Composition of anthocyanin multilayer layer-by-layer edible films using polyelectrolyte complexes of polysaccharide-protein nanoparticles

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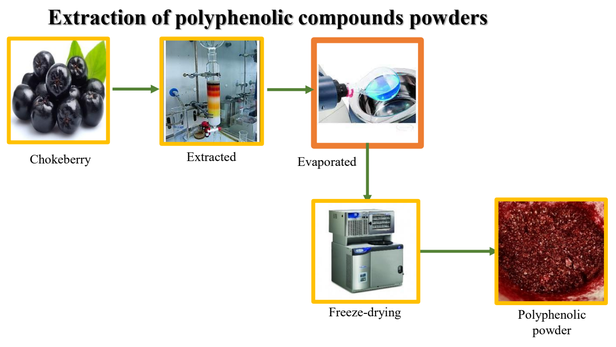
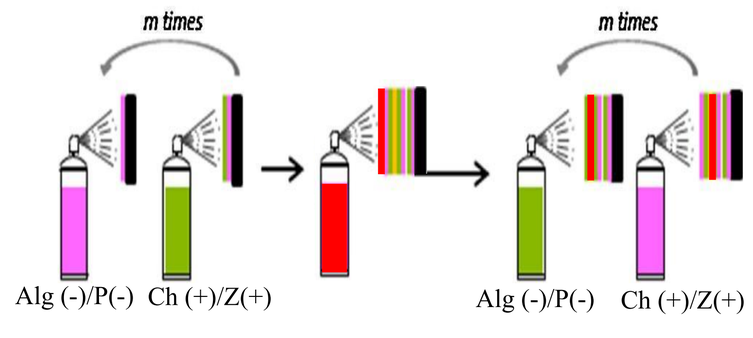
Introduction and Aim

Anthocyanins are water-soluble flavonoids with strong antioxidant, anti-inflammatory, and microbiota-modulating effects, supporting the prevention of chronic diseases such as IBD and certain cancers [1]. However, their use in functional foods is limited due to poor bioavailability (<1%) and instability under pH, temperature, and light [2]. Layer-by-layer (LbL) encapsulation using natural polymers offers a promising strategy to improve anthocyanin stability and delivery [3]. This bottom-up technique allows precise nanoscale control of multilayer structures, with high reproducibility and adaptability [4,5]. Natural biopolymers like polysaccharides and proteins enhance LbL's potential due to their biocompatibility, biodegradability, and safety for food applications [6]. **Therefore, the aim of this study was to determine the composition of multilayer layer-by-layer edible films using a polyelectrolyte polysaccharide-protein complex.**

Experimental





Results

Characterization of zein–anthocyanin complexes

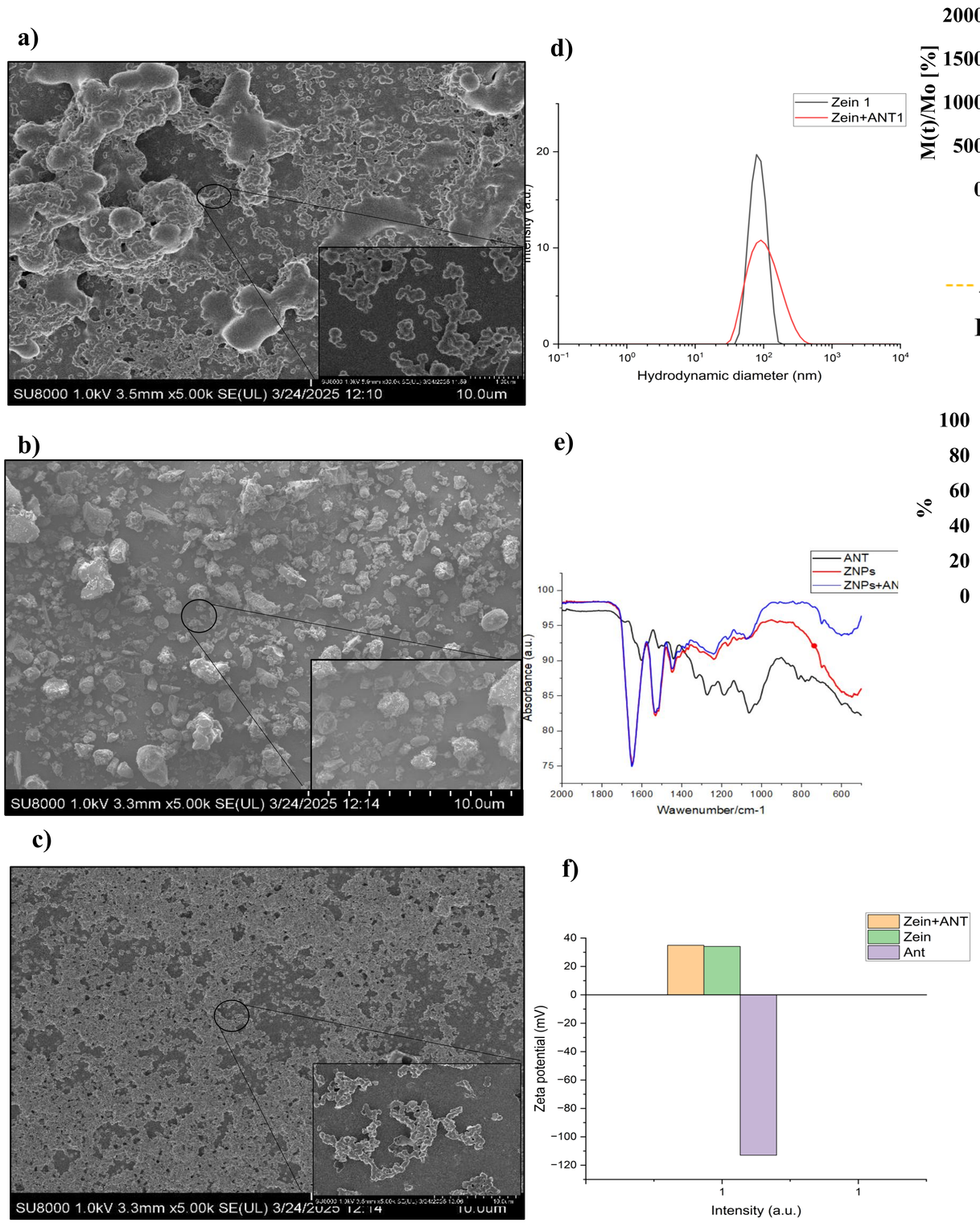


Fig. 1. Characterization of zein–anthocyanin complexes: (a) SEM image of zein nanoparticles (ZNP);(b) SEM image of ZNP coacervated with anthocyanins (ZNP+ANT);(c) SEM image of ANT;(d) Zeta potential values of ANT, ZNP, and ZNP+ANT;(e) Hydrodynamic diameter of ZNP and ZNP+ANT measured by dynamic light scattering (DLS);(f) ATR-FTIR spectra of ANT, ZNP, and ZNP+ANT, indicating functional group interactions during complexation.

Characteristics of edible LbL films

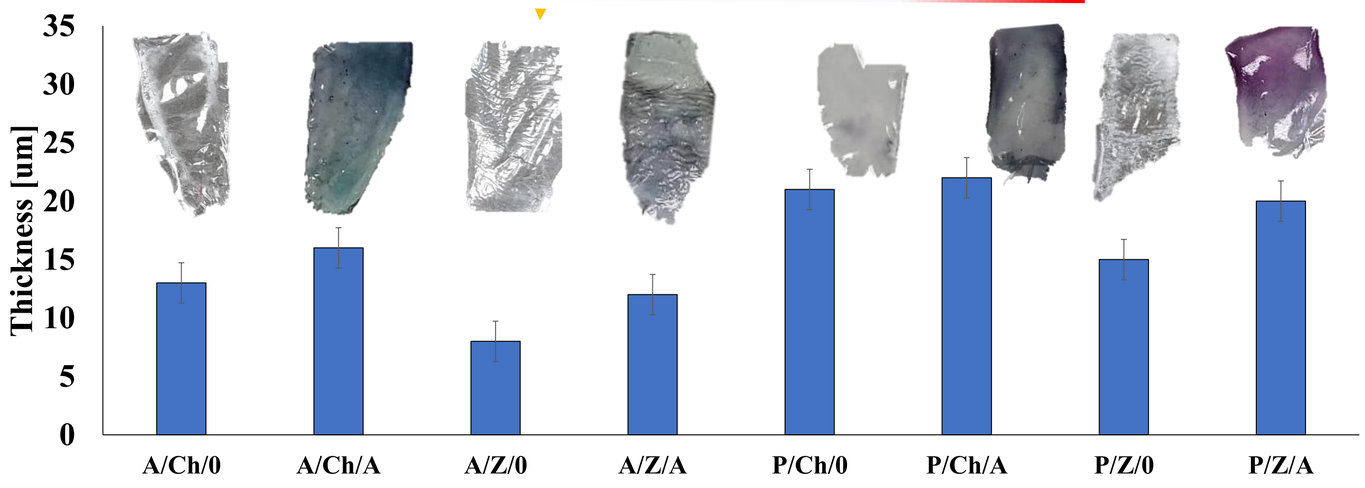


Fig. 2. Composition of preparing films and thickness

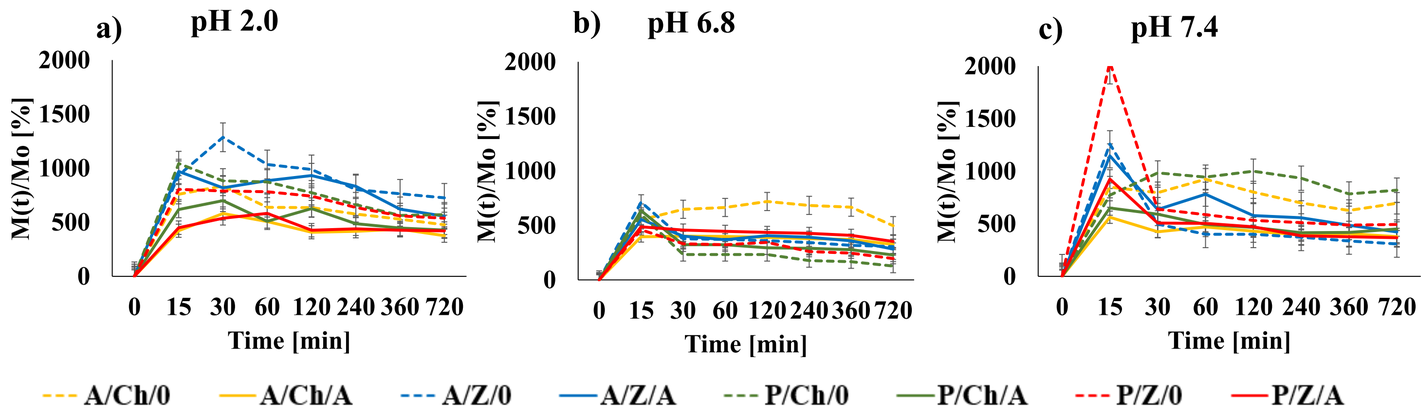


Fig. 3. Swelling assays of film in time at a) pH 2.0, b) pH 6.8, c) pH 7.4

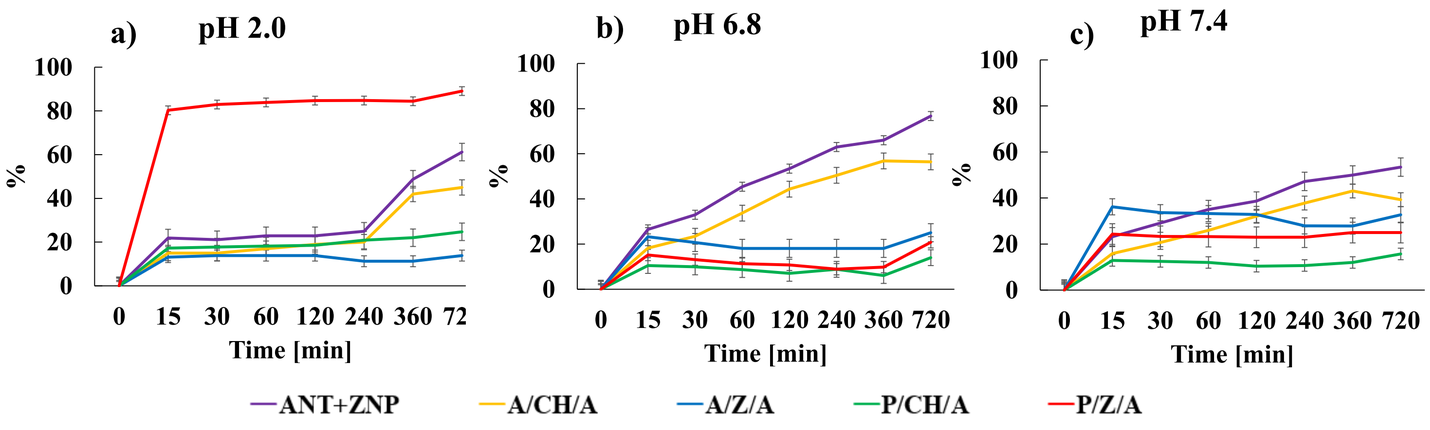
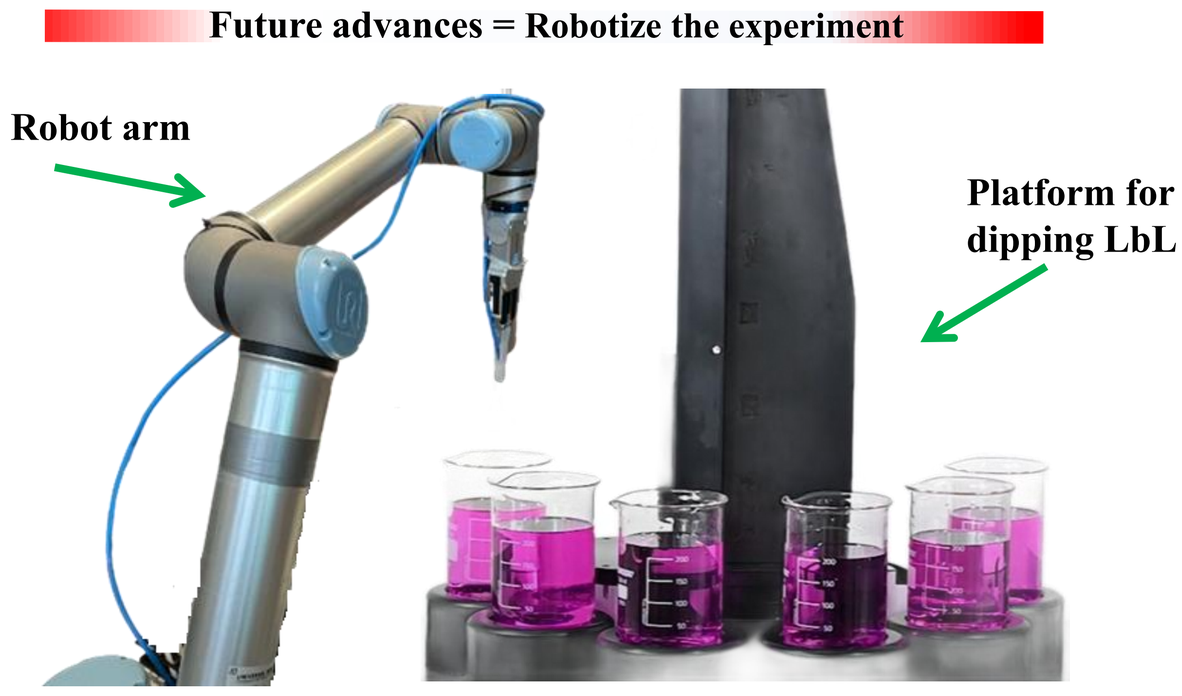


Fig. 4. Degradation of anthocyanins in time at a) pH 2.0, b) pH 6.8, c) pH 7.4

Future advances = Robotize the experiment



Conclusions

This study demonstrated that anthocyanins encapsulated in zein nanoparticles and incorporated into multilayer edible films using a layer-by-layer approach form stable, structured coacervates with distinct morphology and controlled physicochemical properties. The resulting 40-layer films exhibited good mechanical integrity, moderate swelling, and pH-responsive release behavior, effectively protecting anthocyanins under gastric conditions and enabling gradual release at intestinal pH. The composition of the biopolymer matrix (including zein, alginate, pectin, and chitosan) influenced the release profile, confirming its critical role in delivery performance. The most promising results were observed for multilayer complexes combining pectin with either zein or chitosan, which demonstrated enhanced stability and controlled anthocyanin release. These findings highlight the system's potential for use in gut-targeted nutraceutical applications. In future work, the system will be fully automated to enable scalable and precise fabrication of multilayer delivery structures.

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