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Tumbling fluidized-bed process parameters affecting quality of biopolymer coating on surface of pristine urea particles

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ABSTRACT

When pristine urea particles are subjected to spray coating in a fluidized bed, the frequency of substrate particles to come across the spray zone changes due to various hydrodynamic factors of the fluidized bed giving rise to preferential coating. The heterogeneity of coating film on the surface of urea particles results in poor coating quality which ultimately affects the controlled-release properties of the coated product. In this study, the effect of tumbling fluidized bed process parameters is studied on coating quality and the Response Surface Methodology is employed for the optimization of process parameters for better results. In case of coating uniformity w.r.t. coefficient of variance (CV) of coating thickness, atomizing air pressure appears as the most influential parameter. The best coating uniformity in this case is achieved at 2.25 bar of atomizing pressure. The lowest and highest values of atomizing pressure resulted in higher CV of coating thickness. In case of coating uniformity w.r.t. change in coating mass, coating time appears the most influential parameter. Coating mass increases linearly with coating time. For coating uniformity in terms of CV of size distribution, atomizing pressure and fluidizing gas temperature are the most influential parameters. The best coating uniformity is witnessed at 85 °C and 1.90 bar. The optimum values of process parameters and response objectives can be used to scale-up the unit used.

1. Introduction

Pristine urea is highly vulnerable to losses (30–70%) through ammonia volatilization, leaching and surface runoff when applied to soil without engulfment by a barrier layer [1]. Thus, nutrient use efficiency (NUE) of the plants is reduced in addition to environmental pollution through water eutrophication and stratospheric ozone depletion via escape of NH_3, NO, N_2O and N_2 into the atmosphere [2]. To offset these issues, controlled release coated urea (CRCU) is employed that is a purposely designed manure aimed at providing the nutrients in a controlled manner most preferably in synchrony with the metabolic needs of the plants. The CRCU is an environmentally friendly fertilizer that helps to enhance the NUE and eradicate environmental pollution. In addition, the single application of CRCU in one season diminishes the overall cost [3]. The CRCU is produced by the physical intromission of granular urea in an appropriate coating material capable of impeding the spontaneous dissolution and manipulating the nutrient release kinetics as per set standards. Although billions of research dollars have been spent on the development of CRCU, yet the application of CRCU is limited to ornamental and horticultural plants.

Synthetic polymers'-coated controlled-release urea (CRU) is used as an abatement strategy but it offers soil pollution due to non-biodegradability and toxicity of the polymers [2]. Starch is abundantly available naturally occurring polysaccharide polymer which is cheap, biodegradable, renewable, and environmentally friendly [1]. Starch alone, however, is ineffective to be used as coating material for CRF production because of its profound hygroscopic nature, poor mechanical properties and weak dimensional stability [4]. Starch blends with certain appropriate materials can be used to develop efficient control release devices. Borax or di sodium tetraborate can also be used as a crosslinker for the chemical modification of starches [5]. It can enhance the mechanical properties of starch films by crosslinking reaction with the hydroxyl groups of starch [6]. However, it is recommended that the quantity of borax should not exceed 10% in the starch films lest the adverse effect is experienced with respect to the starch film properties [5]. In addition to work as a crosslinker, borax also contributes to the provision of boron which acts as an essential micronutrient for the plants in the soil [7].

When urea is coated in a fluidized bed, a certain degree of heterogeneity of the coating mass or film thickness may arise which distorts the coating quality. This can be attributed to the preferential coating of part of the substrate due to unequal opportunities to pass through the spray zone [8]. The investigation of urea coating quality in terms of coating uniformity (CU) is important due to substantial dependence of the controlled-

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release characteristics on CU. The CU is essential for consistent release profile, good controlled release, core stability, and economics of the coating process [4,9]. In addition, the understanding of the factors affecting CU is helpful as a focal point for the scale-up efforts [10].

Most of studies related to the investigation of CU include glass beads or pharmaceutical tablets as the substrates and CV of coating mass or change in weight gain has been reported as the measure of CU. For instance, Chang et al. [11] studied the effect of coating time on the inter-unit CU of glass beads coated with an aqueous suspension in a pan coater. The effect of tablet size, batch holdup, pan speed, and inclination of rotation axis on CU was studied using oval shaped acetaminophen tablets as the substrate and methacrylic acid as the coating suspension [12]. Another study reported coating of placebo tablets in a rotary pan using Opadry® as the coating suspension [13]. Black Opadry II® was utilized as a coating suspension in another study for the coating of lactose non-pareils in a pan coater as reported by Sahni [14]. A recent study on the investigation of the effect of coating time on inter-tablet coating uniformity has reported that the coating uniformity can be enhanced by longer coating time and optimization of the process parameters. The ibuprofen tablets in this study were coated in a rotating pan coater using a water soluble polymer available as Kallicoat® in the commercial market [9]. All of the aforementioned studies reported CV of coating mass or weight gain as a measure of CU. Some of the authors reported CU in terms of coating mass distribution or coating-per-pass distribution. For instance, CU of glass beads in terms of the coefficient of variance (CV) of coating mass distribution was studied by using aqueous sodium chloride and dextran as the coating suspensions in a tumbling fluidized bed coater [15]. The coating mass was determined by following the same method as described by Chang [11]. Placebo tablets were coated in Wurster-type fluidized bed using an aqueous solution of hydroxypropyl cellulose, hydroxypropylmethyl cellulose, sodium bicarbonate, and a blue dye. The concept of coating-per-pass-distribution was used to study the CU [10]. In another study from the same author, the bioconvex tablets were coated in the drum coater using methacrylic acid copolymer combined with some other additives [16]. The CU was reported in terms of the coating mass variance and CV of coating thickness such that 10 measurements were taken for coating thickness of each cross section of the selected tablets. Shaari et al. [17] determined the CU of placebo tablets coated in a Wurster fluidized bed utilizing Monte Carlo simulations and investigating the coating mass distribution as a measure of the coating uniformity. The ratio of the mass of the coating solution deposited on the surface the pallets to the pallet surface area was reported as a measure of the CU of thickness of pallets coated in a Wurster fluidized bed chamber [18]. The coating dispersion consisted of hydroxypropyl methylcellulose, polyethylene glycol, and tartrazine coloring agent.

To the best of author's knowledge, no studies have so far been reported in literature regarding the use of tumbling fluidized bed coater with tangential spray orientation for the investigation and optimization of the effect of fluidized bed process parameters on CU of borax-modified-starch based CRU (BMS-CRU). This study investigates the effect of fluidizing gas temperature, coating time, and atomizing air pressure on CU of CRU produced in a tumbling fluidized bed using borax-modified starch biopolymer as coating material. The CRCU thus produced would contribute to the agriculture sector in terms of enhancement of the nutrient use efficiency and crops yield, alleviation of pollution through mitigation of volatilization and leaching, and achievement of process economy due to reduced frequency of fertilizer application.

2. Materials & methods

2.1. Materials

Tapioca starch was purchased from the local distributor marketed by Kapal ABC®, Malaysia. It was preserved at −20 °C to avoid any microbial activity. Borax ($Na_2B_4O_7.10H_2O$) was provided by R&M Chemicals®, Malaysia. Borax is used not only as a crosslinker, it's a source boron micronutrient for the plants. Copper chloride ($CuCL_2$) (99.995% pure) was provided by Sigma Aldrich® and used as received. $CuCl_2$ is added in the coating formulation to provide copper as a micronutrient to the plants. Urea particles were provided by PETRONAS Fertilizer Keddah (PFK)®, Malaysia. Urea was sieved and particles of 2.0 mm were used in the coating process.

2.2. Methods

2.2.1. Synthesis of spray formulation

Five grams of Tapioca starch (S) was blended and heated with 100 ml of deionized water at 80 °C for 30 min. 0.25 g of borax (B) was subsequently added and the stirring was continued for 5 min. at 80 °C. Lastly, 0.5 g $CuCl_2$ (C) was added into the starch-borax solution and the agitated for 15 min. at the same temperature. The final solution (SBC) was allowed to cool at room temperature and used as spray formulation.

2.2.2. Urea coating in tumbling fluidized bed coater

The schematic arrangement of the coating process is presented in Fig. 1 and the detailed coating process is explained elsewhere [1]. The peristaltic pump facilitates the transportation of hot coating solution at the tip of tangentially mounted spray nozzle and atomizing air through the two-fluid nozzle makes the spray. The hot fluidizing gas from the bottom of tumbling fluidized bed coater (TFBC) fluidizes urea particles while passing through the annular space between rotor plate and TFBC wall. Coating starts at steady state temperatures and ends with a 10 minute drying session. The controlled release coated urea (BMS-CRU) is then subjected to various tests for the evaluation of CU.

2.2.3. Evaluation of coating uniformity of BMS-CRU

2.2.3.1. CU in terms of CV of coating thickness. Variable pressure Field Emission Scanning Electron Microscope (FESEM) by Zeiss Supra 55 VP® (Germany) was used to determine coating thicknesses of the cross-sections of coated particles. Five particles were randomly picked from each sample and cut with a sharp knife to get the cross sections. The particles' cross sections were mounted on a stainless steel holder with an electrically conductive double sided tape. FESEM micrographs were captured using an accelerating voltage of 15 kV with 500–100,000× magnification. One cross-section was examined under FESEM from 40 equally spaced points (10 points for each quarter) for accurate measurements.

2.2.3.2. CU in terms of coating mass variation. Randomly chosen 100 pristine particle cores were weighed individually on a semi-micro scale. Mass of a single particle was determined from the mean of the 100 cores. After coating the particles in TFBC, 100 coated particles were randomly chosen, completely dried in an air dryer, and weighed on individual basis. The change in mass per particle was recorded and reported as the measure of coating uniformity.

2.2.3.3. CU in terms of CV of size distribution. The size distribution of the population of coated particles is used as another indicator of the coating uniformity. Narrower the size distribution, superior the inter-particle coating uniformity and vice versa. 50 particles were randomly chosen from each BMS-CRU sample and examined their diameters using the ERWEKA TBH325TD (Ottostrasse 20–22, Heusenstamm, Germany). The equipment was first calibrated for diameter measurement. Each particle was subjected to diameter testing for ten times and the mean was reported. The CV in particles' diameter is reported as a measure of coating uniformity.