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Construction of Lignin-based Aqueous Coacervates and Their Mechanisms for Enhancing Pesticide Targeted Delivery

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Abstract. This review systematically explores the construction strategies and functional mechanisms of lignin-based aqueous coacervates for targeted pesticide delivery. By integrating lignin valorization with coacervate technology, we analyze the structural characteristics of different lignin types, compare various coacervate construction methods, and elucidate targeting mechanisms including environmental responsiveness and passive accumulation. Key findings demonstrate that lignin-based coacervates significantly enhance pesticide stability, bioavailability, and environmental compatibility. Despite challenges in scalability and precise control, these systems hold great promise for advancing sustainable agriculture through intelligent delivery platforms.

Keywords: Lignin; Aqueous coacervates; Pesticide delivery; Targeted; Sustainable agriculture.

1. Introduction

Based on a systematic analysis of the correlation between low pesticide utilization efficiency and environmental pollution, the potential for lignin valorization, and the advantages of aqueous coacervates in drug delivery systems, this review focuses on the construction strategies of lignin-based aqueous coacervates and their application mechanisms in the targeted delivery of pesticides.

Firstly, it systematically elaborates on the structural characteristics and functional modification pathways for different types of lignin. Secondly, it compares and analyzes the construction methods among three categories of aqueous coacervates. Finally, from three dimensions—environmentally responsive active targeting, passive targeting delivery, and auxiliary application technologies—it provides an in-depth analysis of the molecular mechanisms and application efficacy of lignin-based aqueous coacervates. This review provides a theoretical basis for the high-value utilization of lignin and the design of intelligent delivery systems for green pesticides.

1.1. Low Pesticide Utilization Efficiency and Environmental Pollution Status

Pesticides face the dual challenges of low utilization efficiency and environmental pollution. Globally, annual pesticide usage exceeds 2.4 million tons, but the effective foliar utilization of traditional formulations is less than 10%, with a significant portion lost due to weak adhesion and photolysis [1–3]. Formulations such as indoxacarb emulsifiable concentrate, with high organic solvent content, exhibit reduced deposition efficiency, compelling frequent high-dose applications [4].

Inefficient use leads to soil degradation, water eutrophication, and residues threatening human health [2–4]. Active ingredients can harm non-target organisms like *Harmonia axyridis*, damaging biodiversity and inducing pest resistance [1, 2, 4]. Nanotechnology and intelligent delivery systems offer promising solutions [1–3, 5, 6].

1.2. Valorization Potential of Lignin

Lignin accounts for 15%-30% of biomass dry weight and is the only renewable aromatic resource [7–9]. However, industries like pulping generate over 50 million tons of lignin by-products annually, with only 1%-2% utilized; the rest is often incinerated, wasting resources [8–10].

Its value is prominent: lignin-derived biochar exhibits a methylene blue adsorption capacity of 178.17 mg/g [11]; it can be complexed with PVA for UV-blocking films [8]; and converted to microbial lipids for biodiesel [9]. Challenges such as the complex structure persist [7]. Reviewing its value is crucial for the circular economy.

1.3. Advantages of Aqueous Coacervates in Drug Delivery

Aqueous coacervates, formed via non-covalent interactions, offer programmable architectures and biocompatibility [12, 13]. They address challenges like the low bioavailability of hydrophobic drugs and susceptibility to degradation [14–16].

Core advantages include mild preparation (encapsulation efficiency >90%) [12, 16], responsiveness to stimuli like pH for controlled release [12, 13, 16], adaptability to various routes, biodegradability, and potential to overcome multidrug resistance [16]. Systematizing their advantages is significant for precision delivery.

As illustrated in Fig 1, aqueous coacervates offer multiple advantages in pesticide delivery systems.

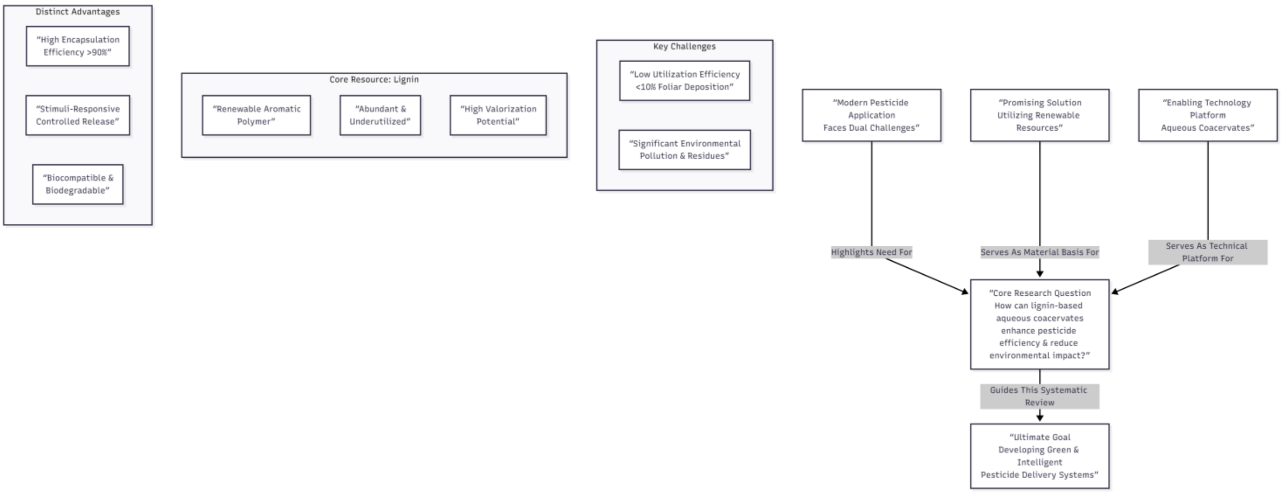

Figure 1. Schematic diagram of the advantages of aqueous coacervates in pesticide delivery systems

2. Lignin Sources

2.1. Alkali Lignin

Alkali Lignin is extracted via alkaline processes (e.g., kraft or soda). Its main characteristics include abundant phenolic hydroxyl groups, conferring good water solubility and reactivity.

Hu et al. (2021) investigated composite systems of xylan-rich hemicelluloses with alkali lignin, confirming that pH and ionic strength regulate aggregation. Introducing cationized xylan enhanced performance, providing a basis for biomimetic material design and biomass utilization [17].

Zhu Guodian's team (2023) used Dissipative Particle Dynamics (DPD) simulations to study Alkali Lignin self-assembly after graft modification. With increasing grafting degree, AL transitioned from random aggregates to multilamellar spherical micelles with a hydrophobic core and hydrophilic shell. This transition process is shown in Fig 2. This enables controlled hydrophobic-hydrophilic balance for drug carrier applications [18].

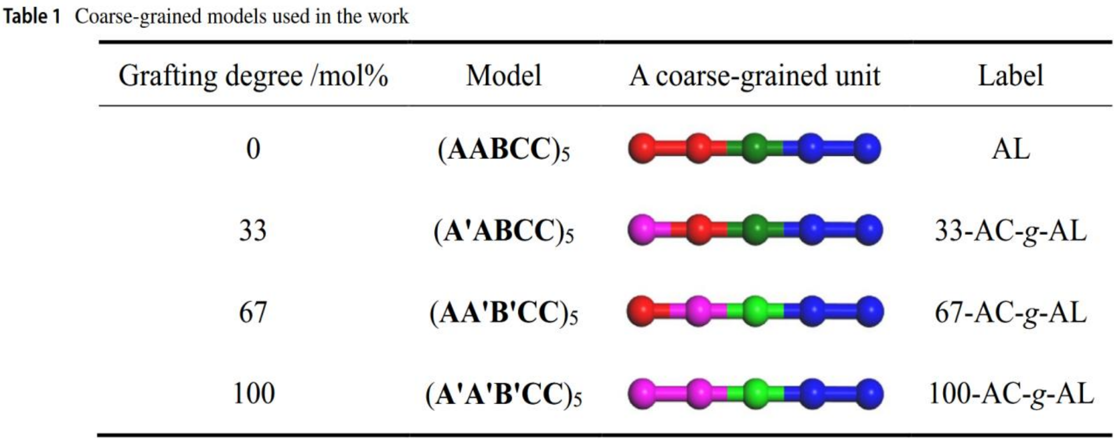

Table 1 Coarse-grained models used in the work

Grafting degree /mol%	Model	A coarse-grained unit	Label
0	**(AABCC)**$_5$		AL
33	**(A'ABCC)**$_5$		33-AC-*g*-AL
67	**(AA'B'CC)**$_5$		67-AC-*g*-AL
100	**(A'A'B'CC)**$_5$		100-AC-*g*-AL

Figure 2. Schematic diagram of the self-assembly transition process of alkali lignin[18]

Nilza's team (2024) isolated strain *Aspergillus ochraceus* DY1, achieving a 60.19% degradation rate for Alkali Lignin. The strain exhibited laccase-like activity and unique degradation products, showing potential for wastewater treatment and lignin valorization [19].

These studies provide multiple pathways for the functional modification and valorization of alkali lignin.

2.2. Enzymatically Hydrolyzed Lignin

Enzymatically hydrolyzed lignin is obtained by degrading the lignin-carbohydrate complex via enzymatic treatment. It retains the aromatic structure but has lower molecular weight and enhanced water solubility.

Yuan et al. (2021) systematically reviewed the influence of lignin structure on enzymatic hydrolysis efficiency. Lignin inhibits cellulase via non-productive adsorption, with efficiency differing between high S/G ratio and G-unit-rich lignin. Strategies like additive regulation (e.g., Mg^{2+}) and genetic engineering can improve efficiency, supporting bioethanol conversion optimization [14].

Khan's team (2022) used Sunburst™ technology to pretreat birch for hydrolysis lignin (HL). Alkaline laccase treatment increased lignin β-O-4 bonds by ~20%, verifying industrial feasibility for enzymatic catalytic conversion [20].

Gen Li's team (2025) studied Enzymatically Hydrolyzed Lignin solvolysis, finding dioxane and ethanol optimal for liquefaction and monomer yield. The process involves synergistic physical dissolution and chemical depolymerization, aiding fuel applications [21].

These works lay foundations for efficient utilization of enzymatically hydrolyzed lignin.

2.3. Organosolv Lignin

Organosolv Lignin is extracted using organic solvents (e.g., ethanol, acetone). It typically has higher purity, controllable molecular weight, and retains more active groups than traditional lignin.

In-Gyu Choi's team (2021) studied ethanol organosolv lignin (EOL), finding intensified extraction conditions decreased β-O-4 bonds but increased phenolic hydroxyl content, enhancing thermal stability. This provides a reference for EOL as a petroleum-based plastic substitute [22].

Lenarda et al. (2023) studied spruce organosolv lignin-derived carbon catalysts. Pore structure and oxygen content affected activity, with carbonyl/quinone groups as key sites. The balance between oxygen content and pore structure determines catalytic activity, informing biomass-derived carbon catalyst design [23].

Edita et al. (2025) optimized lignin fractionation, achieving 81.6% delignification at 160°C. High temperatures reduced molecular weight and increased ethoxylation, providing a fractionation strategy and industrial pathway for valorization [24].

These studies support organosolv lignin development in material substitution and catalysis.

3. Construction Methods of Aqueous Coacervates

3.1. Small Molecules/Simple Biomolecule-Based

Construction involves molecular self-assembly, liquid-liquid phase separation (LLPS), and dynamic covalent chemistry via non-covalent interactions or reversible covalent bonds, regulated by environmental factors.

Riahna's team (2022) enhanced C3M stability using irreversible amide (EDC) and reversible disulfide (DTBP) crosslinks. Crosslinked C3Ms showed improved stability under high salt/pH; DTBP-crosslinked micelles reverted with reducing agents, supporting controlled drug delivery [25].

Professor Ian W. Hamley's team (2024) studied minimal peptide LLPS, clarifying the pH-conformation-phase separation relationship and proposing a droplet stability framework. This offers insights into primitive life origins and artificial organelle design [26].

3.2. Synthetic Polymer-Based

Construction uses molecular design, crosslinking, and environmental regulation via hydrophilic/hydrophobic balance and dynamic bonds.

Wang Yilin and Fan Yaxun's team (2025) pioneered bio-based NaDC/polyamine copolymers for seed coating via phase separation. The coating enhanced germination, provided fungus and cold resistance, encapsulated functional molecules, and showed low toxicity, offering a sustainable agricultural solution [27].

Timothy J. Deming's group (2023) synthesized PEGylated block copolymers forming PCMs with 95% poly(A) encapsulation efficiency and physiological stability, providing new ideas for polynucleotide delivery [28].

Kembaren et al. (2022) regulated enzyme-containing C3M salt stability by adding a polyglutamate tail to laccase CotA. Modified micelles maintained monodispersity in high-salt environments while retaining enzyme activity, balancing stability and function [29].

3.3. Polymer-Nanoparticle-Based

Construction relies on polymer-nanoparticle interactions (electrostatic, H-bonding) regulated by pH, ions, and temperature.

Lee A. Fielding's team (2024) used PISA to prepare functionalized nanogels complexed with bPEI, creating ultra-stretchable (strain>1000%), self-healing hydrogels. Mechanical properties were regulable by Ca^{2+}, and adding graphene enabled conduction, providing a new paradigm for smart soft materials [30].

These methods provide diverse pathways for functional coacervates across fields.

4. Targeted Delivery Mechanisms

4.1. Environmentally Responsive Active Targeting

This mechanism uses environmentally responsive carriers (e.g., pH-, enzyme-, light-sensitive) to trigger release at target sites like acidic lesions or pest digestive tracts.

The team at Shanxi Agricultural University (2024) developed pH-responsive Thi@ZIF-8. The system showed acidity-responsive release and migration, low plant toxicity, and superior antifungal activity and persistence, offering insights for eco-friendly pesticide design [31].

Cen et al. (2022) constructed photothermal-responsive AVM@CS/CMA/PDA microcapsules. The PDA layer provided sustained-release and UV shielding. Under 808 nm NIR, photothermal conversion efficiency reached 14.93%, enhancing nematode eradication, applicable in pest control [3].

4.2. Passive Targeting Delivery

Passive targeting relies on carrier properties (size, charge) and organism physiology (e.g., EPR effect, cuticle retention) for directional accumulation.

A joint team (2022) reviewed MOFs like UiO-66-NH2/SL for sustained pesticide release and adsorption, conforming to pseudo-second-order kinetics, showing environmental sustainability and providing ideas for efficient pesticide use [32].

Mo et al. (2021) developed lignin-based Av-NDs with azobenzene crosslinking, achieving near-zero-order release at pH=7, enhancing abamectin photostability and prolonging efficacy, offering a new solution for pesticide delivery optimization [33].

4.3. Targeted Pesticide Application Assistance Technology

This involves biological recognition, carrier delivery, physical positioning (drones/sensors), and chemical guidance (attractants).

Wang Yunfei et al. (2025) reviewed LiDAR, multispectral imaging, and YOLO algorithms for orchard targeted spraying, identifying challenges like high-precision data registration, providing a systematic technical route for precision agriculture [34].

These technologies collectively advance pesticide targeted delivery from theory to practice.

5. Conclusion and Future Directions

This review has examined the development of lignin-based aqueous coacervates for improving pesticide targeting and efficiency. Beginning with an analysis of pesticide utilization challenges and environmental concerns, it established the rationale for advanced delivery systems that combine lignin valorization with coacervate technology. The discussion systematically addressed the structural characteristics and modification approaches for major lignin types, compared construction methodologies across different coacervate categories, and analyzed multiple targeting mechanisms from environmental responsiveness to application technologies. Collectively, these insights demonstrate how lignin-based coacervates can address key limitations in conventional pesticide applications.

While significant advances have been made in developing lignin-based aqueous coacervates, their transition to industrial application remains hindered by several technical limitations. Future research should focus on innovating lignin sources through precise molecular engineering to obtain more uniform and reactive building blocks. The integration of artificial intelligence with multi-scale simulations will enable intelligent design of coacervates with tailored functionality and enhanced loading capacity. Further progress requires system-level integration that combines these novel delivery systems with precision application technologies like drones and sensors, creating comprehensive plant protection platforms. Finally, thorough life cycle assessments are essential to ensure both environmental compatibility and economic viability. Through coordinated multidisciplinary efforts, lignin-based aqueous coacervates are positioned to become transformative technologies for sustainable crop protection.

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