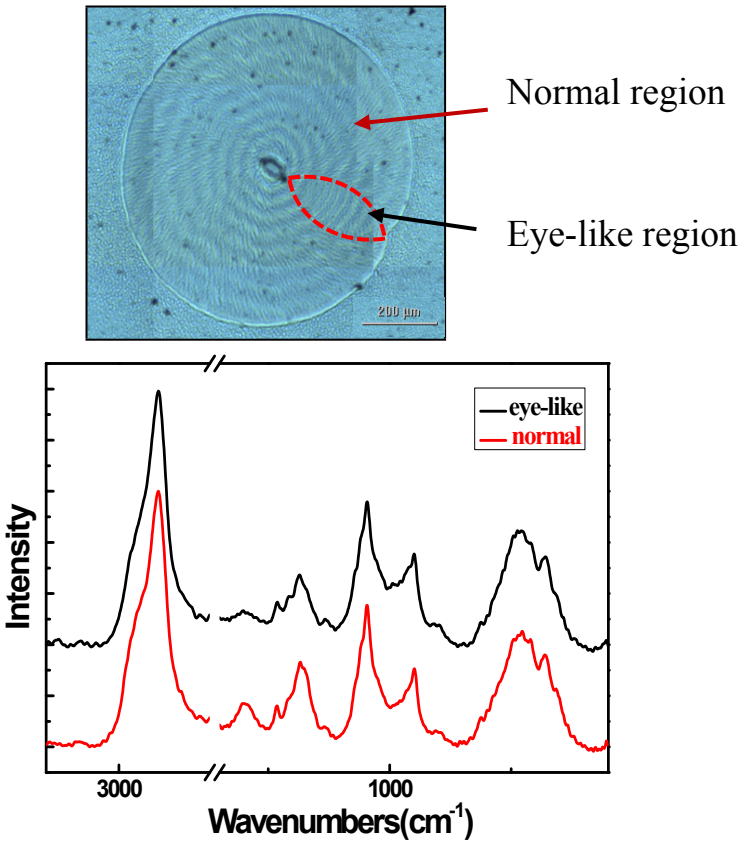

Figure S2. Micro-Raman Spectra of different regions of cellulose spherulite crystallized from 20 wt% MCC/AmimCl solution at 60 °C with 0.5 wt% MWCNTs.

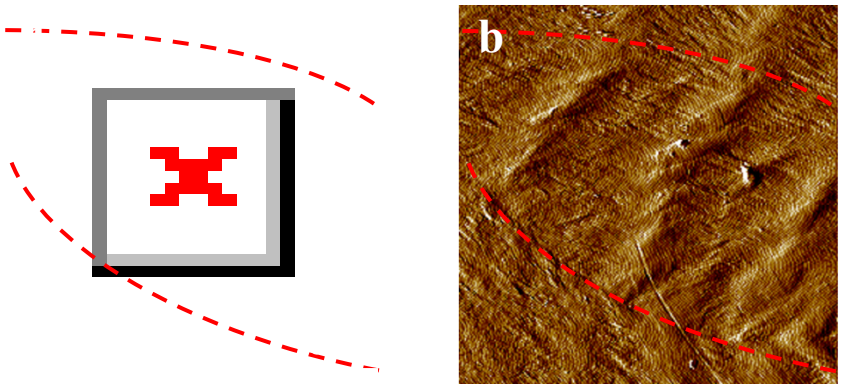

Figure S3. AFM images of cellulose spherulite crystallized from 16 wt% MCC/AmimCl solution at 40 °C with 0.5 wt% MWCNTs. Height (a) and phase (b) images.

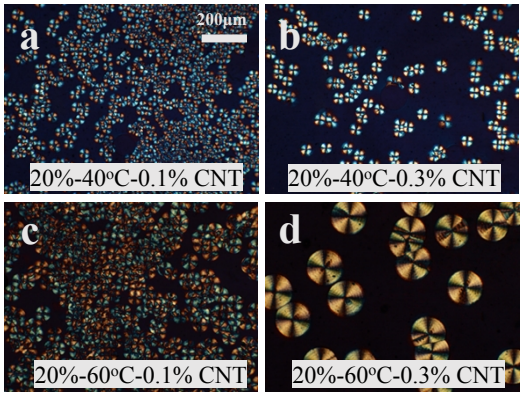

Figure S4. POM images of cellulose spherulites crystallized from 20 wt% MCC/AmimCl solution at 40 °C with 0.1 wt% MWCNTs (20 %-40 °C-0.1 % CNT) (a), and 0.3 wt% MWCNTs (20 %-40 °C-0.3 % CNT) (b); and 20 wt% solution at 60 °C with 0.1 wt% MWCNTs (20 %-60 °C-0.1 % CNT) (c), and 0.3 wt% MWCNTs (20 %-60 °C-0.3 % CNT) (d).