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Experimental Study of the Antibacterial Activity and Modeling of Non-isothermal Crystallization Kinetics of Sintered Seashell Reinforced Poly(Lactic Acid) And Poly(Butylene Succinate) Biocomposites Planned for 3D Printing

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Abstract : The use of additive manufacturing technologies has revolutionized various aspects of our daily lives. In particular, 3D printing has greatly advanced biomedical applications. While fused filament fabrication (FFF) technologies have made it easy to produce or prototype various medical devices, it is crucial to minimize the risk of contamination. New materials with antibacterial properties, such as those containing compounded silver nanoparticles, have emerged on the market. In a previous study, we prepared a newly sintered seashell filler (SSh) from bio-based seashells found along the Mediterranean coast using a suitable heat treatment process. We then prepared a series of polylactic acid (PLA) and polybutylene succinate (PBS) biocomposites filled with these SSh particles using a melt mixing technique with a twin-screw extruder to use them as feedstock filaments for 3D printing. The study consisted of two parts: evaluating the antibacterial activity of newly prepared biocomposites made of PLA and PBS reinforced with a sintered seashell in the first part and experimental and modeling analysis of the non-isothermal crystallization kinetics of these biocomposites in the second part. In the first part, the bactericidal activity of the biocomposites against three different bacteria, including Gram-negative bacteria such as (E. coli and Pseudomonas aeruginosa), as well as Gram-positive bacteria such as (Staphylococcus aureus), was examined. The PLA-based biocomposite containing 20 wt.% of SSh particles exhibited an inhibition zone with radial diameters of 8mm and 6mm against E. coli and Pseudo. Au, respectively, while no bacterial activity was observed against Staphylococcus aureus. In the second part, the focus was on investigating the effect of the sintered seashell filler particles on the non-isothermal crystallization kinetics of PLA and PBS 3D-printing composite materials. The objective was to understand the impact of the filler particles on the crystallization mechanism of both PLA and PBS during the cooling process of a melt-extruded filament in (FFF) to manage the dimensional accuracy and mechanical properties of the final printed part. We conducted a non-isothermal melt crystallization kinetic study of a series of PLA-SS and PBS-SS composites using differential scanning calorimetry at various cooling rates. We analyzed the obtained kinetic data using different crystallization kinetic models such as modified Avrami, Ozawa, and Mo's methods. Dynamic mode describes the relative crystallinity as a function of temperature; it found that time half crystallinity (t1/2) of neat PLA decreased from 17 min to 7.3 min for PLA+5 SSh and the (t1/2) of virgin PBS was reduced from 3.5 min to 2.8 min for the composite containing 5wt.% of SSh. We found that the coated SS particles with stearic acid acted as nucleating agents and had a nucleation activity, as observed through polarized optical microscopy. Moreover, we evaluated the effective energy barrier of the non-isothermal crystallization process using the Iso conversional methods of Flynn-Wall-Ozawa (F-W-O) and Kissinger-Akahira-Sunose (K-A-S). The study provides significant insights into the crystallization behavior of PLA and PBS biocomposites.

Keywords : avrami model, bio-based reinforcement, dsc, gram-negative bacteria, gram-positive bacteria, isoconversional methods, non-isothermal crystallization kinetics, poly(butylene succinate), poly(lactic acid), antbactirial activity
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