CONTROLLED/SLOW-RELEASE FERTILIZER COATING FROM POLYSACCHARIDES: A MINI REVIEW OF LIGNIN AS A REINFORCEMENT MATERIAL

LAPISAN PUPUK TERKENDALI/LEPAS LAMBAT DARI POLISAKARIDA: TINJAUAN MINI TENTANG LIGNIN SEBAGAI BAHAN PENGUAT

\mathbf{S} avira Astri Adriana $^{\mathrm{1)}}$, Farah Fahma $^{\mathrm{1})^*}$, Titi Candra Sunarti $^{\mathrm{11}}$, Anuraga Jayanegara $^{\mathrm{21}}$, Rini Purnawati $^{\mathrm{11}}$, **Lisman Suryanegara3) , RM Muhammad Nur Fauzan4)**

¹⁾Department of Agroindustrial Technology, Faculty of Agricultural Engineering and Technology, IPB University, Bogor 16680, Indonesia Email [: farah_fahma@apps.ipb.ac.id](mailto:farah_fahma@apps.ipb.ac.id)

2)Department of Nutrition and Feed Technology, Faculty of Animal Science, IPB University Bogor, 16680, Indonesia 3)Research Center for Biomass and Bioproduct, National Research and Innovation Agency, Bogor, Indonesia 4)Graduate School of Science and Technology, Agricultural Sciences, University of Tsukuba, Tsukuba, Ibaraki, Japan

Paper: Accepted June 18, 2024; Corrected July 25, 2024; Approved July 30, 2024

ABSTRAK

Controlled/slow-release fertilizers (CSRF) terus dikembangkan karena adanya kebutuhan untuk meningkatkan produktivitas pertanian dan mengurangi dampak ekonomi, sosial, dan lingkungan akibat pencucian pupuk konvensional ke alam. Selain itu, penggunaan polimer biodegradable sebagai bahan CSRF terus menjadi prioritas dalam pengembangan CSRF. Oleh karena itu, tinjauan ini mensintesis penelitian terkini tentang CSRF berbasis lignin dan polisakarida, dengan menekankan kompatibilitas dan kinerjanya dalam aplikasi pertanian. Lignin, biopolimer yang berlimpah dan terbarukan, dievaluasi efektivitasnya sebagai agen pelepasan lambat dalam CSRF. Penelitian telah menunjukkan potensi lignin untuk meningkatkan profil pelepasan nutrisi dan kelestarian lingkungan bila digunakan sendiri atau dikombinasikan dengan polimer lain. Polisakarida, yang dikenal karena biokompatibilitas dan biodegradabilitasnya, juga telah dieksplorasi. Memasukkan lignin ke dalam CSRF berbasis polisakarida telah disorot, khususnya dalam matriks pati, selulosa, kitosan, dan natrium alginat. Komposit ini menawarkan sifat mekanik yang lebih baik, pelepasan unsur hara yang terkontrol, dan peningkatan retensi air tanah. Tantangan dan arah masa depan terkait CSRF berbasis lignin dan polisakarida juga ditinjau. Temuan-temuan ini menggarisbawahi pentingnya pengembangan teknologi pupuk berkelanjutan untuk memenuhi kebutuhan pangan di masa depan sekaligus memitigasi dampak lingkungan.

Kata kunci: lignin, matriks komposit, pupuk terkontrol/pelepasan lambat, polisakarida, urea

ABSTRACT

Controlled/slow-release fertilizers (CSRF) continue to be developed because of the need to increase agricultural productivity and reduce the economic, social, and environmental impacts of conventional fertilizers leaching into nature. Additionally, the use of biodegradable polymers as CSRF materials continues to be a priority in CSRF development. Therefore, this review synthesizes the current research on lignin- and polysaccharide-based CSRFs and emphasize their compatibility and performance in agricultural applications. Lignin, an abundant and renewable biopolymer, was evaluated for its effectiveness as a slow-release agent in CSRF. Studies have demonstrated the potential of lignin to improve nutrient release profiles and environmental sustainability when used alone or in combination with other polymers. Polysaccharides, which are known for their biocompatibility and biodegradability, have also been explored. The incorporation of lignin into polysaccharide-based CSRFs has been highlighted, particularly in starch, cellulose, chitosan, and sodium alginate matrices. These composites offer improved mechanical properties, controlled nutrient release, and enhanced soil water retention. The challenges and future directions regarding lignin- and polysaccharide-based CSRF are also reviewed. These findings underscore the importance of developing sustainable fertilizer technologies to meet future food demands while mitigating environmental impacts.

Keywords: controlled/slow-rele ase fertilizer, composite matrix, lignin, polysaccharide, urea

INTRODUCTION

With the global population projected to rise to nine billion by 2050, future food demand is expected to increase significantly (Vermoesen *et al.,* 2024). Nitrogen fertilizer has proven effective in boosting crop yields over the past six decades (Tiwari and Pal, 2022). However, the misuse of conventional fertilizers has led to elevated nitrogen levels in the soil, resulting in environmental, societal, and economic issues due to nitrogen leaching (Zulfiqar *et al*., 2019). Environmental damage includes harmful algal blooms, eutrophication, and water contamination (Bibi *et al.,* 2016). Socially,

Jurnal Teknologi Industri Pertanian 34 (2): 193-203 193 **Coressponding Author*

contaminated water sources can cause health problems, including methemoglobinemia, in children (dos Santos *et al.,* 2024). Economically, the cost of mitigating fertilizer leaching damage is substantial (Norse and Ju, 2015). Sustainable and environmentally friendly fertilizer management practices are crucial for mitigating these impacts.

Controlled-release fertilizers (CSRF) release nutrients gradually over an extended period, in contrast to conventional fertilizers that release nutrients quickly. CSRFs effectively enhance crop total nitrogen (TUN) uptake and yield of crop by synchronizing nutrient release with crop demand, thereby reducing environmental nitrogen release (Zhang *et al.,* 2014). Lignin, a renewable and relatively hydrophobic substance, has considerable potential as a sustainable raw material because of its abundance and role as a structural component of lignocellulosic biomass (Maheshwari, 2018). It is cost-effective and sustainable, given its availability as a by-product of the paper industry and biomass refining (Chen *et al.,* 2020; Kumar *et al.,* 2023).

Studies have demonstrated the effectiveness of lignin as a reinforcement agent in composite matrices owing to its slow-release properties (Abbas *et al.,* 2022). Most natural biopolymers, such as polysaccharides, within plant cell walls, exhibit high hydrophilicity, whereas lignin is predominantly hydrophobic (Zhao *et al.,* 2016). Therefore, compared to other reinforcing biomaterials, lignin is a particularly promising solution for creating controlled slow-release fertilizers (CSRF). Lignin has been used as a biocoating material to improve the slow nutrient release profile of fertilizers, both alone and in combination with other polymers (Jiao *et al.,* 2018; dos Santos *et al.,* 2021; Elhassani *et al.,* 2023). It significantly extends the nitrogen release time and can also slow the release of phosphorus when used to coat phosphate fertilizers (Fertahi *et al.,* 2024). Additionally, lignin-incorporated chitosan-based coatings and lignin-clay nanohybrid composites have shown promise in extending the life of coated urea fertilizers and in controlling nitrogen release while increasing soil water retention (Elhassani *et al.,* 2023; Zhang *et al.,* 2020).

Owing to their long carbohydrate chains, polysaccharides are promising candidates for sustainable composites because of their non-toxic, biocompatible, and biodegradable properties (Shah *et al.,* 2022). However, the variability in natural fiber properties poses a challenge in developing highperformance natural fiber-reinforced composites (Saravanan and Ganesan, 2017). CSRFs made from a single type of natural film-forming coating polymer often exhibit both desirable and undesirable properties. Consequently, mixing biopolymers with biocomposite materials is an alternative approach to improve CSRF coatings (Firmanda *et al.,* 2024).

Given the extensive research on lignin and polysaccharide mixtures, a literature review focusing on the CSRFs from these materials is necessary, particularly regarding the interactions between lignin and different polysaccharides. This paper reviews the current knowledge and research on lignin and various polysaccharides as CSRF composites and analyze their compatibility, properties, and challenges to optimize CSRF performance.

MATERIAL AND METHODS

An exploratory literature review was conducted using articles from national and international journals, sourced from databases such as Scopus, Science Direct, PubMed, Google Scholar, and Research Gate. The keywords employed included controlled-release fertilizer (CRF), lignin, urea, material, composite matrix, slow-release fertilizer (SRF), and polysaccharides. The search spanned from August 2023 to May 2024. The collected articles facilitated a detailed discussion of CSRF, lignin as a reinforcement material in CSRF, and ligninpolysaccharide-based CSRF.

Previous studies have investigated the incorporation of lignins into polysaccharide composites for various applications, such as biodegradable materials and drug delivery systems. However, a significant gap remains in understanding their specific impact on the CSRF performance requirements. Existing studies have primarily focused on mechanical properties and general biodegradability, often neglecting how lignin incorporation affects nitrogen release kinetics, water retention, and hydrophobicity. A comprehensive examination of these factors could optimize ligninpolysaccharide interactions and enhance the CSRF efficiency, sustainability, and functionality.

This article provides an in-depth examination of CSRF and their types. Initially, the role of lignin in CSRF formulations was explored, followed by an analysis of the properties of various polysaccharides, including starch, cellulose, chitosan, and alginate. These polysaccharides serve as matrices in CSRF, with lignin acting as a reinforcement agent. The characteristics of each polysaccharide composite and their corresponding CSRF properties, such as hydrophobicity, water retention, and nitrogen release, offer a holistic understanding of their interactions and potential optimization.

RESULTS AND DISCUSSION

Controlled/Slow-Release Fertilizer (CSRF)

Slow-release fertilizers release nutrients gradually, aligning with the plant's nutritional needs and improving the application efficiency. The release mechanism is influenced by environmental factors such as temperature, pH, humidity, and microbial activity, as well as the characteristics of the composite material (Firmanda *et al.,* 2023). Slow-release and controlled-release fertilizers are often used

interchangeably but differ primarily in their nutrientrelease mechanisms. Slow-release fertilizers depend on natural processes, whereas controlled-release fertilizers use engineered solutions for precise nutrient release (Dave *et al*., 1999; Rajan *et al.,* 2021).

Commercially, two main methods are used to produce slow- and controlled-release fertilizers: encapsulation and hydrophobicity (Purnomo and Saputra, 2021). Encapsulation involves coating fertilizer granules with a low-solubility layer of organic and inorganic materials to regulate nutrient dissolution by slowing water penetration and dissolution rates. Techniques such as pan, rotary drum, and fluidized-bed coating have been employed to produce controlled-release urea, with the fluidizedbed process yielding high-quality, uniformly thick layers (Behin and Sadeghi, 2016; Tzika *et al.,* 2003). The hydrophobic method uses a solid material matrix mixed and molded with fertilizer powder, which is commonly used in underdeveloped countries (Purnomo and Saputra, 2021). This method involves chemically modified slow-release fertilizers, in which chemical reactions between nutrients and active groups in the matrix and filler regulate the nutrient release (Lu *et al.,* 2022).

Lignin as a Reinforcement Material in CSRF

Lignins are complex phenolic and organic polymers essential for plant structural tissues (Kai *et al.,* 2017; Ebrahimi *et al.,* 2024). Annually, approximately 1.65 million tons of commercial technical lignin is produced, with lignosulfonate comprising ~80%, kraft lignin ~15%, and hydrolyzed and soda lignin ~5% (Dessbesell *et al.,* 2020). The physical and chemical properties of lignin vary significantly based on the type, source, and isolation procedure and are influenced by factors such as molar mass, polydispersity, and multi-branched structure (Kai *et al.,* 2017; Ariyanta *et al.,* 2023). Technical lignin can exhibit different structures and impurities depending on the plant species and isolation method used (Ruwoldt *et al.,* 2024). Owing to its functional aromatic structure, which primarily includes hydroxyl and methoxy chemical groups on aromatic rings, lignin has significant potential applications as an additive, coating, fertilizer, and plant growth promoter (Ariyanta *et al.,* 2023). The increase in phenolic hydroxyl or sulfonated groups during the technical processing of lignin can enhance its reactivity and hydrophobicity, making it suitable for slow-release fertilizers (Chen *et al.,* 2020).

Lignin can serve as a coating agent for fertilizers by leveraging its excellent slow-release properties (Chen *et al.,* 2020). Lignin-based CSRF are categorized into physically impeded (coating/encapsulation) and chemically modified (crosslinking) types (Lu *et al.,* 2022). Modified lignin coatings on urea can inhibit surface biodegradability in soil, prolonging fertility by reducing the degradation of urinary enzymes (Majeed *et al.,* 2015; Lu *et al.,* 2022). However, the high heterogeneity, complex structure, and low reactivity of lignin can result in uneven coatings, large surface porosities, and cracks (Kumar *et al.,* 2023). Therefore, modifications such as surface functionalization or composites with other compounds are necessary to utilize lignin effectively as a CSRF coating (Mulder *et al.,* 2011).

Lignin is widely used as a reinforcement material in composite matrices for CSRF production. Common composite matrices include polyvinyl alcohol (PVA) (dos Santos *et al*., 2021; Liu *et al.,* 2024), polysaccharides (Elhassani *et al.,* 2023; Latha *et al.,* 2023), rubber (Boonying *et al.,* 2023), and clays (Zhang *et al.,* 2020). The efficiency of lignin-coated urea CSRF depends on the coating's percentage and composition (García *et al.,* 1998). Although significant progress has been made in developing lignin-based CSRF, future efforts should focus on applying these advancements to practical agricultural production.

Lignin-Polysaccharide Based CSRF

Polysaccharides are complex carbohydrates composed of long chains of monosaccharide units that are linked by glycosidic bonds. Polysaccharides are widely distributed in nature, found in animals, plants, and microorganisms, and exhibit diverse functional properties (Chen and Huang, 2017). Different types of polysaccharides are classified based on their monosaccharide compositions and structural characteristics. The sources of polysaccharides vary greatly, ranging from plant materials, such as agricultural and forestry waste (Mohammed *et al.,* 2021) to microbial sources, such as fungi, bacteria, and microalgae (Galasso *et al.,* 2019). Polysaccharides have wide applications in various industries, including pharmaceutical, cosmetic, food, and paper production (Ullah *et al.,* 2021). Polysaccharides have also received significant attention in sustainable agriculture because of their potential as controlled-release fertilizers. This complex carbohydrate exhibits a series of characteristics that make it suitable for these applications, including its ability to slowly release nutrients over time, biodegradability, and its potential to improve soil health (Nechita and Iana-Roman, 2020). Therefore, polysaccharides are an environmentally friendly alternative to CSRF.

However, unmodified polysaccharide-based CSRF have several weaknesses, including hydrophilicity and relatively low mechanical strength, which affect their performance under various soil moisture conditions (Chiaregato *et al.,* 2022). Therefore, chemical modification of polysaccharides has emerged as a promising approach. An example of this chemical modification is the crosslinking of starch by reaction with sodium tetraborate, which can enhance its viscosity and solubility because pure starch becomes a highly branched long-chain biopolymer (Naz *et al.,* 2014).

In addition to chemical modification, the incorporation of reinforcing agents, such as lignin, into polysaccharide-based CSRF offers further improvements (Chiaregato *et al.,* 2022). Lignin and polysaccharide-based CSRFs (Figure 1) have emerged as promising solutions that offer many benefits over conventional fertilizers (Gu *et al.,* 2014), as shown in Table 1. When incorporated into fertilizers, lignin and polysaccharides can act as slowrelease agents that gradually release nutrients such as nitrogen over time to offset plant uptake and increase wettability or the ability of the soil to absorb water, which is critical for effective plant nutrient uptake. This advantage aligns with increasing demand for sustainable and environmentally responsible agricultural practices.

Lignin-Starch Based CSRF

Starch is a polysaccharide produced by plants for energy storage and is primarily composed of two D-glucose polymers: lightly branched amylose, with long glucan chains, and highly branched amylopectin, which contains many shortchain groups (Wang *et al.,* 2015). Most commercially available starches are derived from grains, such as corn, rice, and wheat, or tubers such as potatoes and cassava (tapioca) (Jiang *et al.,* 2020).

Starch shows potential for use in CSRF composites owing to its several key properties. It has excellent film-forming capabilities, which allow it to adhere to fertilizer granules and form a continuous barrier that controls nutrient release, providing a gradual nutrient supply to plants (Lin *et al.,* 2024). Additionally, starch can retain water, enhancing the ability of coated fertilizer granules to reduce nutrient loss through leaching (Cai *et al.,* 2014). Its costeffectiveness and availability make it an attractive option for large-scale slow-release fertilizer production (Jiang *et al.,* 2020).

However, natural starch has limitations, such as suboptimal mechanical and thermophysical properties, reduced suitability for urea coatings, and sensitivity to humidity (Chen *et al*., 2020; Jiang *et al.,* 2020). The uneven layer thickness and high biodegradation rate can affect the stability and commercialization of slow-release fertilizers (SRF) derived from starch (Channab *et al*., 2023; Majeed *et al.,* 2018). The addition of lignin as a filler to the starch matrix can enhance the functional and mechanical properties of CSRF. Lignin's incorporation can improve starch biodegradation and nitrogen release properties (Majeed *et al.,* 2013). Nanotechnology can further enhance the performance of lignin and starch coatings in slow-release fertilizers, making them biodegradable and environmentally friendly (Chen *et al.,* 2020). The role of lignin in protecting starch morphology has been confirmed through optical microscopy, which showed fewer changes in the starch particle structure (Majeed *et al.,* 2018).

Research has demonstrated that lignin in the starch matrix positively affects nitrogen release, water absorption, biodegradability, and plant growth in CSRF. The urea release efficiency of CSRF is improved by chemical crosslinking and hydrogen bondings between alkaline lignin (AL), polyvinyl alcohol (PVA), and corn starch (CS), as well as the aggregation properties of AL (Yang *et al.,* 2023). Lignin-modified tapioca starch films can function as effective soil conditioners and slow-release systems (Man *et al.,* 2014). Increasing the lignin content enhances the hydrophobicity of urea-cross-linked starch-lignin composite films, reducing water absorption (Sarwono *et al.,* 2018; Ariyanti *et al.,* 2013). The slow mineralization in these films indicates that lignin controls the biodegradability of starch (Majeed *et al.,* 2016). Rapid plant growth trials have shown that CSRF from composites of AL, PVA, and CS can significantly reduce fertilizer application rates by up to 70% (Yang *et al.,* 2023).

Figure 1. Illustration of lignin-polysaccharide based CSRF with physically impeded method (coating) (a) and chemically modified method (crosslinking) (b).

Lignin-Cellulose Based CSRF

Cellulose is a commercially important polysaccharide. Cellulose is the most abundant, water-insoluble, bio-derived, fibrous polymer (Ranjha *et al.,* 2023). Cellulose is in growing demand because of its renewability, non-toxicity, economic value, biodegradability, exceptional mechanical properties, high surface area, and biocompatibility (Chen *et al.,* 2023).

Cellulose is an excellent film-forming material as it can form hydrocolloids in suitable solvent systems (Khalil *et al.*, 2017). Currently, these composites are used in most engineering fields because they strengthen the polymer matrix, as

demonstrated by mechanical characterization (Chen *et al.,* 2023).

The use of cellulose in slow-release fertilizer (SRF) coatings offers several advantages. Cellulose is hydrophilic, meaning that it absorbs and retains water (El Bouchtaoui *et al.,* 2022b). This property enables the gradual release of water into the fertilizer core, facilitating the dissolution and subsequent release of nutrients. Its high hydrophilicity helps to maintain soil moisture and promote plant growth (Firmanda *et al.,* 2022). Additionally, the large surface area and high porosity of cellulose enhance its ability to absorb and retain nutrients (Voon *et al.,* 2016). Cellulose is also resistant to chemical degradation, providing long-term protection to the fertilizer core (Rinaldi and Schüth, 2009).

Methylcellulose (MC) is a significant cellulose derivative formed by replacing hydroxyl groups with methoxy groups (Viera *et al.,* 2007). However, because of its hydrophilicity, methylcellulose alone has a limited barrier capacity, making it less suitable for slow-release systems (El Bouchtaoui *et al.,* 2022b). To address this, methylcellulose must be combined with other biopolymers. Incorporating lignin as a filler with cellulose creates a lignin-cellulose layer for the SRF, forming a robust, long-lasting barrier that controls nutrient release (El Bouchtaoui *et al.,* 2022b).

Several studies have examined the effects of lignin in the cellulose matrix on CSRF, including nitrogen release, water absorption, biodegradability, and plant growth. The methylcellulose/lignin (MC/LGe) coating layer effectively delayed fertilizer release for over 30 days (El Bouchtaoui *et al.,* 2022b). Urea fertilizer coated with lignocellulose from sugarcane bagasse showed slower nitrogen release than pure urea fertilizer, even after 60 days (Elhassani *et al.,* 2019).

The MC/LGe film layer also increased the mechanical resistance of the fertilizer, thereby enhancing the water retention capacity of the soil. This is evidenced by the increased contact angle of the MC/LGe composite film with a higher lignin content, indicating improved wettability of the outer surface (El Bouchtaoui *et al*., 2022b). This methylcellulose/lignin-based CSRF fertilizer significantly increased wheat leaf area, chlorophyll content, biomass, root architecture, and fertilization efficiency (El Bouchtaoui *et al.,* 2022b). Improved soil structure due to the addition of CSRF with a methylcellulose/lignin-based coating benefits air circulation and nutrient absorption (Li *et al.,* 2023).

Lignin-Chitosan Based CSRF

Chitosan (CS) is a biodegradable and biocompatible polysaccharide derived from fungi, crustaceans, and insects (Román-Doval *et al.,* 2023). As a highly deacetylated derivative of chitin, chitosan is a natural polymer that has significant agricultural applications (Wu and Liu, 2008). Its biocompatibility, biodegradability, non-toxicity, and drug adsorption ability make chitosan an effective carrier for the controlled delivery of agrochemicals and genetic materials in agriculture (Virmani *et al.,* 2023).

Chitosan enhances root growth and exhibits antimicrobial, antifungal, and antiviral properties, thus benefiting horticultural plants (Román-Doval *et al*., 2023). As a coating material for slow-release fertilizers, chitosan offers advantages such as biodegradability, biocompatibility, and non-toxicity. Its amino groups allow chitosan to form complexes with other compounds, although it is difficult to dissolve in water or acidic solutions (Riseh *et al.,* 2023). Chemical modification of the reactive amino groups of chitosan can improve its physicochemical properties, enhancing its applicability in various agricultural contexts (Virmani *et al.,* 2023).

The polymer compound of chitosan can bond with other natural polymers, enhancing fertilizer effectiveness by meeting plant nutrient needs and improving soil texture (Riseh *et al.,* 2023). The combination of lignin and chitosan in fertilizer coatings offers several benefits. This combination improves the hydrophobic-hydrophilic balance, reduces water vapor permeability, and enhances nutrient release efficiency (Wu and Liu, 2008). Both lignin and chitosan exhibit chelating properties, enabling them to bind and release nutrients in a controlled manner, thereby preventing nutrient loss and ensuring efficient uptake by plants (Chen *et al.,* 2020).

Furthermore, lignin- and chitosan-based coatings provide additional stability against urease hydrolysis and nitrification by microorganisms after fertilizer is applied to the soil. This results in improved nutrient utilization, reduced environmental impact, and increased crop yield. Research on a CSRF fertilizer formulation with chitosan, polyvinyl alcohol (PVA), and 3% lignin demonstrated favorable physicochemical properties, including a balanced hydrophobic-hydrophilic nature, enhanced elasticity tolerance, and reduced water vapor permeability (Elhassani *et al.,* 2023). Studies on chitosan-lignin nanocomposite fertilizers have indicated a slow release of 30−35% total nitrogen over 15 days in the soil (Latha *et al.,* 2023). Urea granules were coated with a chitosan-PVA-lignin formulation using controlled spraying and drying conditions to achieve optimal adhesion between the fertilizer and coating (Elhassani *et al*., 2023). This study demonstrated that lignin and chitosan effectively serve as molds for retaining and releasing nitrogen in a controlled manner, thereby improving the nitrogen use efficiency (Latha *et al.,* 2023).

Lignin-Alginate Based CSRF

Alginate is a salt of alginic acid and a naturally occurring hydrophilic anionic polysaccharide (Taib *et al.,* 2022). With a relative molecular mass of approximately 106, alginate is a long-chain polymer

consisting of (1-4) cross-linked D-mannuronic acid and (1-4) cross-linked guluronic acid (Taib *et al.,* 2022).

Alginate is extracted from brown seaweed and has been widely used in various applications, including food, pharmaceutical, and biomedical fields (El Bouchtaoui *et al.,* 2022a). Sodium alginate has outstanding features, such as high biocompatibility, biodegradability, renewability, and abundant hydroxyl and carboxyl groups with high adsorption affinity towards heavy metal ions (Gao *et al.,* 2020). These characteristics make it an ideal coating material for slow-release fertilizers. However, the mechanical strength, stability, and heat resistance of sodium alginate are relatively low (Gao *et al.,* 2017).

These studies have shown that the ligninalginate layer creates a uniform and compact polymer layer on the fertilizer surface, correcting any irregularities and ensuring controlled release of nutrients over a long period (El Bouchtaoui *et al.,* 2022a). Additionally, increasing the lignin content in the CSRF coating formulation improved the slowrelease behavior, resulting in a release period of over one month in the soil compared to only four days for the uncoated fertilizer (El Bouchtaoui *et al.,* 2022a). This research was also supported by the release rate of a controlled-release formulation (CRF), which can be controlled by mixing activated carbon in alginatebased CRF with kraft lignin (Fernández-Pérez *et al.,* 2011). In addition, biodegradability was also proven by the new green hydrogel synthesized by connecting lignosulfonate, sodium alginate, and konjac flour, which has good degradability and can be degraded by 20% when buried in soil for six months (Song *et al.,* 2020).

Future Challenges

The development of lignin and polysaccharide-based coatings for controlled-release fertilizers (CSRF) is a promising area of research because of the favorable physical and chemical properties, biodegradability, renewability, and low cost of these materials. However, several challenges must be addressed to fully realize their potential. Polysaccharides, for instance, exhibit high hydrophilicity, which can affect the performance of coatings. Additionally, lignin dispersion within the polysaccharide matrix is problematic, necessitating further investigation to identify the most economically feasible polysaccharide materials for CSRF production. The complex and expensive process of modifying lignin and the inherent mechanical and thermophysical limitations of natural polymers such as starch further complicate the development of effective coatings (Chen *et al.,* 2020). Research efforts must also focus on optimizing the timing of nutrients release to match plant growth requirements. For lignin-cellulose and lignin-alginate coatings, achieving optimal layer thickness, homogeneity, and stability is essential to ensure controlled nutrient release and prevent issues such as cracking or peeling (El Bouchtaoui *et al.,* 2022a; El Bouchtaoui *et al.,* 2022b). For lignin and chitosanbased coatings, standardization and evaluation methods are crucial for improving the preparation process and overall performance of fertilizers (Boarino and Klok, 2023). Establishing uniform evaluation protocols and emphasizing CSRF testing in agricultural applications will provide valuable data on fertilizer efficiency, potentially reducing reliance on conventional fertilizers. Overall, the continued exploration and development of lignin- and polysaccharide-based coatings hold significant promise for advancing sustainable agricultural practices.

CONCLUSIONS AND RECOMMENDATION

Conclusions

The increasing global population necessitates innovative solutions to sustainably meet future food demands. The overuse of conventional nitrogen fertilizers has led to environmental, social, and economic challenges due to nitrogen leaching. CSRF particularly those incorporating lignin and polysaccharides, are promising solution to these issues. Lignin, a renewable and abundant biopolymer, has demonstrated significant potential as a reinforcing agent in CSRFs, owing to its slow-release properties and environmental benefits. Polysaccharides such as starch, cellulose, chitosan, and sodium alginate contribute to the sustainability and efficiency of CSRFs by improving their nutrient release profiles, water retention, and biodegradability. Combining lignin with these polysaccharides enhances the functional properties of CSRFs, making them more effective and environmentally friendly. However, challenges such as high polysaccharide hydrophilicity, lignin dispersion issues, and complex, costly modification processes must be addressed to optimize the timing of nutrient release and ensure coating stability and effectiveness. This review highlights the need for continued research on lignin-polysaccharide interactions to optimize the performance of CSRFs, ultimately contributing to sustainable agricultural practices and food security for the growing global population.

Recommendation

Future research should focus on addressing the challenges identified in this review to advance the development and application of CSRF incorporating lignin and polysaccharides. Innovative approaches to enhance the hydrophilicity of polysaccharides and improve lignin dispersion within polymer matrices are critical. Additionally, research on cost-effective and scalable polysaccharide modification techniques is essential to make CSRFs viable for widespread agricultural adoption.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the financial support provided by the *Direktorat Jenderal Pendidikan Tinggi, Riset, dan Teknologi, Kementerian Pendidikan, Kebudayaan, Riset, dan Teknologi,* the Republic of Indonesia. This research was funded under the Fundamental Research Regular scheme, contract number 027/E5/PG.02.00.PL/2024, dated June 11, 2024.

REFERENCES

- Abbas A, Wang Z, Zhang Y, Peng P, She D. 2022. Lignin-based controlled release fertilizers: A review. *International Journal of Biological Macromolecules*. 222 Pt B:1801–1817.
- Ariyanta HA, Sari FP, Sohail A, Restu WK, Septiyanti M, Aryana N, Fatriasari W, Kumar A. 2023. Current roles of lignin for the agroindustry: Applications, challenges, and opportunities. *International Journal of Biological Macromolecules*. 240:124523.
- Behin J and Sadeghi N. 2016. Utilization of waste lignin to prepare controlled-slow release urea. *International Journal of Recycling of Organic Waste in Agriculture*. 5(4):289–299.
- Bibi S, Saifullah, Naeem A, Dahlawi S. 2016. Environmental impacts of nitrogen use in agriculture, nitrate leaching and mitigation strategies. In Hakeem, K., Akhtar, J., Sabir, M., editor. *Soil science: agricultural and environmental prospectives*. Cham: Springer.
- Boarino A and Klok HA. 2023. Opportunities and challenges for lignin valorization in food packaging, antimicrobial, and agricultural applications. *Biomacromolecules*. 24(3):1065–1077.
- Boonying P, Boonpavanitchakul K, Amnuaypanich S, Kangwansupamonkon W. 2023. Natural rubber-lignin composites modified with natural rubber-graft-polyacrylamide as an effective coating for slow-release fertilizers. *Industrial Crops and Products Journal*. 191: 116018.
- El Bouchtaoui FZ, Ablouh EH, Kassem I, Kassab Z, Sehaqui H, El Achaby M. 2022a. Slow-release fertilizers based on lignin–sodium alginate biopolymeric blend for sustained N–P nutrients release. *Journal of Coatings Technology and Research*. 19(5):1551–1565.
- El Bouchtaoui FZ, Ablouh EH, Mhada M, Kassem I, Salim MH, Mouhib S, Kassab Z, Sehaqui H, El Achaby M. 2022b. Methylcellulose/lignin biocomposite as an eco-friendly and multifunctional coating material for slowrelease fertilizers: Effect on nutrients management and wheat growth. *International Journal of Biological Macromolecules*. 221:398–415.
- Cai J, Cai C, Man J, Zhou W, Wei C. 2014. Structural and functional properties of C-type starches. *Carbohydrate Polymers*. 101:289–300.
- Channab B, El Idrissi A, and Zahouily M, Essamlali Y, White JC. 2023. Starch-based controlled release fertilizers: A review. *International Journal of Biological Macromolecules*. 238(124075):124075.
- Chen J, Fan X, Zhang L, Chen X, Sun S, Sun R-C. 2020. Research progress in lignin-based slow/controlled release fertilizer. *ChemSusChem*. 13(17):4356–4366.
- Chen L and Huang G. 2017. Antitumor Activity of Polysaccharides: An Overview. *Curr Drug Targets*. 19(1). 89-96.
- Chen Z, Aziz T, Sun H, Ullah A, Ali A, Cheng L, Ullah R, Khan FU. 2023. Advances and applications of cellulose bio-composites in biodegradable materials. *Journal of Polymers and the Environment*. 31: 2273–2284.
- Chiaregato CG, França D, Messa LL, dos Santos Pereira T, Faez R. 2022. A review of advances over 20 years on polysaccharide-based polymers applied as enhanced efficiency fertilizers. *Carbohydrate Polymers.* 279:119014.
- Dave AM, Mehta MH, Aminabhavi TM, Kulkarni AR, Soppimath KS. 1999. A review on controlled release of nitrogen fertilizers through polymeric membrane devices. *Polymer-Plastics Technology and Engineering*. 38(4), 675–711.
- Dessbesell L, Paleologou M, Leitch M, Pulkki R, Xu C (Charles). 2020. Global lignin supply overview and kraft lignin potential as an alternative for petroleum-based polymers. *Renewable and Sustainable Energy Reviews.* 123:109768.
- Ebrahimi M, Acha V, Hoang L, Martínez-Abad A, López-Rubio A, Rhazi L, Aussenac T. 2024. Extraction of homogeneous lignin oligomers by ozonation of Miscanthus giganteus and vine shoots in a pilot scale reactor. *Bioresource Technology.* 402:130804.
- Elhassani CE, Essamlali Y, Aqlil M, Nzenguet AM, Ganetri I, Zahouily M. 2019. Ureaimpregnated HAP encapsulated by lignocellulosic biomass-extruded composites: A novel slow-release fertilizer. *Environmental Technology & Innovation Journal.* 15: 100403.
- Elhassani CE, El Gharrak A, Essamlali Y, Elamiri S, Dânoun K, Aboulhrouz S, Zahouily M. 2023. Lignin nanoparticles filled chitosan/polyvinyl alcohol polymer blend as a coating material of urea with a slow-release property. *Journal of Applied Polymer Science*. 140(16): e53755.
- Fernández-Pérez M, Garrido-Herrera FJ, González-Pradas E. 2011. Alginate and lignin-based formulations to control pesticides leaching in

a calcareous soil. *Journal of Hazardous Materials.* 190(1):794–801.

- Fertahi S, Bertrand I, Amjoud M, Oukarroum A, Arji M, Barakat A. 2019. Properties of coated slow-release triple superphosphate (TSP) fertilizers based on lignin and carrageenan formulations. *ACS Sustainable Chemistry & Engineering Journal*. 7(12):10371–10382.
- Fertahi S, Bertrand I, Ilsouk M, Oukarroum A, Amjoud M, Zeroual Y, Barakat A. 2020. Impact of plasticizers on lignin-carrageenan formulation properties and on phosphorus release from a coated triple superphosphate fertilizer. *Industrial & Engineering Chemistry Research*. 59(31):14172–14179.
- Fertahi S, Elhaissoufi W, Bargaz A, Touchaleaume F, Habibi Y, Oukarroum A, Zeroual Y, Barakat A. 2024. Lignin-rich extracts as slow-release coating for phosphorus fertilizers. *Progress in Organic Coatings Journal.* 190:108394.
- Firmanda A, Fahma F, Syamsu K, Mahardika M, Suryanegara L, Munif A, Gozan M, Wood K, Hidayat R, Yulia D. 2024. Biopolymer-based slow/controlled-release fertilizer (SRF/CRF): Nutrient release mechanism and agricultural sustainability. *Journal of Polymers and the Environment* 12(2):112177.
- Firmanda A, Fahma F, Syamsu K, Sari YW, Suryanegara L, Wood K, Saito Y. 2023. Factors influencing the biodegradability of agro-biopolymer based slow or controlled release fertilizer. *Journal of Polymers and the Environment*. 31(5):1706–1724.
- Firmanda A, Fahma F, Syamsu K, Suryanegara L, Wood K. 2022. Controlled/slow-release fertilizer based on cellulose composite and its impact on sustainable agriculture: review. *Biofuels, Bioproducts and Biorefining.* 16(6):1909–1930.
- Galasso C, Gentile A, Orefice I, Ianora A, Bruno A, Noonan DM, Sansone C, Albini A, Brunet C. 2019. Microalgal derivatives as potential nutraceutical and food supplements for human health: A focus on cancer prevention and interception. *Nutrients.* 11(6): 1226.
- Gao X, Guo C, Hao J, Zhao Z, Long H, Li M. 2020. Adsorption of heavy metal ions by sodium alginate based adsorbent-a review and new perspectives. *International Journal of Biological Macromolecules*. 164:4423–4434.
- Gao X, Zhang Y, and Zhao Y. 2017. Biosorption and reduction of Au (III) to gold nanoparticles by thiourea modified alginate. *Carbohydrate Polymers*. 159:108–115.
- García C, García L, Vallejo A, Cartagena MC, Díez JA. 1998. Forecasting by laboratory tests of nitrogen leached and absorbed in soil‐plant system with urea‐based controlled‐release fertilizers coated with lignin. *Commun*

Communications in Soil Science and Plant Analysis. 29(15–16):2479–2491.

- Gu N, Zhao LP, Zhao XM. 2014. A review and perspective on slow and controlled release fertilizer in China. *Applied Mechanics and Materials*. 535:222–225.
- Jiang T, Duan Q, Zhu J, Liu H, Yu L. 2020. Starchbased biodegradable materials: challenges and opportunities. *Advanced Industrial and Engineering Polymer Research*. 3(1):8–18.
- Jiao G-J, Xu Q, Cao S-L, Pai P, She D. 2018. Controlled-release fertilizer with lignin used to trap urea/hydroxymethylurea/ ureaformaldehyde polymers. *Bioresources*. 13(1): 1711-1728.
- Kai D, Chow LP, Loh XJ. 2017. Lignin and its properties. In *Functional materials from lignin.* Volume 3. Singapore: World Scientific.
- Khalil HPSA, Tye YY, Saurabh CK, Leh CP, Lai TK, Chong EWN, Nurul Fazita MR, Mohd Hafiidz J, Banerjee A, Syakir MI. 2017. Biodegradable polymer films from seaweed polysaccharides: A review on cellulose as a reinforcement material. *Express Polymer Letters*. 11(4):244–265.
- Kumar R, Næss G, and Sørensen M. 2023a. Slowrelease fertilizers using lignin: Challenges and future prospects. *Biofuels Bioprod Biorefin*. 17(5):1368–1381.
- Latha M, Subramanian KS, Sundara Sharmila DJ, Raja K, Rajkishore SK, Chitdeshwari T. 2023. Urea-lignin/chitosan nanocomposite as slowrelease nanofertilizer. *ACS Agricultural Science & Technology Journal.* 3(6):463–476.
- Li H, Wang N, Zhang Li, Wei Y, Zhang Long, Ma Y, Ruso JM, Liu Z. 2023. Engineering and slowrelease properties of lignin-based double-layer coated fertilizer. *Polymers for Advanced Technologies.* 34(6):2029–2043.
- Lin Z, Cheng H, He K, McClements DJ, Jin Z, Xu Z, Meng M, Peng X, Chen L. 2024. Recent progress in the hydrophobic modification of starch-based films. *Food Hydrocolloids Journal*. 151:109860.
- Liu Y, Cao L, Wang L, Qi Y, Zhao Y, Lu H, Lu L, Zhang D, Wang Z, Zhang H. 2024. Preparation and application of degradable lignin/poly (vinyl alcohol) polymers as urea slow-release coating materials*. Molecules*. 29(8): 1699.
- Lu J, Cheng M, Zhao C, Li B, Peng H, Zhang Y, Shao Q, Hassan M. 2022. Application of lignin in preparation of slow-release fertilizer: Current status and future perspectives. *Industrial Crops and Products Journal.* 176: 114267.
- Maheshwari NV. 2018. Agro-industrial Lignocellulosic Waste: An Alternative to Unravel the Future Bioenergy. In Kumar A, Ogita S, Yau Y-Y, editor. *Biofuels:*

Greenhouse Gas Mitigation and Global Warming: Next Generation Biofuels and Role of Biotechnology. New Delhi: Springer India.

- Majeed Z, Mansor N, Ajab Z, Man Z, Sarwono A. 2018. Kraft lignin ameliorates degradation resistance of starch in urea delivery biocomposites*. Polymer Testing Journal*. 65:398–406.
- Majeed Z, Mansor N, Man Z, Wahid SA. 2016. Lignin reinforcement of urea-crosslinked starch films for reduction of starch biodegradability to improve slow nitrogen release properties under natural aerobic soil condition. *E-Polymers*. 16(2):159–170.
- Majeed Z, Ramli NK, Binti Mansor N, Man Z. 2013. Lignin loading effect on biodegradability and nitrogen release properties of urea modified tapioca starch in wet soil. *Key Engineering Materials.* 594–595:798–802.
- Majeed Z, Ramli NK, Mansor N, Man Z. 2015. Lignin modified urea fertilizer's biodegradation and nitrogen release under reduced soil condition. *Applied Mechanics and Materials.* 699:981– 987.
- Man Z, Sarwono A, Bustam MA, Azizli KAM. 2014. Starch based soil conditioner and slow release system. In *Applied Mechanics and Materials*. Volume 625. Bäch: Trans Tech Publications Ltd.
- Mohammed ASA, Naveed M, Jost N. 2021. Polysaccharides; classification, chemical properties, and future perspective applications in fields of pharmacology and biological medicine (a review of current applications and upcoming potentialities). *Journal of Polymers and the Environment.* 29(8):2359–2371
- Mulder WJ, Gosselink RJA, Vingerhoeds MH, Harmsen PFH, Eastham D. 2011. Lignin based controlled release coatings*. Industrial Crops and Products*. 34(1):915–920.
- Naz MY, Sulaiman SA, Ariwahjoedi B, Shaari KZK. 2014. Characterization of Modified Tapioca Starch Solutions and Their Sprays for High Temperature Coating Applications. *The Scientific World Journal.* 2014(1):375206.
- Nechita P and Iana-Roman MR. 2020. Review on polysaccharides used in coatings for food packaging papers. *Coatings*. 10(6): 566.
- Norse D and Ju X. 2015. Environmental costs of China's food security. *Agriculture, Ecosystems & Environment*. 209:5–14.
- Purnomo CW, Saputra H. 2021. Chapter 6 Manufacturing of slow and controlled release fertilizer. In Lewu FB, Volova T, Thomas S, K.R. R, editor. *Controlled release fertilizers for sustainable agriculture*. Cambridge: Academic Press.
- Rajan M, Shahena S, Chandran V, Mathew L. 2021. Chapter 3 - Controlled release of fertilizers concept, reality, and mechanism. In Lewu FB,

Volova T, Thomas S, K.R. R, editor. *Controlled release fertilizers for sustainable agriculture*. Cambridge: Academic Press.

- Ranjha MMAN, Shafique B, and Aadil RM, Manzoor MF, Cheng J-H. 2023. Modification in cellulose films through ascent cold plasma treatment and polymerization for food products packaging. *Trends in Food Science & Technology.* 134:162–176.
- Rinaldi R and Schüth F. 2009. Acid hydrolysis of cellulose as the entry point into biorefinery schemes. *ChemSusChem*. 2(12):1096–1107.
- Riseh RS, Vazvani MG, and Kennedy JF. 2023. The application of chitosan as a carrier for fertilizer: A review. *International Journal of Biological Macromolecules*. 252 (126483): 126483.
- Román-Doval R, Torres-Arellanes SP, Tenorio-Barajas AY, Gómez-Sánchez A, Valencia-Lazcano AA. 2023. Chitosan: Properties and its application in agriculture in context of molecular weight. *Polymers (Basel).* 15(13):2867.
- Ruwoldt J, Syverud K, and Tanase-Opedal M. 2024. Purification of soda lignin. *Sustain Chem Env 6.* 100102.
- dos Santos ACS, Henrique HM, Cardoso VL, Reis MHM. 2021. Slow release fertilizer prepared with lignin and poly(vinyl acetate) bioblend. *International Journal of Biological Macromolecules*. 185:543–550.
- dos Santos RC, da Silva RA, dos Santos MM, Bovo AB, da Silva AF. 2024. Assessing nitrate contamination in groundwater for public supply: A study in a small Brazilian town. *Groundwater for Sustainable Development.* 25:101084.
- Saravanan S and Ganesan K. 2017. Mechanical testing of epoxy bonded eco friendly natural fibre composite material. *International Journal of Computer Aided Engineering and Technology*. 9(2):241–250.
- Sarwono A, Man Z, Bustam MA, Subbarao D, Idris A, Muhammad N, Khan AS, Ullah Z. 2018. Swelling mechanism of urea cross-linked starch–lignin films in water. *Environmental Technology.* 39(12):1522–1532.
- Sarwono A, Man Z, and Bustam MA. 2013. Improvement of hydrophobicity of urea modified tapioca starch film with lignin for slow release fertilizer. *Advanced Materials Research*. 626(350–354).
- Shah N, Khan WA, Rehan T, Lin D, Tetik H, Haider S. 2022. 12 - Polysaccharides-metal oxide composite: A green functional material. In Haider S, Haider A, editor. *Renewable polymers and polymer-metal oxide composites.* Amsterdam: Elsevier.
- Song B, Liang H, Sun R, Peng P, Jiang Y, She D. 2020. Hydrogel synthesis based on

lignin/sodium alginate and application in agriculture. *International Journal of Biological Macromolecules*. 144:219–230.

- Taib MNAM, Antov P, Savov V, Fatriasari W, Madyaratri EW, Wirawan R, Osvaldová LM, Hua LS, Ghani MAA, Edrus SSAO Al, *et al.,* 2022. Current progress of biopolymer-based flame retardant. *Polymer Degradation and Stability*. 205: 110153.
- Tiwari AK and Pal DB. 2022. Chapter 11 Nutrients contamination and eutrophication in the river ecosystem. In S. Madhav, S. Kanhaiya, A. Srivastav, V. Singh, & P. Singh, editor. *Ecological Significance of River Ecosystems*. Amsterdam: Elsevier.
- Tzika M, Alexandridou S, and Kiparissides C. 2003. Evaluation of the morphological and release characteristics of coated fertilizer granules produced in a Wurster fluidized bed. *Powder Technology.* 132(1):16–24.
- Ullah A, Rahman L, Yazdani MB, Irfan M, Khan WS, Rehman A. 2021. Cell wall polysaccharides. In Inamuddin, M.I. Ahamed, R. Boddula and T. Altalhi, editor. *Polysaccharides*. Hoboken: John Wiley & Sons, Inc.
- Vermoesen E, Bodé S, Brosens G, Boeckx P, Van Vlierberghe S. 2024. Chemical strategies towards controlled release in agriculture. *Reviews in Chemical Engineering*. 40(2):247– 277.
- Viera RGP, Filho GR, de Assunção RMN, S. Meireles C da, Vieira JG, de Oliveira GS. 2007. Synthesis and characterization of methylcellulose from sugar cane bagasse cellulose. *Carbohydrate Polymers.* 67(2):182–189.
- Virmani T, Kumar G, Sharma A, Pathak K, Akhtar MS, Afzal O, Altamimi ASA. 2023. Amelioration of cancer employing chitosan, its derivatives, and chitosan-based

nanoparticles: Recent updates. *Polymers*. 15(13): 2928.

- Voon LK, Pang SC, and Chin SF. 2016. Highly porous cellulose beads of controllable sizes derived from regenerated cellulose of printed paper wastes. *Materials Letters.* 164:264–266.
- Wang Shujun, Li C, Copeland L, Niu Q, Wang Shuo. 2015. Starch retrogradation: A comprehensive review. *Comprehensive Reviews in Food Science and Food Safety.* 14(5): 568–585.
- Wu L and Liu M. 2008. Preparation and properties of chitosan-coated NPK compound fertilizer with controlled-release and water-retention. *Carbohydrate Polymers*. 72(2): 240–247.
- Yang Z, Su W, Fang J, Qian Y, Li H. 2023. A degradable mulch film with fertilizer slowrelease function enhanced by lignin. *ACS Applied Polymer Materials Journal*. 5(9): 6864–6874.
- Zhang S, Fu X, Tong Z, Liu G, Meng S, Yang Y, Helal MID, Li YC. 2020. Lignin-clay nanohybrid biocomposite-based double-layer coating materials for controllable-release fertilizer. *ACS Sustainable Chemistry & Engineering Journal.* 8(51): 18957–18965.
- Zhang Y, Liang X, Yang X, Liu H, Yao J. 2014. An eco-friendly slow-release urea fertilizer based on waste mulberry branches for potential agriculture and horticulture applications. *ACS Sustainable Chemistry & Engineering Journal.* 2(7): 1871-1878.
- Zhao C, Ma Z, Shao Q, Li B, Ye J, Peng H. 2016. Enzymatic hydrolysis and physiochemical characterization of corn leaf after H-AFEX pretreatment. *Energy Fuels*. 30(2): 1154-1161.
- Zulfiqar F, Navarro M, Ashraf M, Akram NA, Munné-Bosch S. 2019. Nanofertilizer use for sustainable agriculture: Advantages and limitations. *Plant Science*. 289: 110270