

**Supplementary File 1:** Determining the preparation conditions of CS-LA complexes for HPP treatments

The preparation conditions of CS-LA complexes significantly affect the degree of complexation. The first influencing factor we explored was the lipid concentration, as the appropriate lipid concentration was essential to improve the degree of complexation. In previous research, it was demonstrated that subjecting potato starch and myristic acid to 400 MPa HPP treatment resulted in a higher degree of complexation and improved physicochemical properties. Therefore, 400 MPa was selected to investigate the impact of varying LA additions on the encapsulation efficiency (EE) and complex index (CI) of SLC, as shown in [Supplementary Fig. S1](#). Under ambient pressure, with an LA addition of 2%, the EE was only 1.52%, and the CI was 0.03%. As the addition amount increased, the EE initially increased and then decreased, reaching a maximum of 18.28% when 10% of LA was added. The CI gradually increased and leveled off with LA addition, peaking at 1.83% at 10% of LA addition. Further increases in LA addition did not result in significant differences in the complex index ( $p > 0.05$ ). These results indicated that under ambient pressure, the starch was saturated when 10% LA was added. LA interacted with long-chain amylose at lower lipid concentrations through non-covalent bonds, such as hydrogen bonds and van der Waals's force. As the LA concentration increases from 2% to 10%, it can also bind to short-chain amylose to form complexes, leading to an increase in both EE and CI. When the lipid concentration is above 10%, no active sites on the starch are available for further interaction with fatty acids. A similar phenomenon has also been observed by Kawai et al., where the CI of potato starch- fatty acid complex elevated with fatty acid/starch ratios increases from 0 to 1.0 mmol/g but plateaued after 1.0 mmol/g.

Also illustrated in [Supplementary Fig. S1](#), with 400 MPa HPP treatment, both EE and CI increased significantly ( $p < 0.05$ ). At a 2% LA addition, the encapsulation efficiency improved by 15.39% compared to the result observed at ambient pressure, and CI increased by 0.31%. The EE of SLC at 400 MPa showed a similar trend of initial increase followed by a decrease, reaching a maximum of 35.30% at 10% LA addition. The CI increased and plateaued with LA addition, peaking at 3.36% at 10% LA addition. Further increases in LA addition did not result in significant differences in the complex index ( $p > 0.05$ ). These indicated that although HPP could increase the EE and CI of SLC, the maximum was observed when 10% LA was used. It was probably due to the intermolecular interaction between fatty acids, which are prone to form micellar structures with each other at higher concentrations. To maximize starch utilization for fatty acid encapsulation, 10% LA addition was chosen as the condition for subsequent experiments.

Reaction temperature is another key factor in determining the degree of complexation of SLCs. Therefore, EE and CI of the CS-LA complex at ambient and high pressures (400 MPa/10 min) were investigated at 30, 40, 50, 60, and 70 °C, respectively, and the results are shown in [Supplementary Fig. S2](#). It could be observed that as temperature increased from 30 °C to 70 °C, the EE under ambient pressure gradually increased and became stable between 40 °C and 70 °C. Under 400 MPa, EE was not significantly different among the various temperatures. However, at 30 °C and 40 °C, there was a notable difference in EE between the samples at the same temperature but different pressures. In comparison, when the temperatures were above 50 °C, no significant difference was observed between the samples treated at different pressures. The CI exhibited a similar trend. Although at 40 °C, the EE and CI under ambient pressure were significantly different compared to the one at 30 °C, no such difference was observed under 400 MPa. Moreover, the EE and CI at 30 °C were more sensitive to pressure treatment than 40 °C. To avoid the interference of thermal treatment on the

HPP treatment regarding starch structural properties, the reaction temperature was kept at 30 °C for subsequent experiments.