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The Effect of the Structural Arrangement of Binary Bisamide Organogelators on their Self-Assembly Behavior

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Abstract : Low-molecular-weight organogelators form gels by self-assembly into the crystalline network which immobilizes the organic solvent. For single bisamide organogelator systems, the effect of the molecular structure on the molecular interaction and their self-assembly behavior has been explored. The spatial arrangement of bisamide molecules in the gel-state is driven by a combination of hydrogen bonding and Van der Waals interactions. The hydrogen-bonding pattern between the amide groups of bisamide molecules is regulated by the number of methylene spacers; the even number of methylene spacers between two amide groups, in even-spaced bisamides, leads to the antiparallel position of amide groups within a molecule. An even-spaced bisamide molecule with antiparallel amide groups can make two pairs of hydrogen bonding with the molecules on the same plane. The odd-spaced bisamide with a parallel directionality of amide groups can form four independent hydrogen bonds with four other bisamide molecules on different planes. The arrangement of bisamide molecules in the crystalline state and the interaction of these molecules depends on the molecular structure, particularly the parity of the spacer length between the amide groups in the bisamide molecule. In this study, the directionality of amide groups has been exploited as a structural characteristic to affect the arrangement of molecules in the crystalline state and produce different binary bisamide gelators with different degrees of crystallinities. Single odd- and even-spaced single bisamides were synthesized and blended to produce binary bisamide organogelators to be characterized in order to understand the effect of the different directionality of amide groups on the molecular interaction in the crystalline state. The pattern of molecular interactions between these blended molecules, mixing or phase separation, has been monitored via differential scanning calorimetry (DSC) and crystallography techniques; X-ray powder diffraction (XRD) and Small-angle X-ray scattering (SAXS). The formation of lamellar structures for odd- and even-spaced bisamide gelators was confirmed by using SAXS and XRD techniques. DSC results have shown that binary bisamide organogelators with different parity of methylene spacers (odd-even binary blends) have a higher tendency for phase separation compared to the binary bisamides with the same parity (odd-odd or even-even binary blends). Phase separation in binary odd-even bisamides was confirmed by the presence of individual (100) reflections of odd and even lamellar structures. The structural characteristic of bisamide organogelators, the parity of spacer length in binary systems, is a promising tool to control the arrangement of molecules and their crystalline structure.

Keywords : binary bisamide organogelators, crystalline structure, phase separation, self-assembly behavior
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