

## Revealing the Effect of Additive Molecular Structure on Inducing Spherulitic Growth of L-Isoleucine

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Table S1. Detailed information on the materials used

Materials	CAS No.	Specification
L-Ile	73-32-5	Purity: 99%
L-Ala	56-41-7	Purity: 99%
L-Val	72-18-4	Purity: 99%
L-Phe	63-91-2	Purity: 99%
L-Ser	56-45-1	Purity: 99%
L-Tyr	60-18-4	Purity: 99%
L-Lys	56-87-1	Purity: 99%
MC	9004-67-5	Viscosity: 1500 mPa·s
HEC I		Viscosity: 80-120 mPa·s
HEC II	9004-62-0	Viscosity: 1000-1500 mPa·s
HEC III		Viscosity: 8000-12000 mPa·s
CMC-Na I		Viscosity: 50-100 mPa·s
CMC-Na II	9004-32-4	Viscosity: 1200-1500 mPa·s
CMC-Na III		Viscosity: 8000-12000 mPa·s

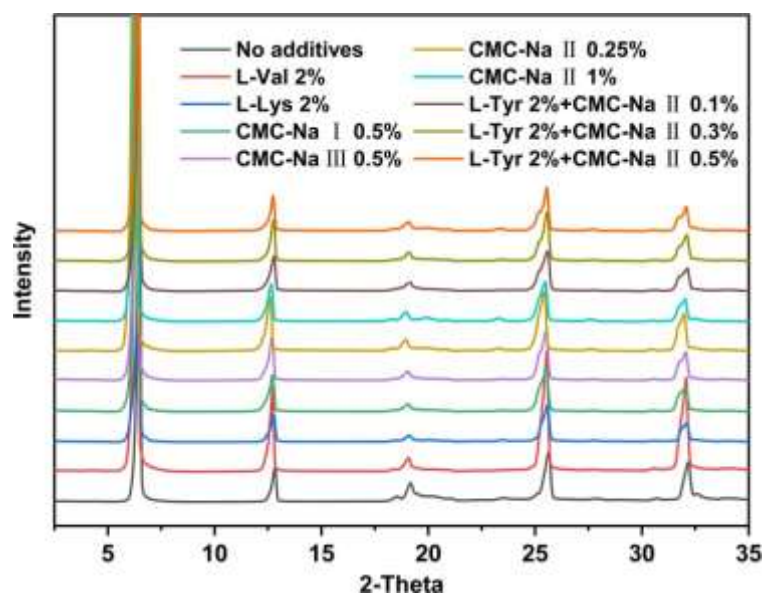


Figure S1. PXRD pattern of pure L-isoleucine crystals and those modified by various additives.

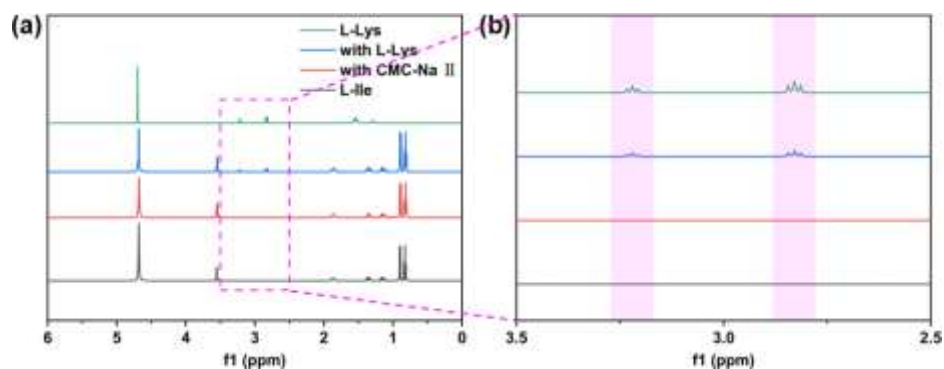


Figure S2.  $^1\text{H}$  NMR spectra (using  $\text{D}_2\text{O}$  as the solvent at 25 °C, operating at 500 MHz) of L-isoleucine products with different additive conditions and pure L-Lys.

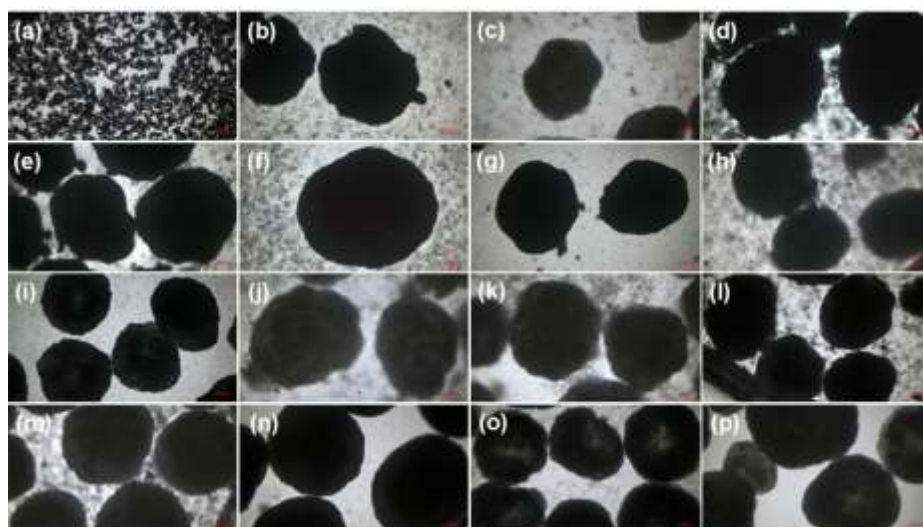


Figure S3. Optical microscope images of L-isoleucine under different additive conditions: (a) no additive, (b) with L-Ala at 2%, (c) with L-Val at 2%, (d) with L-Phe at 2%, (e) with L-Ser at 2%, (f) with L-Lys at 2%, (g) with L-Tyr at 2%, (h) with HEC at 0.5%, (i) with CMC-Na II at 0.5%, (j) with CMC-Na I at 0.5%, (k) with CMC-Na III at 0.5%, (l) with CMC-Na II at 0.25%, (m) with CMC-Na II at 1%, (n) with L-Tyr at 2% and CMC-Na II at 0.1%, (o) with L-Tyr at 2% and CMC-Na II at 0.3%, (p) with L-Tyr at 2% and CMC-Na II at 0.5%.

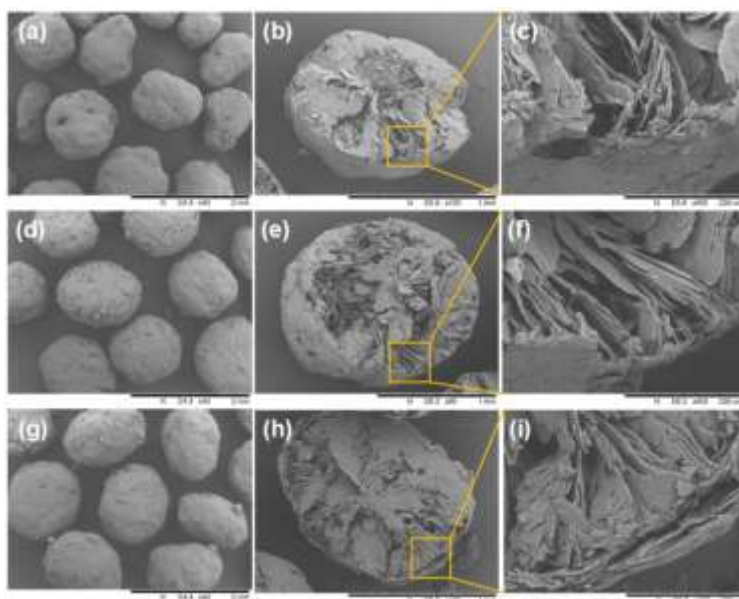


Figure S4. SEM images showing the morphology of L-isoleucine crystals grown in the presence of different amino acid additives: (a-c) L-Ala, (d-f) L-Phe, and (g-i) L-Ser. (a, d, g) Overall surface morphology. (b, e, h) Cross-section morphology. (c, f, i) Close-up views of the cross-sections.

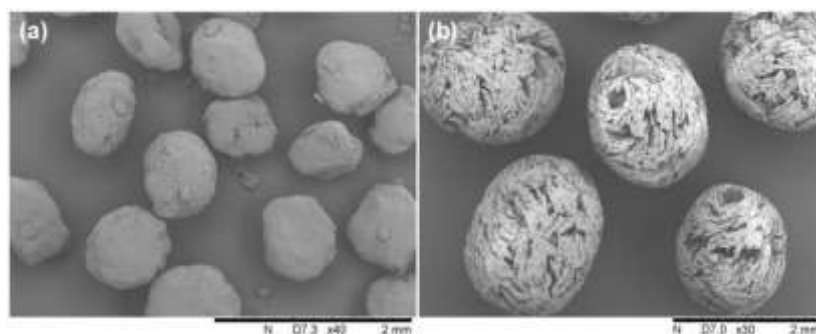


Figure S5. SEM images of L-isoleucine under different additive conditions with: (a) L-Val at 2%, (b) L-Lys at 2%.

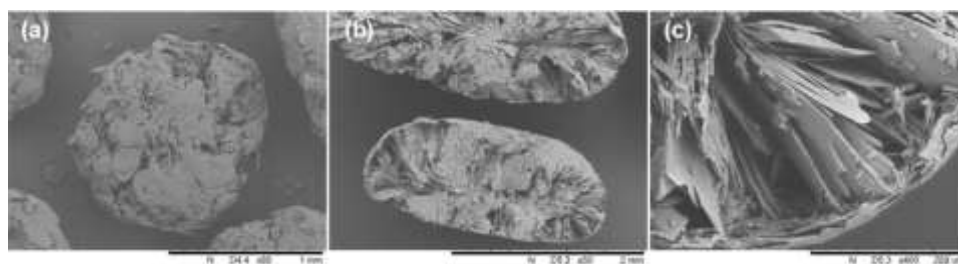


Figure S6. SEM images of L-isoleucine crystals grown with MC as an additive: (a) overall surface morphology, (b) cross-section morphology, (c) close-up view of the cross-section.

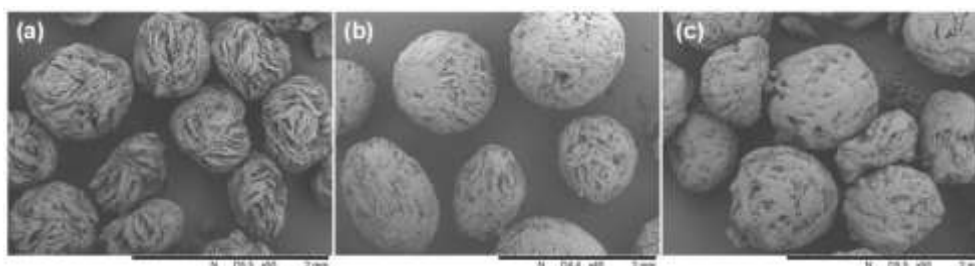


Figure S7. SEM images of L-isoleucine under the action of CMC-Na II at (a) 0.25%, (b) 0.5%, (c) 1%.

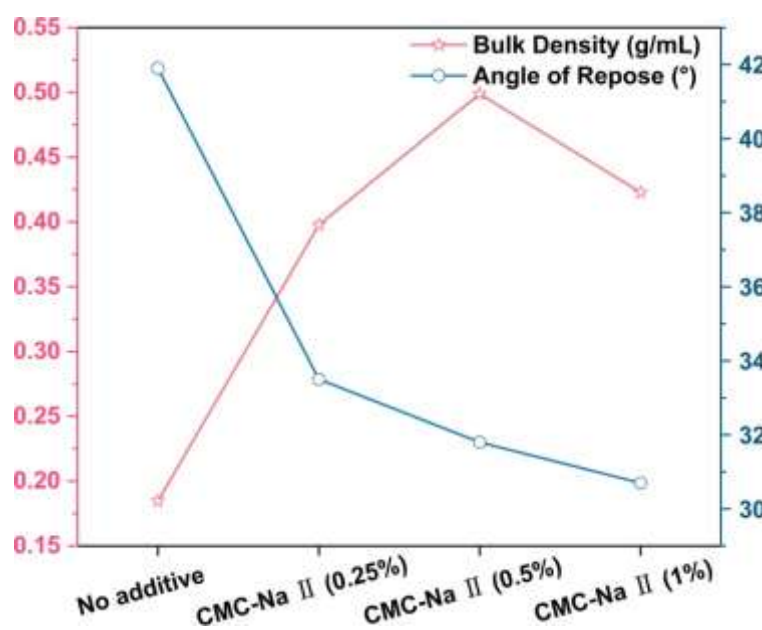


Figure S8. Effect of additive concentration on powder properties of L-isoleucine crystal products.

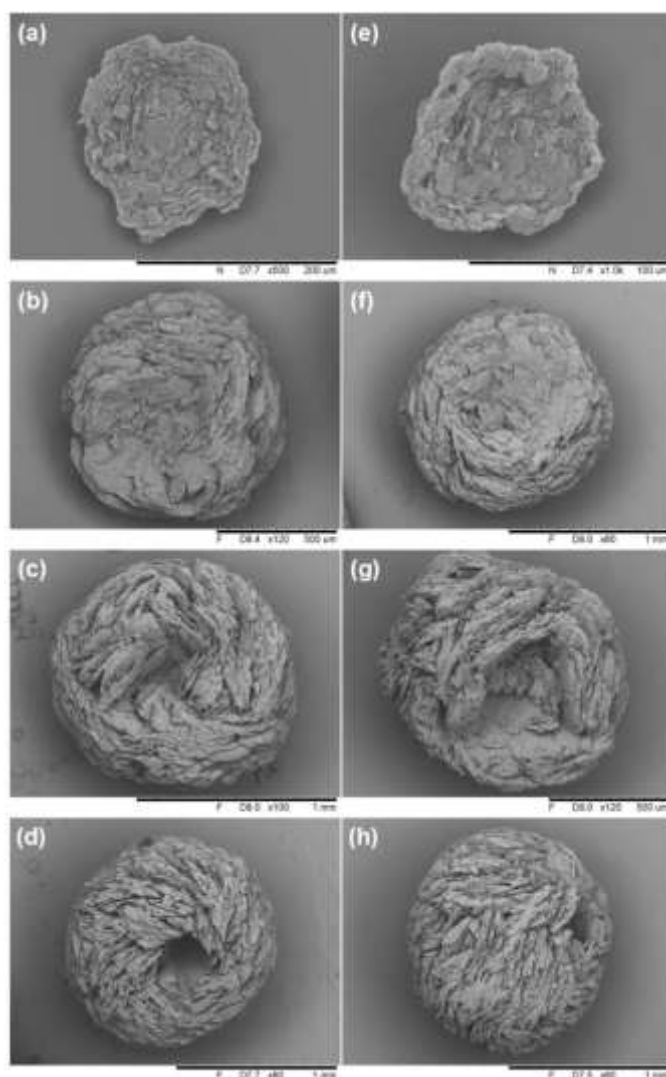


Figure S9. SEM images showing the morphological evolution of the L-isoleucine spherulite evaporated at a rate of 0.5 mL/min, temperature of 70°C, and addition of (a–d) 0.5% CMC-Na II, (e–h) 2% L-Tyr and 0.3% CMC-Na II.

Table S2. The BET specific surface area of L-isoleucine crystal samples under different conditions.

Additives (wt%)	Specific surface area (m <sup>2</sup> /g)
L-Ala (2%)	0.31
L-Ser (2%)	0.49
L-Phe (2%)	0.78
L-Tyr (2%)	1.51
MC (0.5%)	0.39
HEC II (0.5%)	0.75
CMC-Na II (0.5%)	1.09

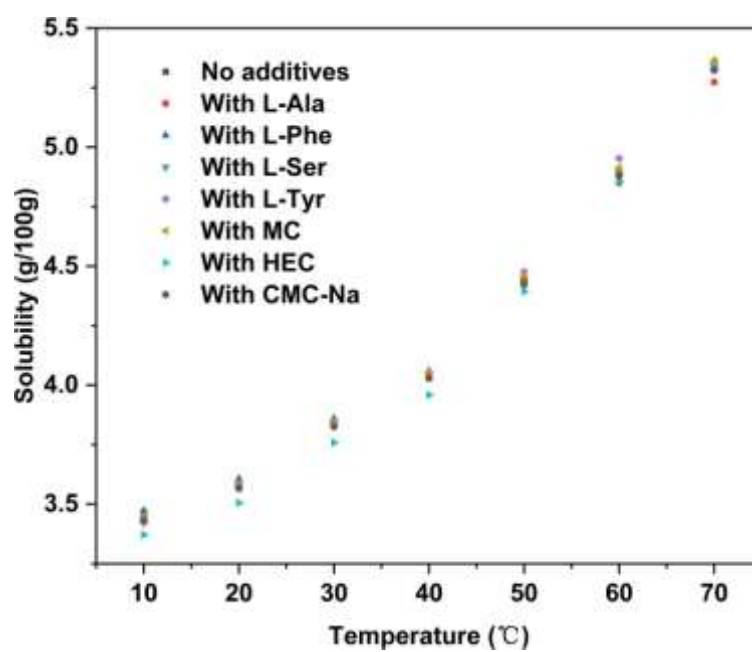


Figure S10. Mass fraction solubility of L-isoleucine in pure water or with additives at different temperatures.