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RECENT PROGRESS IN BIO-BASED POLYLACTIDE FILM FORMING EMULSIONS DEVELOPMENT

Serhii KHLYSTUN,
Ph.D. student of
Igor Sikorsky Kyiv Politechnic Institute
s.khlystun@kpi.ua

Oleksiy MYRONYUK,
D.Sc., As. Prof.
Igor Sikorsky Kyiv Politechnic Institute
o.myronyuk@kpi.ua

Abstract: *Poly(lactic acid) (PLA), a biodegradable biopolymer, faces challenges in coatings due to film formation and stability issues. To enhance performance, researchers use plasticizers, flexibilizers, and cross-linking agents. Emulsification techniques like solvent evaporation, high-shear mixing, and ultrasonic emulsification aid PLA dispersion, with recent focus on solvent-free methods such as reactive extrusion. Stabilizers like polyvinyl alcohol, casein, and pectin improve emulsion stability. However, issues like phase separation, high energy demands, and residual solvents persist. Research into eco-friendly stabilizers, green processing, and PLA nanocomposites aims to enhance mechanical properties and scalability. PLA emulsions show promise for wood finishes, food packaging, and biomedical coatings, reducing volatile organic compound emissions. Advancements in formulation and processing will be key to unlocking PLA-based coatings' full potential.*

Keywords: *PLA, biopolymer, polymer, film, coatings*

In the quest for sustainable development, the transition from fossil-based to renewable or bio-based raw materials in polymer production has become increasingly essential. Using renewable sources reduces reliance on limited fossil resources, lowers greenhouse gas emissions, and decreases the environmental impact of polymer production and disposal. In particular, poly(lactic acid) (PLA), a biopolymer derived from renewable sources like corn starch or sugarcane, has gained attention for its biodegradability and low carbon footprint. However, its application in coatings remains underexplored, partly due to the challenges posed by its inherent properties.

It was indicated that one of the primary limitations of PLA and other biopolymers in coatings is their film-forming ability [1]. Traditional petroleum-based polymers offer a proven track record of forming durable, flexible films critical for coating applications, whereas biopolymers often struggle to meet these standards [2]. To address this, researchers are investigating various approaches to enhance the film-forming properties of PLA and similar biopolymers. By incorporating additives such as plasticizers, flexibilizers, and cross-linking agents, it is possible to improve flexibility, durability, and adhesion, paving the way for biopolymer coatings with enhanced performance.

The synthesis of polylactic acid (PLA) emulsions involves processes aimed at dispersing hydrophobic PLA particles into a continuous aqueous phase. These methods are influenced by the physical and chemical properties of PLA, such as its high hydrophobicity and limited water solubility. Common techniques include: solvent evaporation method, solvent diffusion method, high-shear or ultrasonic emulsification, emulsion-solvent-free method [3].

Solvent evaporation method is one of the most widely used techniques for preparing PLA emulsions. In this method, PLA is first dissolved in an organic solvent (e.g., dichloromethane or acetone). The organic phase is then emulsified into water containing surfactants to stabilize the dispersion. After emulsification, the organic solvent is evaporated, leaving behind PLA particles suspended in the aqueous phase. This method has the following aspects: produces uniform particles with controllable sizes, limited by the need for organic solvents and complex solvent recovery processes, requires volatile organic compounds (VOCs), raising concerns about emissions and solvent disposal.

Solvent diffusion method is similar to the solvent evaporation method, this approach relies on the diffusion of a water-miscible organic solvent (e.g., acetone) into the aqueous phase. This results in the precipitation of PLA as fine particles. This method has the following aspects: suitable for smaller-scale applications with moderate energy input, more feasible than solvent evaporation due to lower solvent recovery requirements, less solvent-intensive but still involves VOCs.

In a high-shear or ultrasonic emulsification method, molten PLA or PLA in a solvent is broken into fine droplets within an aqueous phase using high-shear mixers or ultrasonic waves. Stabilizers or surfactants are used to prevent particle coalescence.

This method produces stable emulsions with relatively low surfactant concentrations, uses high-shear devices that are scalable for industrial production and eliminates organic solvents, making it more eco-friendly.

Recent advances have focused on avoiding solvents altogether by using reactive extrusion or direct melt emulsification. Emulsion-solvent-free method involve mixing molten PLA with water under controlled conditions to form emulsions directly. This method has high efficiency, as it avoids solvent evaporation steps, promising for large-scale production, environmentally sustainable due to the elimination of harmful solvents.

Belletti et al. [4] demonstrated that stabilizing agents and emulsification systems are critical for producing stable PLA emulsions. They prevent phase separation by forming a protective layer around PLA particles in the aqueous phase, ensuring the long-term stability of emulsions.

Surfactants like sodium dodecyl sulfate (SDS) and polyvinyl alcohol (PVA) are commonly used. They reduce the interfacial tension between PLA and water, aiding in the formation of uniform emulsion droplets. The choice of surfactant significantly influences particle size and stability. SDS provides effective stabilization but can have environmental concerns, whereas PVA offers a more eco-friendly option.

Mileti et al. [5] explored polysaccharides (e.g., pectin) and proteins (e.g., casein) as natural stabilizers. These biopolymers can form strong viscoelastic interfaces that enhance the mechanical and thermal stability of PLA emulsions. Their natural origin aligns with the eco-friendly goals of PLA-based systems.

Mixed stabilizers, such as combinations of surfactants and biopolymers, are gaining attention. For instance, systems using both casein and pectin demonstrate enhanced stabilization through synergistic interactions, where proteins provide interfacial activity and polysaccharides add thickening properties.

Methods like high-shear mixing and ultrasonic emulsification are often paired with stabilizing agents. These techniques ensure better dispersion of PLA particles and maximize the effectiveness of the stabilizers used.

Andrzejewski et al. [6] and Jem & Tan [7] described several key challenges in the development of PLA emulsions. Achieving stable emulsions with uniform particle size is challenging due to PLA's hydrophobic nature. The mismatch between the properties of PLA and water often leads to phase separation or coagulation during or after emulsification processes. This instability limits the broader applicability of PLA-based emulsions in coatings and other applications.

Many emulsification methods, such as ultrasonication and high-pressure homogenization, demand significant energy input. While effective, these methods increase production costs and may limit scalability.

Solvent-based methods, often employed to disperse PLA in water, can leave residual organic solvents, posing environmental and health risks. Developing solvent-free or green processing methods remains a critical focus for sustainability.

PLA's low thermal resistance and brittle nature further complicate its use in demanding applications, necessitating modifications or blending with other materials, which can affect biodegradability.

To address these challenges, the following research avenues are promising:

- investigating eco-friendly stabilizing agents, such as bio-based surfactants or polysaccharides, could improve emulsion stability without compromising biodegradability [8].
- techniques like reactive extrusion or direct polymerization may reduce energy consumption and eliminate the need for organic solvents, creating more sustainable production processes [6].
- enhancing PLA properties: research on nanocomposites or bio-additives to improve PLA's thermal stability and mechanical strength could broaden its application in high-performance coatings [9].

PLA emulsions have potential applications across various industries. They can be used as wood finishes, paper coatings for food packaging, and biomedical coatings. These applications reduce volatile organic compound emissions and align with global environmental regulations. The focus is on creating coatings that are both high-performing and eco-friendly.

In conclusion, PLA emulsions offer an exciting avenue for sustainable coatings. Despite challenges like stability and scalability, ongoing research into novel stabilizers and green processing methods provides hope for their widespread use. PLA-based coatings hold great potential to meet both industrial and environmental needs.

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ОСТАННІ ДОСЯГНЕННЯ У СТВОРЕННІ ПЛІВКОУТВОРЮЮЧИХ ЕМУЛЬСІЙ НА ОСНОВІ БІОПОЛІМЕРА PLA

Сергій ХЛИСТУН
Аспірант,
Олексій МИРОНЮК
Д.т.н., доценет,
КПІ ім. Ігоря Сікорського

Анотація: *Полі(молочна кислота) (PLA), біорозкладний біополімер, стикається з проблемами при створенні покриттів через проблеми з утворенням плівки та стабільністю. Для покращення характеристик дослідники використовують пластифікатори, флексибілізатори та зшиваючі агенти. Методи емульгування, такі як випаровування розчинника, змішування з високим зсувом та ультразвукове емульгування, допомагають диспергуванню ПЛА, причому останнім часом основна увага приділяється методам, що не містять розчинників, таким як реактивна екструзія. Стабілізатори, такі як полівініловий* *спирт, казеїн і пектин, покращують стабільність емульсії. Однак залишаються такі проблеми, як фазовий поділ, високі енергетичні вимоги та залишкові розчинники. Дослідження екологічно чистих стабілізаторів, «зеленої» переробки та нанокомпозитів PLA спрямовані на покращення механічних властивостей та масштабованості. Емульсії PLA є перспективними для обробки деревини, пакування харчових продуктів та біомедичних покриттів, зменшуючи викиди летких органічних сполук. Досягнення в розробці рецептур і переробці стануть ключем до розкриття повного потенціалу покриттів на основі PLA.*

Ключові слова: *ПЛА, біополімер, полімер, плівка, покриття*