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Review article

Optimizing coating materials and processes for controlled release coated urea fertilizer: Review of bio-based coating materials and encapsulation methods

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ABSTRACT

The strict legal requirements in the European Union mandating biodegradable components in fertilizers by 2026 underscore the urgent need for innovative solutions in controlled release coated urea (CRCU) fertilizers. Despite the critical importance of optimizing coating materials and processes for CRCU fertilizers, a comprehensive review of optimization methods is notably absent, leaving a significant knowledge gap in the development of efficient and sustainable CRCU technologies. This review addresses this gap by providing an in-depth analysis of bio-based coating materials, encapsulation methods, and fluidized bed processes for CRCU fertilizers. We critically evaluate existing research, identify key challenges and opportunities, and discuss future directions for developing biodegradable and efficient CRCU fertilizers that meet the impending regulatory requirements. This review serves as a timely resource for researchers, industry stakeholders, and policymakers seeking to develop and implement sustainable coating solutions.

1. Introduction

Fertilizers such as urea are crucial for supplementing plant nutrients, but plants' nutrient needs fluctuate throughout their growth cycle, requiring smaller amounts during early stages and larger quantities during root, stalk, and stem development [1]. Unfortunately, conventional fertilizers often result in significant losses, with plants utilizing only about 30 % of the applied nutrients, while the remainder contributes to environmental degradation, including air and water pollution [2]. Excessive fertilizer application can damage young plants and have far-reaching consequences, including soil degradation, water pollution, increased greenhouse gas emissions, reduced plant health, and negative impacts on human health [3,4].

One solution for avoiding the previously mentioned problems is to coat the surface of solid fertilizers to obtain a controlled release fertilizer (CRF). CRF provide a cost-effective and environmentally friendly way to deliver nutrients to plants. These fertilizers are made by coating granules with a polymer that controls nutrient release [4]. Coatings have a broad spectrum of properties and can e.g. be liquid, solid, thermoplastic, and/or reactive [5]. Coatings are applied to fertilizers to maintain their quality during transportation, storage, and handling. These coatings serve various purposes, including dust control, caking prevention, improved flowability, moisture resistance, surface stabilization, enhanced compatibility, and aesthetic appeal [6]. In addition, the coating modifies nutrient release characteristics, which helps to increase fertilizer utilization efficiency and thereby mitigating its ecological footprint [7]. Despite their benefits, CRFs are predominantly used in developed nations, as their high costs and intricate production processes hinder their widespread adoption globally. Currently, effective controlled-release fertilizers (CRFs) often come with drawbacks, including the use of non-biodegradable coating materials, complex production processes, and high costs [8].

Substantial research investments have been dedicated to developing a wide range of coating materials and techniques, resulting in promising as well as non-promising materials and routes. For example, sulfur and starch have been found to be not effective as coating material because of their amorphous nature [8]. Sulfur was initially explored as a coating agent for controlled-release fertilizers due to its potential benefits [9,

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10]. However, its limitations, including poor wettability and adhesion to the core material, made it less viable. Additionally, sulfur coatings require conditioning materials to enhance their effectiveness, increasing production costs. Furthermore, residual sulfur in soil can react with water, leading to soil acidification, which may negatively impact soil health and plant growth [9,10]. To slow down nutrient release, different types of additives have been employed, including sealants, binders, plasticizers, and protective agents, but they add complexity and expense to the process, which has nearly led to the abandonment of sulfur-coated urea production. While polymer-based and superabsorbent CRFs show promise for prolonged release and water retention, their production is hindered by processing challenges, high costs, and environmental concerns, preventing large-scale industrial production [8,11].

While polymeric materials have shown to be promising as coating materials, they are not without issues. Unlike sulfur coatings, they are more resistant to microbial degradation [8]. Also, many polymers require harmful organic solvents such as dichloromethane [12], N, N-dimethyl-formamide (DMF) [13], or chloroform [14], posing environmental risks. Moreover, the non-biodegradable nature of most polymers used in CRFs contributes to soil pollution, resulting in the accumulation of plastic residues, estimated to be as high as 50 kg/ha/year [15]. To address these concerns, researchers are shifting focus to bio-based coating materials, which offer several advantages: they're cost-effective [16,17], biodegradable, biocompatible, and nontoxic, ultimately improving soil quality [18,19]. Biodegradable biopolymers, such as lignin [6,7], bio-polyurethane, chitosan, cellulose derivatives, guar gum, carrageenan, xanthan gum, and natural rubber [8,9], offer a promising alternative. However, these materials can have inconsistent nutrient-release characteristics due to surface heterogeneities. To improve coating uniformity and nutrient release, the impact of process variables has been reported [9,10]. The study in [9] examines how process parameters in fluidized bed spray granulation, including atomizing air pressure, fluidized-bed temperature, spray rate, and fluidizing-air flow rate, interactively affect the coating uniformity and nutrient release duration of CRCU particles. Achieving optimal coating film properties and nutrient-release dynamics depend on:

➢ Coating material selection: Choose materials with suitable properties, such as permeability, mechanical strength, and adhesion.
➢ Coating process optimization: Optimize coating processes, such as fluidized bed coating, to achieve uniform coating thickness and properties.

Using Design of Experiment (DOE) and analysis of variance (ANOVA), we identified the critical factors that impact coating uniformity and nutrient release duration. We then optimized the process using Response Surface Methodology (RSM), a proven approach for modeling and fine-tuning complex processes [10].

Urea is a widely used fertilizer worldwide, attributed to its high nitrogen content (46 %), cost-effectiveness, and convenient application. Coating materials for urea fertilizers are evaluated based on several key factors to enhance their performance. These factors include [8]:

i. Affinity with urea: Strong bonding to ensure effective coating.
ii. Water permeability: Controlled release of urea through the coating.
iii. Urea retention: Preventing immediate urea loss from the coating surface.
iv. Release mechanism: Tailored release of urea to match crop nutrient demands over time.
v. Sustainability and environmental impact: Biodegradability, eco-friendly, and cost-effectiveness.

So far, there is no single material that perfectly meets all these criteria (i-v). Nevertheless, researchers have conceived innovative solutions like chemically-modified lignin-palm stearin-based bio-resin. This bio-resin can significantly achieve the desired traits for effective urea encapsulation and controlled release. Studies of coating uniformity, thickness, release kinetics and their dependence on the process parameters, have been reported by several authors in CRCU production process optimization [20–23].

Fluidized bed technology offers a versatile and efficient method for coating various particles and granules, making it a valuable tool in granular urea encapsulation [22]. Fluidized bed coating technology improves the urea coating process by facilitating superior heat and mass transfer, thorough mixing, and faster processing. However, achieving coating uniformity is still challenging especially with larger particles, compatibility and high chance of solvent explosion. Specifically, suitable eco-friendly coating materials that provide sustained release kinetics and are well compatible with urea are a trending area of research.

Several reviews have discussed various aspects of controlled-release fertilizers (CRFs), including [24–30]:

✓ Membrane applications and technologies: Dave et al. [25] explored membrane-based controlled-release delivery systems and kinetics.
✓ Materials and methods: Azeem et al. [26] reviewed materials and methods for preparing CRFs.
✓ Nutrient release characteristics: Timilsena et al. [27] examined nutrient release profiles and formulations.
✓ Biodegradable polymers: Majeed et al. [24] discussed biodegradable polymers and blends for CRF applications.
✓ Coating materials and methods: Naz et al. [28] reviewed slow-release coating materials and techniques.
✓ Comprehensive review: Sempeho et al. [29] provided a broad overview, including empirical and mechanistic mathematical models.
✓ Mathematical modeling and simulation: Detailed mathematical models describing nutrient release from coated controlled-release fertilizer (CRF) granules have been reviewed [30].

This review examines the potential of bio-based coatings and fluidized bed technology to improve the effectiveness and eco-friendliness of controlled-release coated urea (CRCU) production. It focuses on optimizing coating materials and processes to improve fertilizer performance, reduce environmental impact, and promote sustainable agriculture as summarized in Fig. 1. This review further aims to provide insights into underutilized bio-based materials and optimization methods for developing effective CRCU fertilizers, promoting sustainable agriculture and reduced environmental impact.

1.1. Particle coating technologies for controlled release applications

Particles of varying sizes find applications in diverse industries, including pharmaceuticals, agriculture, wastewater treatment, food technology and plastics [31–33]. However, inhomogeneous sizes, shapes, and compositions can limit their effectiveness. To enhance their performance, particle properties such as flow, size, dustiness, strength, appearance, and dissolution rate can be tailored through modification techniques, allowing for improved functionality and efficiency.

Particles can be made more effective by controlling the release of their active ingredients, a technology known as controlled-release or delayed-release [32]. This involves coating particles with a material that regulates the release rate and duration, and is applied in various fields, including pharmaceuticals and agriculture. Additional applications of this technology include controlled-release viscous breakers for oil well stimulation and carbon black agglomeration, tablet coloring, carbonless paper production, and detergent manufacturing [33]. Additionally, controlled-release particles are useful in pharmaceuticals, enabling prolonged therapeutic effects from a single dose. Controlled-release particles have various applications in different industries, including:

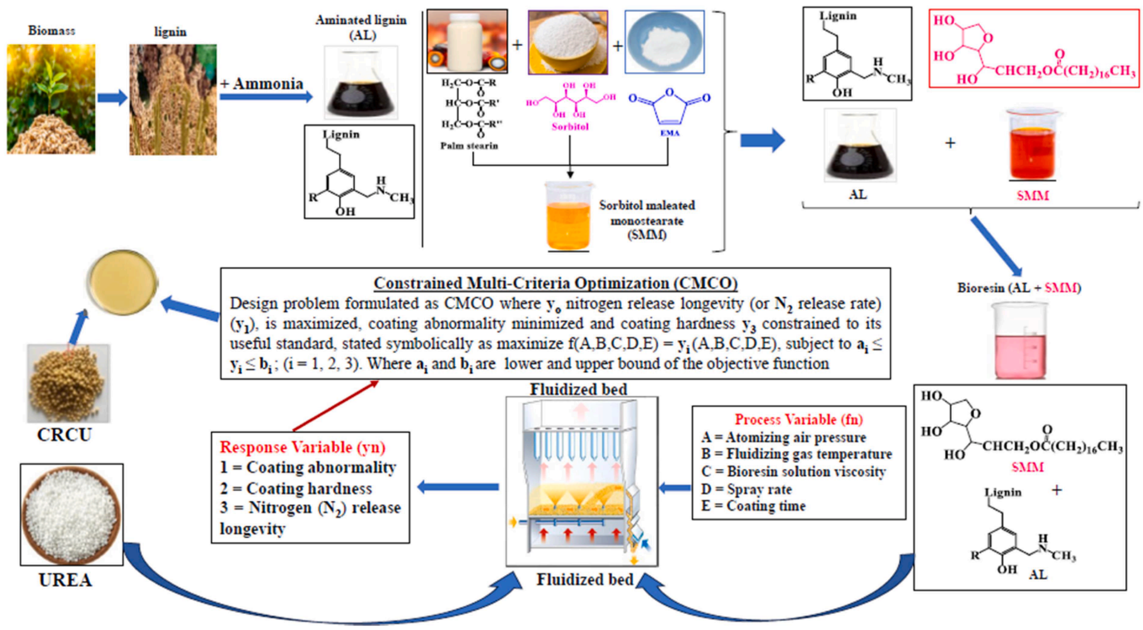

Fig. 1. Schematics of Bio-Coated Urea Fertilizer Process.

Pharmaceutics: Extended-release drugs for prolonged therapeutic effects, taste-masking to improve patient compliance, and targeted delivery to specific body areas, reducing side effects [35].
Agriculture Controlled-release fertilizers and pesticides, along with coated seeds, reduce environmental impact by minimizing application frequency and groundwater runoff, promoting more sustainable agricultural practices [31].

Particle surface modification and controlled-release particle production can be achieved through various methods using equipment that agitates particles, including: pans, mixers, fluid beds, and centrifuges [34]. Spray coating in rotating pans or fluidized beds is a widely used method for producing coated particles [35]. The rotating pan process fluidizes particles in a rotating drum or pan, where a coating solution is sprayed on, and hot gas is used to dry the particles as they move. Despite its long-standing use and improvements, the rotating pan method has limitations, including low reproducibility, making it less suitable for particles requiring high coating uniformity. Recent advancements focus on improving fluidization techniques, like fluidizing particles inside drums, to enhance coating efficiency and uniformity [36]. The conventional fluidized bed has undergone design modifications to enhance coating processes, capitalizing on its excellent mixing and heat transfer. One notable modification was introduced by Wurster in the 1950s, featuring a design with an inner tube (draft tube) that improves circulation within the bed [37]. Another design, by Glatt, uses a central cone to enhance particle circulation. The Kugel coater design has been introduced to further improve the process, featuring a vertical center shaft, multiple nozzles, and inclined perforated slots, replacing the traditional distributor plate [38].

Particles are fluidized by a gas in the fluid-bed coating process, creating a dynamic environment that facilitates efficient and consistent coating. Fluid-bed designs include conventional fluidized beds, spouted beds, and spouted-fluid beds, each offering unique particle coating characteristics. The gas, often preheated, serves as both a heat and momentum carrier [40,39]. The coating material is atomized and directed at the particles through a nozzle, with options for top-down or bottom-up spraying to ensure effective coverage. The material solidifies through evaporation, crystallization, or chemical reaction. The dynamics within the bed are influenced by factors such as bed conditions, properties of the seed particles, and the type of coating material applied. Atomized coating solution droplets may successfully attach to particle surfaces or remain airborne. Unsuccessful droplets are spray-dried, forming fines that can be elutriated or remain in the bed, potentially leading to fines agglomeration or "snowball" growth if captured by larger particles [41]. Depending on bed conditions, particle growth occurs by layered growth or during the collision of wetted particles when liquid and later solids bridges might be formed resulting in agglomeration. Moderately wetted particles may form solid bridges that remain joined or break apart depending on adhesive force strength, potentially leading to dry quenching, agglomeration, or coated particles. The outcome depends on the strength of adhesive forces, determining whether particles remain joined or break apart, resulting in coated particles or agglomerates [43]. Excessive wetting can cause wet quenching, where particles form large wet clumps, defluidizing the bed [42]. The occurring phenomena are summarized in Fig. 2. Fluidized bed spray coating enables producing tailor-made particles with adjusted chemical and mechanical properties for various applications, including food, pharmaceutical, and automotive industries [42], whereby the process conditions influence coating thickness, roughness, texture, and structural and mechanical properties [42–43].

1.2. Coating technologies for controlled release coated urea (CRCU) fertilizers

The coating procedure involves putting a thin layer of material on the surface of fertilizer granules to improve their physical and chemical properties, such as moisture protection and nutrient release. Many coating techniques are used in the commercial sector to provide slow or controlled-release for nitrogen discharge to the soil [44,45]. Fluidized beds, pan coaters, rotary drums, and melting or extrusion technologies are the most frequently used coating processes, which are based on physical principles (see Fig. 3). Alternatively, chemical methods like inverse suspension polymerization, solution polymerization/cross-linking, and microwave irradiation can be applied for coating of slow or

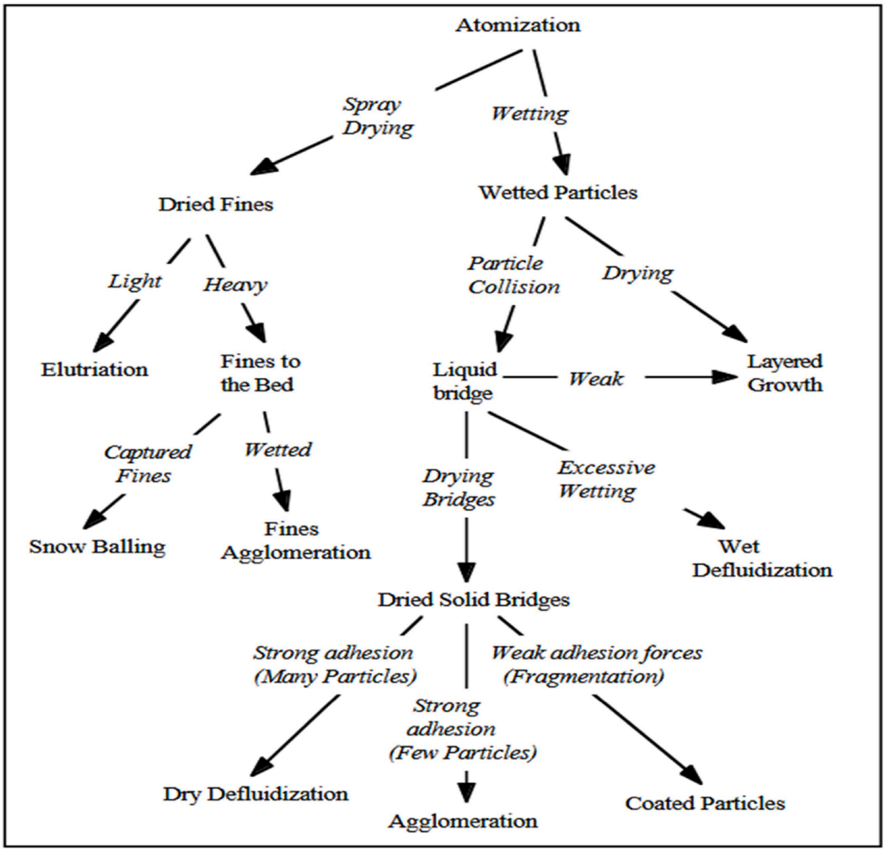

Fig. 2. Illustrates the various phenomena that can occur during the fluidized bed coating process, as documented in reference [43].

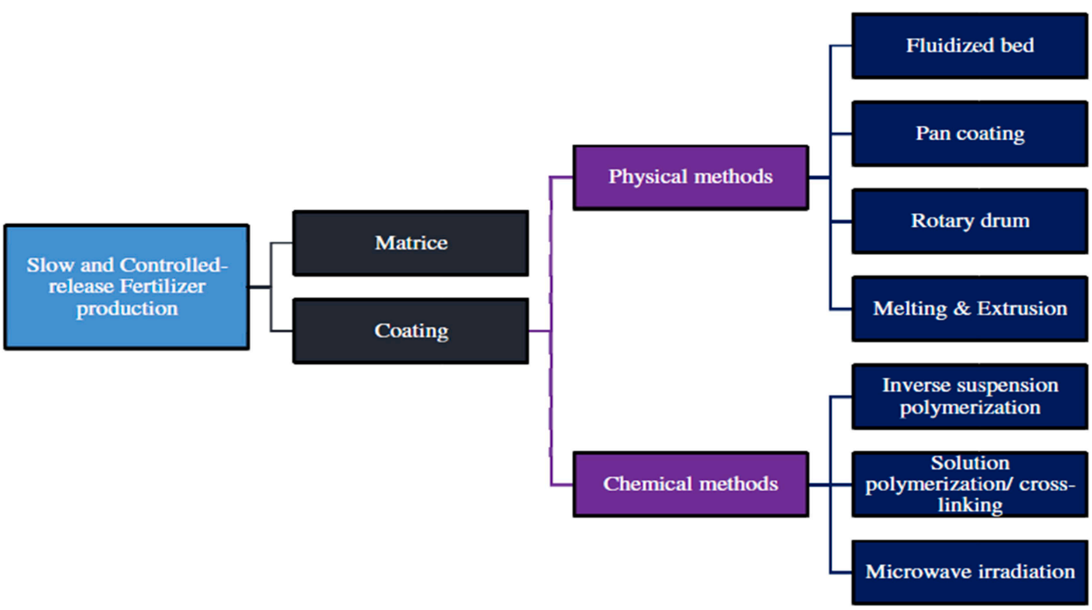

Fig. 3. Coating technologies for fertilizers [46].

controlled-release fertilizers that are also used on an industrial scale. However, all of these methods also have drawbacks, which are briefly explained in the following [46].

1.2.1. Physical methods of urea coating technology

Physical methods of urea coating involve depositing a coating material onto urea particles using physical forces or processes. These methods are widely used in the production of controlled-release fertilizers due to their simplicity, efficiency, and scalability and involve the following [46,50]:

- ✓ ***Spray coating (rotating drum):*** This method involves spraying a coating material, such as polymers or waxes, onto urea particles in a rotating drum.

- ✓ ***Fluidized bed coating***: In this process, urea particles are suspended in a fluidized bed, and a coating material is sprayed onto the particles.
- ✓ ***Extrusion Coating***: This method involves mixing urea with a coating material and then extruding the mixture through a die to form coated granules.
- ✓ ***Pan Coating***: This process involves coating urea particles in a rotating pan using a spray nozzle or another coating device.
- ✓ ***Spray Drying***: In this process, a coating material is sprayed onto urea particles within a hot gas flow, causing rapid solvent evaporation and resulting in a dry, coated product.

1.2.1.1. Fluidized bed, pan and rotary drum coating. In the fluidized bed process, fertilizer grains are suspended in a chamber by hot air and then coated. The coating substance can be gaseous, solid, or liquid. While the method requires advanced equipment and significant energy, it produces thin, homogeneous coatings [47,48]. The fluidized bed coating method produces uniform coatings using liquid or molten materials, offering a wide range of coating options, including hydrophilic, hydrophobic, solvent, and non-solvent materials [49,50]. This method allows for controlled release, which slows down with increased coating thickness. Additionally, coated fertilizers exhibit enhanced strength, being approximately three times more durable than their uncoated counterparts. The fluidized bed coating technique has been extensively studied using various natural and synthetic polymers [49–51]. To initiate the process, the fluidized bed is typically preheated to a temperature range of 45–50 °C for a duration of five minutes using fluidization gas, although temperatures as high as 80 °C have been reported [52]. The coating solution is then atomized and sprayed onto the urea granules at predetermined pressures and flow rates, which can vary from 0.3 MPa to 1.8–2 MPa. Following the coating process, the granules are dried at temperatures between 54–60 °C within the fluidized bed [49,50]. Researchers led by Ali [23] utilized a Wurster fluidized-bed reactor to coat granular urea with a specially formulated suspension derived from almond-shell waste. To ensure optimal conditions, the urea granules underwent thorough air drying and gravimetric analysis to eliminate any residual moisture [23]. The team investigated how various process parameters – including atomizing air pressure, fluidized-bed temperature, spray rate, and fluidizing-air flow rate – impacted the uniformity of the coating film and the duration of nutrient release. The Wurster fluidized-bed reactor's configuration is detailed in Fig. 4 and Table 1 [23]. The Wurster coating process, a bottom spray technique, enables precise control over active ingredient release. By positioning the spray nozzle at the base, particles are coated uniformly as they're accelerated upward through the Wurster tube, concurrently with the spray. As particles dry and fall outside the tube, they're guided back in for repeated coating, resulting in a consistent film. This method achieves thorough coating of particles, regardless of size, while minimizing coating material usage.

Table 1
Experimental process design of Wurster fluid bed coating of granular urea.

Process Variables		Range and Level					
	Acronym	Units	-α	−1	0	1	α
Atomizing air pressure	P_{atom}	Bar	1	2	3	4	5
Fluidizing-air flow Rate	F_{rate}	m^3/h	38	50.5	63	75.5	88
Fluidizing gas temperature	T_{fluid}	°C	45	65	85	105	125
Coating solution viscosity	V_{coat}	cP	200	270	340	410	480
Spray rate	S_{rate}	mL/s	0.02	0.08	0.14	0.2	0.26
Coating time	t_{coat}	Min	30	60	90	120	150
Response Variable							

Coating uniformity: Determined in terms of coefficient-of-variance (CoV) of film thickness.
Nutrient release kinetics: Time for 100 % release of nitrogen in a coated urea fertilizer.

Trung et al. [50] developed a biodegradable coating solution comprising phosphated di-starch phosphate, sodium tetraborate, polyvinyl alcohol, and polyacrylic acid for encapsulating urea using a pan coater. The pan coating system, depicted in Fig. 5, utilized an automated spray nozzle with a 0.8 mm diameter to apply the coating solution at 0.18 MPa pressure and flow rates ranging from 1.5 to 2.5 mL/min [50].

In the rotary drum process, fertilizer granules are coated by tumbling them with the coating material in a rotating cylinder, which can be in liquid or solid form [44]. Although this method offers flexibility and adaptability in coating, it may lead to granule abrasion and degradation. The structure of sprayed coatings is hard to anticipate without experimentation, as they produce more porous membranes than cast coatings. Additionally, achieving a homogeneous coating using a rotary drum requires a significant amount of raw materials, increasing process costs. Research has been conducted using this method with coating materials like gypsum, epoxy, and polyurethane [46]. Typically, urea granules are preheated to 70 °C for about 10 min in the rotary drum, then coated using side spray nozzles at 0.7 MPa pressure while the drum rotates at 60 rpm at a 45° angle. The entire process, including preheating, coating, cooling, and collection, can take one to three hours. Fig. 6 illustrates the rotary drum technique schematically.

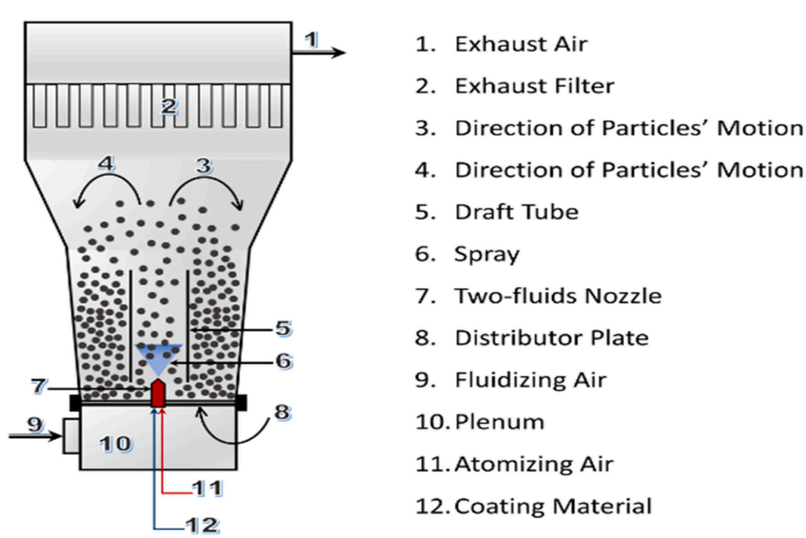

Fig. 4. Schematics of the Wurster fluidized-bed (WFB) reactor for urea coating applied by Ali et al. [23].

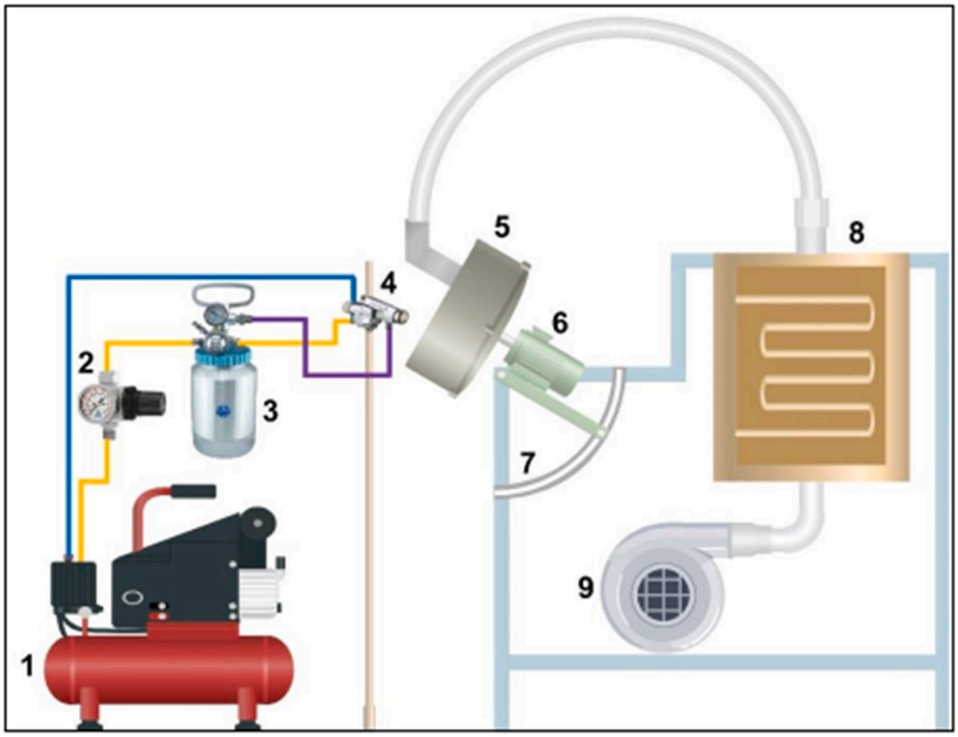

Fig. 5. Illustrates the pan coating system's layout [50], consisting of an (1) air compressor, (2) air regulator, (3) pressure tank, (4) automatic spray gun, (5) rotating pan, (6) motor, (7) inclinometer, (8) air heater, and (9) fan.

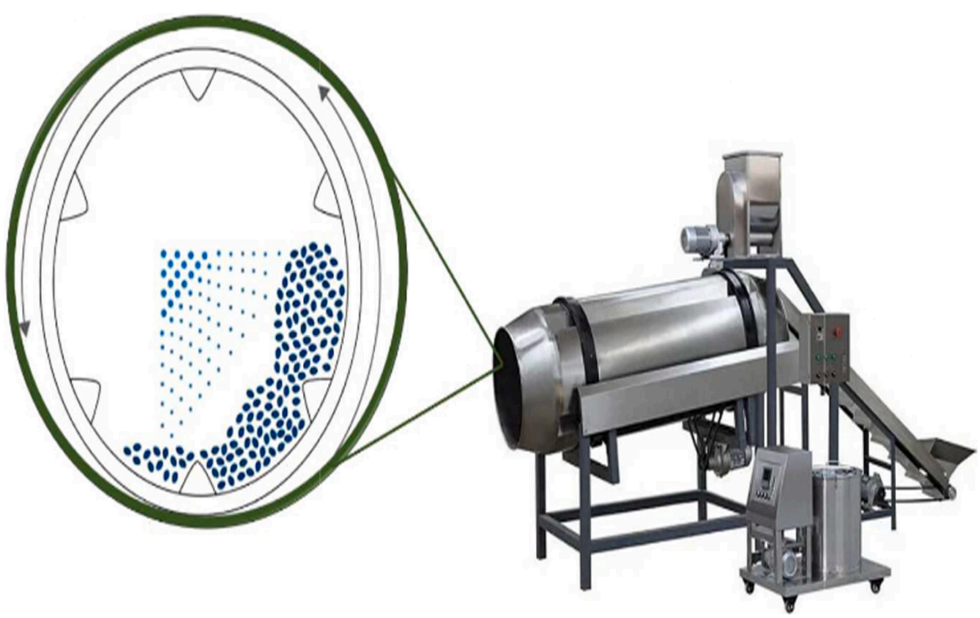

Fig. 6. Schematics of a rotary drum spray coater for urea coating.

Each coating technique has its strengths and weaknesses, and the most suitable method will depend on the specific application, product requirements, and desired properties. A comparative analysis of advantages and disadvantages of the three mentioned processes (fluidized bed, pan coater and rotary drum) is illustrated in Table 1.

1.2.1.2. Melting and extrusion. Spray coating processes typically rely on organic solvents to dissolve resins, regulate evaporation rates, and adjust viscosity, all of which impact the coating's adhesion and durability. However, the evaporation of these solvents poses risks to both human health and the environment [53–55]. To mitigate this issue, eco-friendly alternatives like melting and extrusion using single extruders have been developed. The process utilizes hot melts and requires expensive machinery. Various materials, including polyesters, clay, starch, and minerals, are used in this method to create slow or controlled-release fertilizers. To ensure successful production, careful monitoring of torque and temperature is essential when employing melting and extrusion techniques [56–59]. Bi et al. [58] developed a CRF using an extrusion method with a matrix approach. Their research showed that extruding urea phosphate/polymer composites resulted in high loading efficiency, creating a biodegradable and environmentally friendly controlled-release fertilizer.

1.2.2. Chemical methods

Chemical methods of urea coating involve chemical reactions that form a coating around urea particles, controlling the release of nutrients. These methods offer several advantages, including improved coating uniformity, enhanced durability, and tailored release profiles as described in the following subsections [60].

1.2.2.1. Inverse suspension polymerization. Inverse suspension polymerization uses a water-in-oil emulsion, where monomers and initiators are in the water phase, and stabilizers and solvents are in the oil phase. This process creates spherical polymer particles in the water droplets, which can be used to form fertilizer coating materials. Inverse suspension polymerization requires constant stirring due to its thermodynamic instability, with hydrophilic monomers and initiators suspended in a hydrocarbon continuous phase. The process allows for solvent recovery at a lower cost due to the insoluble nature of the product [61–63]. It also offers a faster reaction rate, potentially enhancing encapsulation efficiency. However, contamination risks necessitate additional purification steps.

1.2.2.2. Solution polymerization/cross-linking. In solution polymerization, monomers and initiators are mixed in a solvent, reducing viscosity and simplifying the process. However, the presence of cross-linking agents complicates solvent recovery, and the slower reaction rate can result in less efficient encapsulation [64]. These methods are used to develop hydrogel-based slow-release fertilizers from synthetic polymers, such as polyvinylpyrrolidone with acrylic acid and acrylamide, or natural polysaccharides like starch, alginate, and gelatin. Commonly used cross-linkers and initiators include N, N-methyl bisacrylamide and ammonium persulfate. Solution polymerization/cross-linking specifically involves [64–67];

- ➢ ***Graft Polymerization***: This method involves grafting monomers onto a polymer backbone, creating a coating around urea particles.
- ➢ ***Cross-Linking***: This process involves cross-linking polymers to form a network structure, which can be used to coat urea particles.

1.1.2.3. Microwave irradiation. The microwave irradiation process uses electromagnetic waves to heat and activate reactants in chemical reactions. This technique can be applied to coat fertilizers with materials like polymers, biopolymers, clays, and metals, enhancing their properties and performance. When macroradicals from different chains

Table 2
Comparative analysis of physical methods of urea fertilizer coating processes.

Advantages	Disadvantages
Fluidized bed coating [46,49,50]	
Uniform coating: Fluidized bed coating ensures uniform coating thickness and distribution on particles or granules.	Complexity: Fluidized bed coating requires specialized equipment and expertise.
Efficient heat and mass Transfer: The fluidized bed's dynamic nature enables efficient heat and mass transfer, promoting uniform drying and coating.	Attrition: The fluidization process can cause particle attrition or breakage.
Scalability: Fluidized bed coating can be easily scaled up for large-scale production.	Agglomeration: Particles can agglomerate if not properly fluidized or if the coating solution is not optimized.
Flexibility: The technique can accommodate various coating materials and substrates.	Difficulty in controlling and measuring coating thickness: Achieving precise control over coating thickness can be challenging.
Improved product quality: Fluidized bed coating can enhance product quality by providing a consistent and controlled coating process.	High energy consumption: Fluidized bed coating can require significant energy input, particularly for large-scale operations.
Pan coating [46,49,50]	
Well-established and widely used technique: Pan coating is a well-established and widely used technique in the pharmaceutical and food industries.	Limited scalability: Pan coating is generally limited to small to medium-scale production, making it less suitable for large-scale industrial applications.
Simple equipment and operation: The equipment and operation of pan coating are relatively simple, making it easy to implement and maintain.	Potential for uneven coating distribution: If the pan speed, spray rate, or drying conditions are not optimized, coating distribution can be uneven, leading to variability in product quality.
Suitable for small to medium-Scale production: Pan coating is suitable for small to medium-scale production, making it ideal for research and development or small-batch production.	Requires careful control of process parameters: Pan coating requires careful control of process parameters, such as pan speed, spray rate, and drying conditions, to achieve optimal coating uniformity.
Easy to clean and maintain: Pan coating equipment is relatively easy to clean and maintain, reducing downtime and increasing productivity.	May not be suitable for very small or very large particles: Pan coating may not be suitable for very small or very large particles, as they may not fluidize or coat evenly.
Can achieve high coating uniformity: Pan coating can achieve high coating uniformity, particularly if the pan speed, spray rate, and drying conditions are carefully controlled.	
Rotary drum spray coating [46]	
High capacity and scalability: Rotary drum spray coating is suitable for large-scale production, making it ideal for industrial applications.	Requires specialized equipment and expertise: Rotary drum spray coating requires specialized equipment and expertise, which can increase costs and complexity.
Suitable for large-scale production: The technique can handle large quantities of material, making it suitable for high-volume production.	Potential for particle attrition or breakage: The rotary drum process can cause particle attrition or breakage, particularly if the particles are fragile or the drum speed is too high.
Can handle a wide range of particle sizes: Rotary drum spray coating can handle a wide range of particle sizes, including small and large particles.	Difficulty in controlling coating thickness: Achieving precise control over coating thickness can be challenging due to the complexity of the rotary drum process.
Allows for continuous processing: The technique allows for continuous processing, reducing batch-to-batch variability and increasing productivity.	May require additional drying steps: Depending on the product and coating material, additional drying steps may be required, increasing processing time and costs.
Can achieve uniform coating distribution: Rotary drum spray coating can achieve uniform coating distribution, particularly if the drum speed, spray rate, and drying conditions are optimized.	Can be energy-intensive: Rotary drum spray coating can be energy-intensive, particularly for large-scale operations, which can increase costs and environmental impact.
Melting and extrusion [58]	
Advantage	**Disadvantage**
It is simple, low-cost, and solvent-free	Its equipment is expensive, and hot melting is involved

combine, they form covalent bonds and cross-linked structures. Under microwave radiation (320 W for 390 s), materials like biochar, cotton stalks, and maize cobs can create semi-interpenetrating networks [64]. This method was selected based on its simplicity, efficiency, and energy-saving benefits. However, its application in developing slow or controlled-release fertilizers remains limited and has not yet gained widespread adoption [68].

A detailed overview of the advantages and disadvantages of inverse suspension polymerization, solution polymerization/cross-linking, and microwave irradiation in urea coating for controlled-release coated fertilizer is given in Table 2.

2. Bio-based coating materials for CRCU fertilizers

The development of CRCU fertilizers has led to a surge in research on bio-based coating materials, offering promising solutions for more sustainable agricultural practices. These materials offer a sustainable and environmental friendly alternative to traditional coating materials. Bio-based coating materials can be derived from various sources, including plants, animals, and microorganisms. They can be used to create coatings with tailored properties, such as controlled release rates, improved durability, and enhanced nutrient use efficiency. Specially formulated CRFs are designed to synchronize nutrient delivery with plant demand, releasing essential nutrients in a gradual and timed manner [71]. This targeted approach boosts nutrient utilization efficiency and promotes higher crop yields [69]. A perfect controlled-release fertilizer features a coating made from natural or semi-natural, eco-friendly macromolecules that slow down nutrient release, allowing a single application to support crop growth throughout its lifecycle. Recently, controlled-release coated urea (CRCU) fertilizers have garnered considerable interest for their potential to enhance fertilizer efficiency, minimize environmental impact, and foster sustainable agricultural practices [69].

2.1. Types and classification of bio-based materials

The classification of CRFs is diverse and has been categorized in various ways, as illustrated in Fig. 7, according to the literature [8,23].

i. There are two primary types of organic compounds: natural organic compounds (e.g., animal manure, sewage sludge) and synthetic organic compounds with limited solubility. The synthetic group includes condensation products, such as urea formaldehyde (UF), isobutylidene diurea (IBDU), and cyclodiurea (CDU), which can be either biologically or chemically degradable.
ii. Water-soluble fertilizers can be designed with physical barriers to regulate nutrient release, often as granules or cores encapsulated with water-repellent polymers. These coated granular CRFs fall into two main categories: those coated with organic polymers, such as thermoplastics or resins, and those coated with inorganic materials, like sulfur or minerals. The matrix material controlling release can be hydrophobic, including polyolefins or rubber, or gel-forming polymers known as hydrogels. Hydrogels, being hydrophilic, slow down fertilizer dissolution by retaining large amounts of water through swelling.
iii. Inorganic compounds with low solubility, such as metal ammonium phosphates (e.g., KNH_4PO_4 and $MgNH_4PO_4$) and partially acidulated phosphate rock (PAPR), can provide a slow release of nutrients.
iv. The controlled release of nutrients from coated fertilizers occurs through mechanisms such as diffusion, erosion, chemical reaction, swelling, or osmosis [62]. When applied, irrigation water infiltrates the coating, condenses on the fertilizer core, and partially dissolves the nutrients, initiating the release process as shown in Fig. 8a. Fertilizer core with polymer coating is shown in Fig. 8b: As water penetrates the coating and core granule, fertilizer dissolution occurs, generating osmotic pressure. This pressure leads to controlled nutrient release through the swollen coating membrane. The increasing osmotic pressure causes the

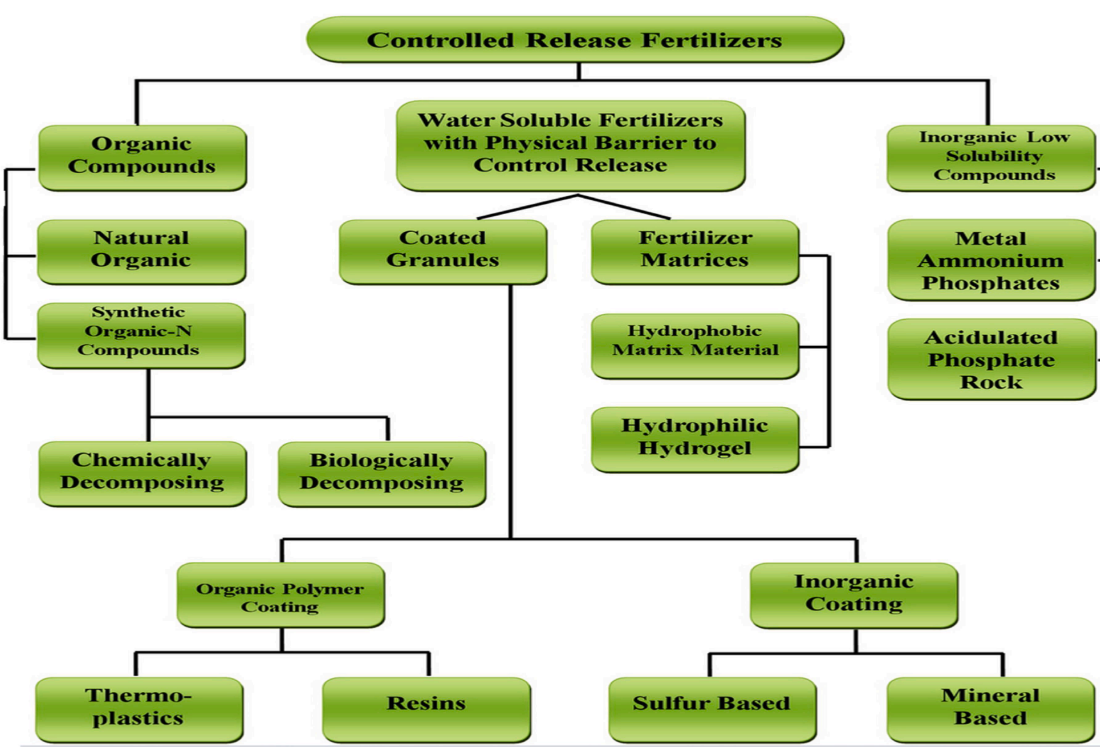

Fig. 7. Classification of controlled release fertilizers [8].

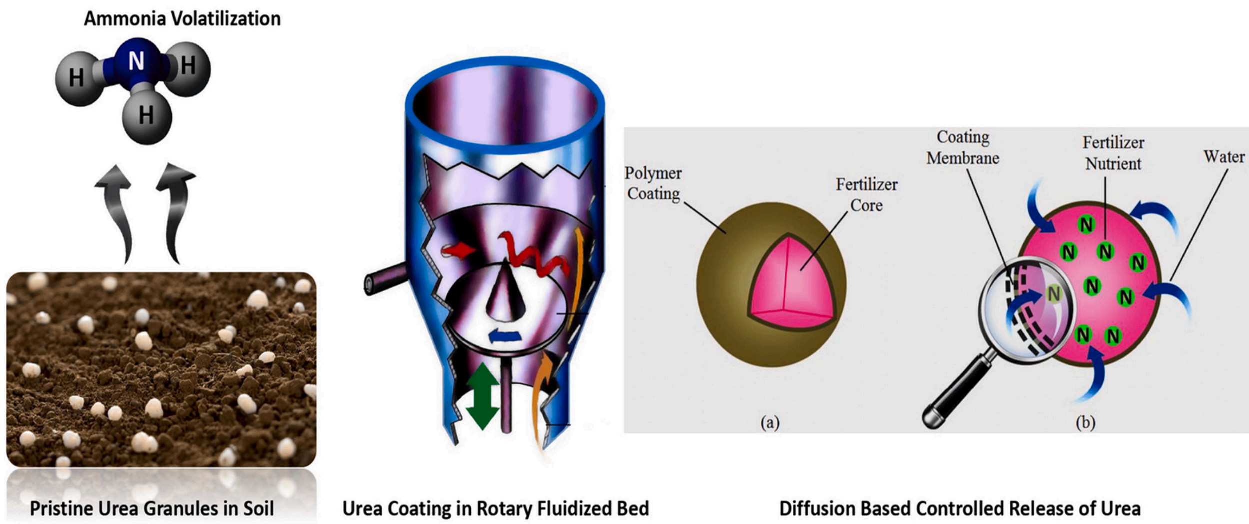

Fig. 8. Coating and diffusion mechanism of CRCU [16].

granule to swell, triggering two concurrent processes. Two possible release mechanisms can occur:

Failure mechanism (catastrophic release): If osmotic pressure exceeds the coating's resistance, it ruptures, releasing the entire core contents suddenly.

Diffusion mechanism: If the coating withstands the pressure, nutrients are released slowly through diffusion, driven by concentration or pressure gradients, or a combination of both.

The failure mechanism typically occurs in coatings with low strength, such as sulfur or modified sulfur, whereas polymer coatings like polyolefin tend to exhibit a diffusion-based release mechanism due to their greater durability.

Following the affair with sulfur, polymeric materials, bio-based materials have emerged as a promising option for coating urea fertilizers, offering a sustainable and environmentally friendly alternative to traditional coating materials. The use of biodegradable bio-based materials for coating fertilizers offers significant benefits for both agricultural fields and the environment [70]. By naturally breaking down, these materials can minimize the need for plastic-based coatings in controlled-release fertilizers (CRFs). Various plant and animal-derived resources are commonly employed as coating materials, providing a sustainable alternative for fertilizer applications [69–83]. Although bio-based coating materials have been investigated as a more sustainable approach to producing CRFs, their release control properties are limited by their high hydrophilicity and low mechanical resistance. To overcome these limitations, bio-based materials are usually chemically modified, combined with inorganic and plasticizers to form composites and blends, or used as main components for synthetic polymerization [73,75,84]. Several types of bio-based materials have been modified for CRCU coating, including (see Table 3):

- Polysaccharides: Such as starch [84–87], chitosan [88–91] and cellulose [91–93] which are abundant, renewable, and biodegradable.
- Natural oils and proteins: Like sunflower, soybean, hemp, castor oil etc. [94–96], which can be used to create biodegradable and renewable coatings.
- Lignin: A complex organic polymer found in plant cell walls, which can be used to create sustainable and biodegradable coatings [97]. Lignin is a promising natural polymer for controlled-release fertilizers due to its water-repelling properties, high carbon content, and biomass origin. Although it typically requires blending with other materials to form effective coatings, acetylation modification enables the creation of 100 % lignin-based coatings [96]. As lignin decomposes in soil, it converts into humic substances with phenolic groups, enhancing organic carbon content and inhibiting nitrification. This makes lignin a valuable component in sustainable fertilizer technologies [97–103].
- Natural waxes: Like beeswax and carnauba wax, which can provide a hydrophobic and biodegradable coating [104–108].
- Bioplastics: Such as polylactic acid (PLA) and polyhydroxyalkanoates (PHA), which are biodegradable and renewable [109–111].

Bio-based materials offer several benefits for CRU coating, including:

- Sustainability: Bio-based materials are renewable, biodegradable, and can reduce the environmental impact of fertilizer production.
- Biocompatibility: Bio-based materials are generally non-toxic and biocompatible, making them suitable for agricultural applications.
- Cost-effectiveness: Some bio-based materials may be less expensive than traditional coating materials [99,101].

3. Optimization methods for CRCU fertilizer

Manufacturing controlled-release coated urea fertilizers using fluidized bed technology involves complex interactions between process variables, coating materials, and fertilizer properties [23,85]. The optimization of the urea coating process in a fluidized bed is crucial for producing controlled-release coated urea (CRCU) with uniform coating and desired nutrient-release characteristics [23,85]. Understanding nutrient release kinetics in controlled-release fertilizers (CRFs) is crucial for optimizing application timing and rates, ensuring synchronized nutrient delivery with crop demands [141,144]. This knowledge enables manufacturers to design effective, efficient CRFs tailored to specific crops and growing conditions by analyzing coating and formulation impacts on nutrient release [142]. Key Optimization objectives include the following;

- **Coating Uniformity**: Achieving uniform coating thickness and distribution is essential for controlled release.
- **Release Rate**: Optimizing the release rate of urea to match crop demands and minimize losses.
- **Coating Material Efficiency**: Minimizing coating material usage while maintaining effectiveness.
- **Process Efficiency:** Optimizing process conditions to reduce energy consumption, processing time, and costs [143–154].

Table 3
Comparative analysis of chemical methods of urea fertilizer coating process.

Advantages	Disadvantages
Inverse suspension polymerization [46,61–63]	
i. Uniform coating: Inverse suspension polymerization allows for uniform coating of urea particles.	i. Complex process: Inverse suspension polymerization is a complex process requiring careful control of reaction conditions.
ii. Controlled particle size: This method enables control over particle size and distribution.	ii. Surfactant requirements: Surfactants are often required to stabilize the suspension, which can add complexity and cost.
iii. High encapsulation efficiency: Inverse suspension polymerization can achieve high encapsulation efficiency.	iii. Potential for agglomeration: Particles may agglomerate during the polymerization process.
iv. Mild reaction conditions: Reaction conditions are relatively mild, reducing the risk of urea degradation.	iv. Scalability issues: Scaling up the process while maintaining uniform coating quality can be challenging.
Solution polymerization/cross-linking [46,64–67]	
i. Tailored polymer structure: Enables the formation of polymers with tailored structures and properties, allowing for precise control over nutrient release rates.	i. Toxic chemicals: May involve the use of toxic chemicals or solvents, posing environmental and health risks.
ii. Improved mechanical strength: Cross-linking can enhance the mechanical strength and durability of the polymer coating, reducing damage during handling and application.	ii. Variable cross-linking: Can result in polymers with varying degrees of cross-linking, affecting nutrient release rates and fertilizer performance.
iii. Flexibility: Offers flexibility in terms of monomer selection, cross-linking agents, and reaction conditions, enabling the design of polymers with specific properties.	iii. Solvent removal: Requires the removal of solvents after polymerization, which can add complexity and cost to the process.
iv. High encapsulation efficiency: Can achieve high encapsulation efficiency, ensuring effective nutrient retention and release.	iv. Potential for urea fegradation: Harsh reaction conditions or prolonged reaction times may lead to urea degradation, reducing fertilizer efficacy.
Microwave irradiation [46,64,68]	
i. Rapid heating: Provides rapid and uniform heating, enhancing reaction rates and efficiency, and reducing processing times.	i. Limited control: Requires careful control of microwave power and reaction conditions to avoid overheating or non-uniform heating.
ii. Energy efficiency: Can be more energy-efficient than conventional heating methods, reducing production costs.	ii. Scalability issues: May not be suitable for large-scale production due to limitations in microwave penetration depth and potential hotspots.
iii. Improved coating uniformity: Enhances coating uniformity and consistency, ensuring precise control over nutrient release rates.	iii. Equipment requirements: Requires specialized equipment, which can add to the initial investment and maintenance costs.
iv. Tailored polymer properties: Allows for the synthesis of polymers with specific properties and structures, tailored to the desired release rate.	iv. Potential for urea Degradation: High-energy microwave radiation may lead to urea degradation if not carefully controlled.

Most in literature reported optimization methods for production of CRFs [85] are based mainly on empirical models and regression analysis as e.g.:

- Response surface methodology (RSM) [143,149–159]: A statistical technique used to model and optimize the relationships between process variables and responses (Table 4).
- Artificial Neural Networks (ANNs): A machine learning approach that can model complex relationships and predict optimal conditions.
- Genetic Algorithm (GA) [16]: A search heuristic that mimics natural selection to find optimal solutions.
- Particle Swarm Optimization (PSO) [21]: This is a social behavior-inspired optimization technique that utilizes a population-based approach

Babar et al. [23] reported that several process parameters significantly impact the coating uniformity and nutrient-release characteristics of CRCU in a fluidized bed. These include:

- **Atomizing Air Pressure**: Higher pressures can lead to smaller droplet sizes, resulting in more uniform coatings. Optimal pressure ranges from 3.2 to 3.5 bar.
- **Fluidized-Bed Temperature**: Temperature affects the drying rate of the coating solution, influencing coating uniformity and adhesion. Optimal temperatures range from 78 to 80 °C.

Table 4
Advantages of Bio-based Coatings for Controlled-Release Fertilizers.

Bio-based materials	Coating method	Fertilizer	Advantage	Ref.
Lignin, clay, and alginate	Encapsulation	Urea	Coatings demonstrated improved water absorption and reduced urea release compared to single-layer alginate coatings	[112]
100 % Acetylated lignin	Fluidized bed	Urea	Reduced nitrogen release	[113]
Waste kitchen oils (WKO)	Pan/rotary drum spray coating	Phosphorus	Enhanced membrane properties, including thermal resilience, crosslinking, and surface structure, lead to reduced and more controlled nutrient release.	[114]
Bio-based polyurethane coating and alginate hydrogel	Rotary drum coating	Copper, selenium, and nitrogen	Extend nutrition release time to 42 days and demonstrated a fungal inhibition	[115]
Almond-shell lignocellulosic waste	Fluidized-bed	Urea	Extend the nitrogen release time	[23]
Castor oil and polyurethane	Rotating drum	Urea	Hydrophobic modification enhanced the controlled release of coated fertilizers and elucidated the underlying mechanism of how hydrophobicity influences nutrient release	[116]
Epoxidized soybean oil acrylate	UV-curing encapsulation	Urea	Bio-based coatings incorporating stearic acid, Mg(OH)2, and paraffin wax exhibited superhydrophobic properties, enabling slow-release fertilizers to extend nitrogen release up to 63 days..	[117]
Corncob liquefaction for bio-polyurethane productio	Rotating drum spraying	Urea	Shows extreme super hydrophobicity	[118]
Lignin	Spray coating	Urea, di-ammonium phosphate and triple superphosphate	Fertilizers showed improved mechanical resistance and enhanced soil water retention capabilities.	[119–123, 126]
Acrylate epoxidized soybean oil	Rotary drum spray coating	Urea	Enhanced mechanical properties (hardness and toughness) of urea granules resulted in a greatly reduced release rate.	[124]
Corn starch/carboxymethyl cellulose composite	Infusement, layer-by-layer assembly procedure.	Urea Particles	Cumulative release reached 84 % on the seventh day	[125]
Poly(ε-caprolactone)-grafted lignocellulose from walnut wood.	Encapsulation, Melt extrusion and granulation	Urea	Shows acceptable super hydrophobicity	[127]
Castor oil polyurethane enhanced with liquefied starch polyol and siloxane.	Rotating drum coating	Urea	Increased water repellency and prevented water ingress	[128]
Bio-polyurethane formulation incorporating soybean oil and polyols	Encapsulation	Urea	Reduced rate of controlled nutrient release.	[129]
Bio-polyurethane formulation modified with locust sawdust	Rotary drum coating	Urea	Reduced rate of controlled nutrient release.	[130]
Liquefied apple tree branch polyol enhanced with castor oil.	Spray coating	Urea	Reduced rate of controlled nutrient release.	[131]
Microbial fertilizers formulated with biochar.	Encapsulation	Carbon-based Fertilizer	Reduced rate of controlled nutrient release.	[132]
Nanostructured lignin-clay composite material	Spray coating Double layer	Urea	Improved water holding capacity, improved nutrient utilization efficiency	[133]
Castor oil and other essential oil	Spray coating	Urea	Reduced nutrient release rate	[134]
Chitosan and poly (acrylic acid-co-acrylamide)	Double coat via spraying and dipping	Urea	Reduced nutrient release rate	[135]
Bio polyols modified polyurethane	Rotating drum	Urea	Self-healing bio-nanocomposite-coated CRF	[136]
Castor oil-modified polyurethane	Rotating drum coating	Phosphorous	Better nutrient release rate	[137]
Poly acrylic acid grafted natural rubber.	Pan granulation and spraying	Urea	Extended release of nitrogen, phosphorus, and potassium for 30 days	[138]
Carbon-based slow release fertilizer treated by bio-oil coating	Fluidized bed spraying	Biochar/carbon-based fertilizer	Film-forming/coating properties of the bio-oil strengthened the slow release capacity.	[139]
Composite material made from cassava starch and bagasse.	Gelatinization/ plasticization	Urea	Increased nutrient use efficiency	[140]

- **Spray Rate**: A lower spray rate can yield more uniform coatings, but may increase processing time. Optimal spray rates range from 0.125 to 0.15 mL/s.
- **Fluidizing-Air Flow Rate**: Optimal flow rates ensure uniform particle fluidization and coating. Rates range from 72 to 75 m^3/h.
- **Coating time**: As coating time increases, the release time of urea also increases, but beyond an optimal point, the release time decreases due to disintegration of the coating film. This disintegration occurs when substrate granules are coated for an extended period, likely caused by the hydrodynamic forces present in the fluidized bed [85, 143].

Fluidized bed coating processes can be efficiently optimized using response surface methodology (RSM) and central composite design (CCD). These methods help investigate the interactive effects of process parameters on coating uniformity and nutrient-release characteristics. Studies have reported optimal conditions for coating uniformity and nutrient-release performance [23,85]:

- **Coating Uniformity**: Atomizing air pressure of 3.5 bar, fluidized-bed temperature of 80 °C, spray rate of 0.15 mL/s, and fluidizing-air flow rate of 72 m^3/h resulted in a predicted coefficient of variance (CoV) film thickness of 11.5 %.
- **Nutrient-Release Performance**: Atomizing air pressure of 3.2 bar, fluidized-bed temperature of 78 °C, spray rate of 0.125 mL/s, and fluidizing-air flow rate of 75 m^3/h yielded a predicted nutrient-release time of 56 h.
- **Coating thickness:** Optimum film thickness crucial for desirable coating uniformity and effective nutrient-release characteristics [143].

Optimizing the urea coating process in a fluidized bed requires careful consideration of process parameters and coating materials. By understanding the effects of these factors on coating uniformity and nutrient-release characteristics, researchers can develop more efficient and sustainable CRCU production methods. In the coating industry, achieving product quality involves a detailed process design that can be actualized in three major phases: characterization, control, and optimization [144–159].

➢ Characterization: Identifying critical variables that impact the system's output response. This phase separates vital factors from trivial ones, often using experimental designs like full factorial or fractional factorial arrays.
➢ Control: Achieving stable system performance through analysis and consideration of various factors. Advanced process control strategies include signal-to-noise ratio analysis, response surface modeling, and complex modeling techniques.
➢ Optimization: Manipulating significant process variables to achieve the most desirable operating conditions. RSM is often used for optimization, especially when multiple conflicting output characteristics are involved [98,99].

The characterization, control (all the necessary analysis and consideration to achieve desired and stable system performance) and optimization of the fluidized bed coating process (or product) are carried out within a predetermined space of the process parameters which include [20–23]: atomizing air pressure, fluidizing gas temperature, spray rate, spray temperature, coating time, molar ratio of coating materials, viscosity, etc. Each of these factors significantly impacts the system's response variable including, coating uniformity, coating hardness, coating thickness nitrogen release kinetics, biodegradability of the coating material. The coating can serve multiple purposes, such as controlling dust, reducing caking and moisture absorption, improving flowability and appearance, stabilizing the surface, and enhancing compatibility in various applications [22,23,141]. The characterization (or screening) of the major process parameters to the process response are usually achieved through Design-of-Experiment (DOE) and ANOVA while the control and optimization tasks accomplished using the well-known RSM [16,23,85]. Babar et al. [16] utilized a rotary fluidized bed equipment to create controlled-release coated urea granules with a coating material consisting of tapioca starch, polyvinyl alcohol, and citric acid. They chose RSM with a full-factorial CCD to study the effects of five factors at five levels, optimizing the process with a reduced number of experiments. RSM effectively showed how process parameters interact to affect urea release, with fluidizing air temperature and spray rate being the most influential. Optimal conditions were identified as 80 °C fluidizing air temperature, 0.13 mL/s spray rate, 3.98 bar atomizing pressure, 110 min process time, and 70 °C spray temperature. A double coating with modified starch and geopolymer further enhanced controlled release characteristics, yielding promising results for prolonged nitrogen release. Other studies [20,163] provided optimal parameters for the scaled-up controlled release fertilizer production in a rotary fluidized bed coating process. Urea granules are coated with the starch-polyvinyl solution using a two-fluid nozzle atomizer [163] and borax-modified starch biopolymer using a tumbling fluidized bed [20]. In [16,20,163], the global design matrix given in coded notations recommends sixteen (32) experimental runs at 2^5 – full factorial distinct points, eight (10) runs at the 10-unique axial points and ten (10) repeated runs at center point which give rise to a total of fifty experimental runs [23]. The results of these sequence of experiments were recorded in terms of the calculated coefficient of variance of the thickness of the coated urea. To understand the effects of process variables on responses, a predictive model is usually fitted to the data using regression analysis. This model serves as the basis for detailed statistical analysis and optimization exercises [145,154]. Preliminary findings from the steepest ascent method revealed possible curvature effects, prompting the development of a reduced-order quadratic model (ROQM) for further investigation, expressed in Eq. (1) [16,20]:

$$Y_n = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i<} \sum_{j=2}^{k} \beta_{ij} X_i X_j + \sum_{i=1} \sum_{j=1}^{k} \delta_{ij} X_i X_j + \varepsilon \quad (1)$$

[145,154]

Where Y_n is the measured system responses (coating uniformity and nitrogen release longevity). The first term by the right side of the equation β_0 is the intercept, the second terms characterizes the main linear effects of individual process variables, the third term incorporates the interaction effects between the variables, while the fourth term represents the main quadratic effects and ε is the random error of experimentation. $X_i$and Xjrepresents the matrix of the uncoded process variables. The determination of the unknown coefficients of β_0, β_i, β_{ij} *and* $\delta_{ij}$for nth investigation is accomplished via regression analysis implemented on the statistical analysis software Design-Expert sofware [16,145].Table 5

4. Global controlled-release fertilizer market size and growth

The global controlled-release fertilizer market is experiencing significant growth, driven by increasing demand for efficient and sustainable agricultural practices. According to recent market research as shown in Fig. 9, the global controlled-release fertilizer market is projected to reach $3.2 billion by 2029, growing at a compound annual growth rate (CAGR) of 6.0 % from 2022 to 2029 [155]. This growth is attributed to the rising awareness of the benefits of controlled-release fertilizers, including improved crop yields, reduced fertilizer consumption, and minimized environmental impact. The Asia Pacific region dominates the market, accounting for 46.5 % of the global market share in 2022, followed by North America and Europe. The growing demand for controlled-release fertilizers is driven by the increasing need for sustainable agricultural practices, reducing environmental pollution, and improving crop yields. The market is dominated by key players such as Yara, Nutrien Ltd., Mosaic, ICL, Nufarm, Kingenta, ScottsMiracle-Gro,

Table 5
Optimization methods for controlled urea coated fertilizer.

Response Surface Methodology			
Coating materials	Coating methods	Results	Ref.
Mixture of gypsum-sulphur	Rotary pan-equipped process	Coated urea efficiency increased by approximately 59 % (from 22 % to 35 %) under optimal process conditions.	[160]
Modified-starch biopolymer and geopolymer	Rotary fluidized bed	Optimized process conditions resulted in a controlled release of urea with a diffusion coefficient of 1.884 × $10^{(-5)}$ cm^2/s	[16]
SCPU composite: sodium alginate, carboxymethyl starch, polydopamine, and urea.	Encapsulation	The optimal preparation conditions for achieving high encapsulation efficiency and water absorption are: 2.2 % sodium alginate, 5 % carboxymethyl starch sodium, 30 % urea, 1.9 % calcium chloride, and 0.52 % dopamine.	[161]
Organosilicon and nanosilica-modified bio-based polyurethane (BPU)	Encapsulation	The superhydrophobic BPU coated fertilizer (SBPCF) showed significantly improved nutrient release characteristics compared to the regular BPU coated fertilizer (BPCF).	[162]
Waterborne starch biopolymer	Fluidized bed	Promising controlled-release properties achieved with optimal coating thickness and uniform film integrity.	[49]
Borax-modified starch biopolymer	Tumbling fluidized bed	Nitrogen release duration and kinetics are most affected by coating time.	[20]
Starch modified with polyvinyl alcohol (St-PVOH)	Rotary fluidized bed coating process, where urea granules are coated with the St-PVOH solution using a two-fluid nozzle atomizer.	The diffusion coefficient of nitrogen release from the coated urea ranged from 4.43 × $10^{(-11)}$ m^2/s to 6.98 × $10^{(-11)}$ m^2/s, depending on the fluidized bed process variables.The coating thickness and uniformity were affected by the process variables, with a coefficient of variance (CV) ranging from 10.3 % to 23.1 %.The study demonstrated the potential of using fluidized bed technology to produce controlled-release urea with improved nutrient release characteristics.	[163]
Modified starch biopolymer	Fluidized-bed	The coated urea showed controlled-release properties, with longer coating times resulting in extended nutrient release. Coating time and fluidizing gas temperature were the key factors influencing release characteristics and coating uniformity.	[164]

Table 5 (*continued*)

Response Surface Methodology			
Coating materials	Coating methods	Results	Ref.
Starch-based biopolymer	Fluidized bed granulator	The particle size growth rate increased by 30–50 % with increasing binder concentration. The granule size distribution was affected by binder properties and operating conditions, with starch-based binders producing more uniform granules. The optimal binder concentration for granule growth was found to be around 10–20 % (w/w).	[165]
Starch-based biopolymer	Tumbling fluidized-bed coating process	Coating uniformity improves at moderate atomizing pressure, while coating mass increases linearly with time. Optimal conditions for uniformity are 85 °C and 1.90 bar.	[22]
Chemically modified almond shell lignocellulosic microparticles.	Rotary pan coater	Urea dissolution time increased from a couple of minutes (uncoated) to >20 h (coated). Spray rate was found to be the most influential process parameter affecting nutrient release.	[166]
Fly ash-based geopolymer	The stainless-steel pan coater features a thin cylindrical drum or disk design.	The coated urea showed a prolonged release time of up to 30 days. The pan fill ratio, particle size, and the pan speed were the most influential parameter.	[167]
Almond shell-derived lignocellulosic biopolymer with chemical modifications.	Wurster fluidized-bed reactor.	For optimal coating quality: Coefficient of coating mass variance (CMV) = 6.7 % achieved under conditions of Tfb = 75 °C, Qair = 80 m^3/h, Rspray = 0.17 mL/s, and Pair = 3.1 bar. For optimum diffusion coefficient: 2.2 × 10^{-7} cm^2/s obtained at Tfb = 78 °C, Qair = 75 m^3/h, Rspray = 0.125 mL/s, and Pair = 3.2 bar.	[168]
Modified almond-shell lignocellulosic waste particles	Wurster fluidized-bed reactor.	Coating Uniformity: Optimum conditions for coating-film uniformity were achieved with atomizing air pressure = 3.5 bar, fluidized-bed temperature = 80 °C, spray rate = 0.15 mL/s, and fluidizing-air flow rate = 72 m^3/h, resulting in a predicted coefficient of variance (CoV) of film thickness = 11.5 %. Nutrient Release: Optimum conditions for nutrient-release performance were achieved with atomizing air pressure =	[23]

(continued on next page)

Table 5 (*continued*)

Response Surface Methodology			
Coating materials	Coating methods	Results	Ref.
		3.2 bar, fluidized-bed temperature = 78 °C, spray rate = 0.125 mL/s, and fluidizing-air flow rate = 75 m^3/h, resulting in a predicted nutrient-release time of 43.65 h. [1]	
Urea-based SCPU composite with sodium alginate, carboxymethyl starch, and polydopamine	The SCPU was prepared using a two-step method, involving the formation of gel-like particles and subsequent coating with polydopamine.	The optimal conditions yielded an encapsulation efficiency of 89.27 % and water absorption of 167.05 %, indicating good water-holding capacity. The optimal preparation conditions were: Sodium alginate: 2.2 %, Carboxymethyl starch sodium: 5 %, Urea: 30 %, Calcium chloride: 1.9 %, Dopamine: 0.52 %.	[169]
Molten urea	Two-fluid nozzle FBG with external mixing setup	Coating Performance (CP): Maximum CP achieved was 60 % under optimum conditions. Morphology: The coated urea seeds had a compact molten urea layer, as observed by SEM. Particle Size Distribution: Proper Size Guide Number (SGN) and Uniformity Index (UI) were achieved under specific conditions.	[170]
Biodegradable polymer consisting of: Phosphated di-starch phosphate, Polyvinyl alcohol, Polyacrylic acid	Pan coater	The optimized pan coating process resulted in a coating efficiency of 93.45 %. The coated urea fertilizer showed a controlled release profile, with a significant reduction in urea release rate compared to uncoated urea. The optimal pan coating conditions were determined to be: Pan speed: 20 rpm, Spray rate: 10 g/min, Atomizing air pressure: 2 bar, Coating time: 30 min	[50, 171]
Machine Learning and nature inspired optimization			
Neem oil, acacia oil and bentonite clay with and without heat	Rotating drum coater	The machine learning and Response Surface Methodology (RSM) models predicted that the coated USG reduced nitrate leaching by 64.71 % compared to uncoated USG, with the sulfur content in the coating material having a significant impact on nitrate leaching reduction.	[172]
Tapioca starch	Rotary fluidized bed system	The Gaussian Process Regression (GPR) model accurately predicted nutrient release time from biopolymer-coated fertilizer, achieving an R^2 value of 1 and RMSE value of 0.003. The model utilizes key input parameters including diffusion coefficient, coefficient of variance of coating thickness, coating mass thickness, coefficient of variance of size distribution, and surface hardness to simulate and predict outcomes.	[21]

Koch Industries, Helena Chemicals, SQM, and Haifa Chemicals, who are focusing on developing innovative products and strategies to meet the growing demand for controlled-release fertilizers. The Asia Pacific region dominates the market, while North America is expected to record USD 2 billion by 2032. The coated and encapsulated fertilizers segment dominates the market, with the agricultural segment holding a major share of 60 % in 2024 [156]. This growth reflects a shift towards sustainability in today's fertilizer and agriculture processes, as controlled-release fertilizers play a vital role in reducing nutrient losses, minimizing environmental pollution, and promoting efficient fertilizer use, ultimately enhancing crop productivity while minimizing the environmental footprint of agriculture

5. Challenges and future perspectives

The development of CRCU fertilizers has made significant progress in recent years, with various bio-based coating materials and fluidized bed technologies being explored (see Fig. 9). However, there are still challenges and several areas that require further research and development to improve the efficiency, sustainability, and cost-effectiveness of CRCU production [154]. Alkyd resins are known for their excellent adhesive and cohesive properties, which make them suitable for various applications [173–186]. Their compatibility with a wide range of polymers and extensive formulation flexibility makes them suitable for producing a broad spectrum of coating materials, encompassing everything from paints and varnishes for wood and metal to specialized coatings like road marking paints, anticorrosive paints, nitrocellulose lacquers, two-component isocyanate curing coatings, acid curing coatings, and stoving enamels [74,154]. Noteworthy, alkyd resin has not been used for coating of urea fertilizer. Most raw materials used in alkyd synthesis come from biologically renewable sources, except for phthalic anhydride, which is petrochemically derived. Their biodegradability makes them ecologically appealing binders. However, due to their hydrophobic nature, alkyds often require modification to function effectively in aqueous environments [74]. Neem oil, rapeseed oil, sunflower oil and cherry seed oil are unsaturated triglyceride resources from proven inedible industrial crops with annual production exceeding 19.2 and 2.2 million metric tons for rapeseed and sunflower oil in Europe, respectively [187]. Neem oil's natural insecticidal and fungicidal properties enhance fertilizer effectiveness while its alkyd can form a hydrophobic matrix, capable of controlling urea release through diffusion. Lignin is nature's second most abundant bio-renewable polymer. Aminated lignin (AL) can promotes coating adhesion and agglomeration growth, can enhance mechanical strength of the granules, reduces breakage and dust formation and consequential ammonia release to environment. Hydrogen bonding integrity and humectant properties of sorbitol as a hydrophilic monomer to maleic anhydride is suitable for waterborne alkyd (WA) resin solutions. These exceptional properties align well with

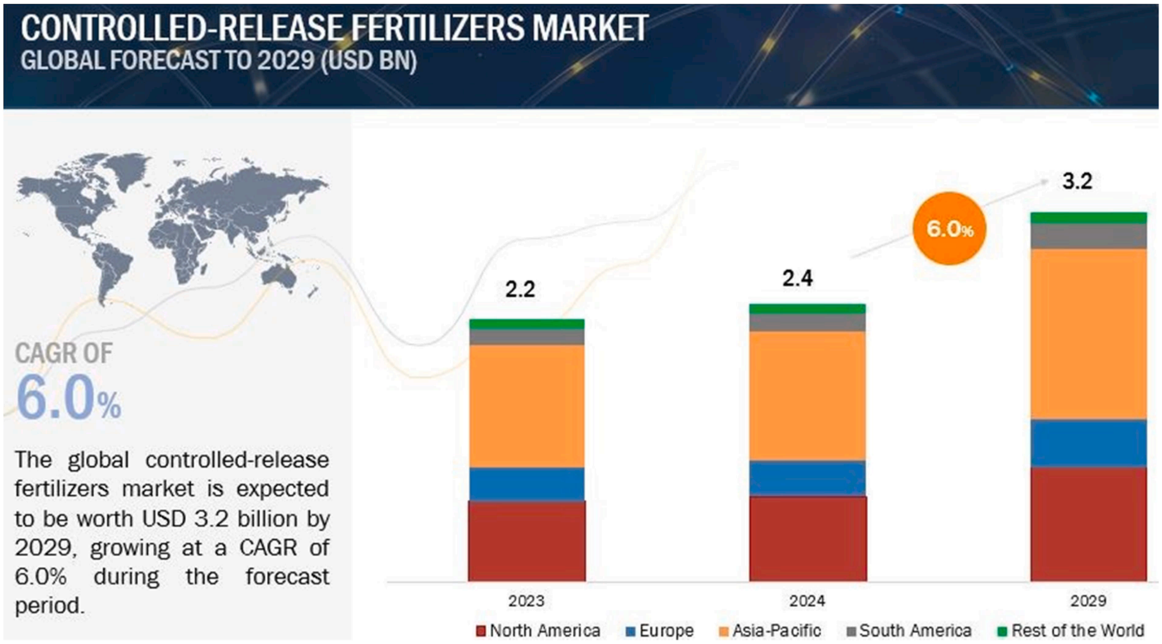

Fig. 9. Global controlled-release fertilizer market size and growth [155].

the future development of zero VOC (Volatile Organic Compound) coatings. The fulfilment of sustainability goals makes waterborne coating material for CRCU an excellent fit with EU's strategy [176]. The EU's stringent regulations mandate that fertilizer materials become biodegradable by July 2026, driving the need for material changes [188]. The European Fertilizing Products Regulation poses a challenge for the fertilizer industry, particularly for controlled-release fertilizers. To comply with the upcoming ban on non-biodegradable coating materials in 2026, manufacturers must find alternative materials that maintain their integrity during the intended release period but degrade rapidly afterward. While reducing microplastic release is a worthy goal, developing biodegradable coatings is hindered by the lack of standardized biodegradability criteria and testing methods. The industry supports reducing microplastic pollution but needs clear guidelines to develop compliant products. The European Commission is tasked with establishing these standards by 2024 to facilitate a smooth transition. Given the 2024 deadline for establishing standards, the industry will have a tight two-year window to develop and finalize biodegradable coating materials that meet the new regulatory requirements, posing a significant challenge. From the fore-going review, the biggest obstacle in CRCU fertilizer is the lack of a coating material that addresses all essential unresolved issues associated with uncoated urea, including inconsistent nutrient release profiles and environmental impact. Moreover, existing technologies struggle with achieving uniform coating and granules for tailored release, with no reported literature on continuous granulation and coating of urea in a fluidized bed. We don't yet fully understand how different desirable but conflicting CRCU response variable influence their performance, production process and scale-up. Future research should focus on developing low-cost, biodegradable materials for controlled-release fertilizers, exploring blends and binders that balance adsorption and hydrophobicity. Field testing under various environmental conditions is necessary to validate results and study the release patterns over the fertilizer's lifespan. Additionally, upscaling procedures and methods need to be developed to bring these fertilizers to practical and commercial use.

5.1. Challenges

The production of Controlled Release Coated Urea (CRCU) fertilizer faces several challenges that must be addressed to ensure its widespread adoption. Scaling up CRCU production while maintaining product quality and reducing costs is a significant challenge. Achieving uniform coating thickness and controlled nutrient release rates remains a challenge, particularly for large-scale production, which can impact fertilizer efficiency and environmental sustainability. Selecting suitable bio-based coating materials that are compatible with urea and can provide sustained release kinetics is crucial, requiring careful consideration of material properties and interactions. Optimizing process parameters, such as atomizing air pressure, fluidized-bed temperature, and spray rate, is essential for achieving desired coating uniformity and nutrient release rates, and reducing the environmental impact of CRCU production, including energy consumption and waste generation, is a significant challenge that warrants attention. Addressing these challenges will be critical to realizing the full potential of CRCU fertilizer and promoting sustainable agriculture practices.

5.2. Future perspectives

Developing new bio-based coating materials that are abundant, renewable, and biodegradable could further reduce the environmental impact of CRCU production. Improving fluidized bed technologies, such as designing new reactor configurations or optimizing process conditions, could enhance coating uniformity and reduce processing time. Designing and developing new fluidized bed reactor configurations that enable continuous and simultaneous granulation and coating of powdered fertilizers will facilitate the development of controlled-release coated urea (CRCU) with tailored particle size and nutrient release kinetics. Applying machine learning algorithms and nature-inspired optimization techniques could help predict and optimize CRCU performance, reducing the need for trial-and-error experiments. A multi-objective problem can be formulated to consider the balance of trade-off between different desirable but conflicting objectives, such as maximizing N2 release (y_1), minimizing coating heterogeneity (y_2), and constraining coating degradability (y_3) to its useful standard. Integrating CRCU technology with precision agriculture practices could optimize fertilizer application, reduce waste, and promote sustainable agriculture. Conducting life cycle assessments of CRCU production could help identify areas for improvement and reduce the environmental impact of the production process, ultimately contributing to a more sustainable and environmentally friendly fertilizer production.

5.2.1. Underutilized bio-based materials

Exploring underutilized bio-based materials, such as alkyd resin, lignin, and natural waxes, alginate, protein could provide new opportunities for CRCU production.

Some underutilized bio-based materials that can be used for urea fertilizer coating include:

Chicken leather protein: This biodegradable protein-based material has shown promise in controlled-release urea production.
Wheat Straw: Combined with sodium alginate, wheat straw can create a biodegradable coating for urea, improving its release characteristics.
K-Carrageenan-Sodium Alginate: This natural polymer blend has demonstrated potential in coating urea, achieving a 94 % release rate over 25 days.
Ethyl Cellulose: A bio-based polymer that can be used to coat urea, controlling its release rate and improving fertilizer efficiency.
Starch-based Materials: Various starches, such as tapioca starch modified with polyvinyl alcohol and citric acid, have been used to create biodegradable coatings for urea.
Lignin: A complex organic polymer found in plant cell walls, lignin could offer a sustainable and renewable coating option.
Natural Waxes: Waxes like beeswax or carnauba wax might provide a biodegradable and water-resistant coating for urea

Some materials that haven't been extensively explored for urea coating include:

Seed-based Biopolymers: Certain seeds, like those from the guar bean or locust bean, contain galactomannans that could be used as biodegradable coating materials.
Alkyd Resins: Alkyd resin, a bio-based material derived from vegetable oils and polybasic acids, could be an interesting option for CRCU coating. Its potential benefits include:

1. Biodegradability: Alkyd resin is biodegradable, making it a sustainable option for CRCU production.
2. Flexibility: Alkyd resin can be formulated to have different properties, such as flexibility and hardness, which could be beneficial for CRCU coating.
3. Adhesion: Alkyd resin has good adhesion properties, which could improve coating uniformity and durability.

Further research is needed to explore the potential of alkyd resin and other underutilized bio-based materials for CRCU production.

By addressing these challenges and exploring new perspectives, researchers and industry professionals can work together to develop more efficient, sustainable, and cost-effective CRCU production methods, promoting sustainable agriculture and reducing environmental pollution.

5.3. Conclusions

- ➢ This review provides a comprehensive analysis of bio-based coating materials, encapsulation methods, and fluidized bed processes for controlled release coated urea fertilizer.
- ➢ Optimizing coating materials and processes is critical for developing efficient and sustainable controlled release coated urea fertilizers.
- ➢ Bio-based coating materials and fluidized bed processes offer a promising approach for promoting sustainable agriculture and reduced environmental impact.
- ➢ Further research is needed to develop scalable and cost-effective coating technologies.

Key challenges need to be addressed, including:

- ➢ Coating uniformity
- ➢ Material selection
- ➢ Scalability
- ➢ Process optimization
- ➢ Environmental impact

Future perspectives can help improve efficiency, sustainability, and cost-effectiveness, including:

- ➢ Developing new bio-based coating materials
- ➢ Advanced fluidized bed technologies
- ➢ Machine learning and optimization
- ➢ Integration with precision agriculture
- ➢ Life cycle assessment

Collaboration between researchers and industry professionals is necessary to develop more efficient, sustainable, and cost-effective production methods, promoting sustainable agriculture and reducing environmental pollution.

CRediT authorship contribution statement

Chigozie Francolins Uzoh: Writing – review & editing, Writing – original draft, Funding acquisition, Conceptualization. **Swantje Pietsch-Braune:** Validation, Project administration. **Lennard Lindmüller:** Visualization, Software. **Stefan Heinrich:** Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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