



Research on permeability coefficient of a polyethylene controlled-release film coating for urea and relevant nutrient release pathways



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ABSTRACT

The permeability coefficient is a key factor that reflects the nutrient release capability of polymer-coated fertilizers. To investigate the permeability coefficient of polyethylene (PE) controlled-release film and to determine the difference between the controlled-release film and a dense membrane, we designed a film permeation device to measure the permeability coefficient of a PE controlled-release film coating for urea, and a mathematical model was used to check the accuracy of these measurements. By measuring the permeation coefficient of a dense, PE membrane, the compactness of the PE controlled-release film was analyzed, and the nutrient release pathway of PE-coated fertilizer was discussed. Research indicated that urea was constantly released through PE controlled-release film and the permeability coefficient remained constant. The permeability coefficient for PE controlled-release film coating on urea with 1–4 months release time was in the range of $7.17\text{--}18.7\text{E-}15\text{ m}^2/\text{s}$ with 2.6 times difference between the maximum and minimum. The permeability coefficient decreased as the release time increased, conforming to the inversely proportional relationship between permeation amount and time in the nutrient release model. It is investigated that the measured values are close to the theoretical values and can be used in model calculation. The urea permeability coefficient of PE dense membrane was $7.11\text{E-}18\text{ m}^2/\text{s}$, which is 1000–2600 times smaller than that of the PE controlled-release film. The contribution of permeability of polymer material itself is negligible. It can be concluded that PE controlled-release film is not a dense membrane but porous and that nutrient release is mainly determined by pore configuration of the film.

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1. Introduction

Polyethylene (PE) controlled-release film coated-urea is a thermoplastic resin-coated fertilizer. PE-coated urea can improve fertilization efficiency, reduce environmental hazards, and balance fertilizer nutrient supply and plant demand, which makes it an environmentally friendly fertilizer [1–3]. The nutrient release rate of coated fertilizer granules is dependent on environmental factors, such as soil moisture, temperature, humidity, and water vapour

permeability and pressure [4–6]. The release rate is also dependent on internal structural factors, such as film properties and structure, solution driving force, and diffusion coefficients [7,8]. Based on Fick's law, assuming that the controlled release coating film is a homogeneous dense film, a controlled nutrient release model can be established [9–11]. The model uses the permeability coefficient (P_s) to evaluate the effects of factors, such as chemical properties and polymer film structure, on nutrient release. Nutrient transport through the controlled release film occurs through a dissolution-diffusion mechanism [12]. Here, the permeability coefficient is defined as the release rate of the nutrients (urea) across the unit film area per unit time (m^2/s). This parameter reflects the degree to which nutrient release from the coated film is controlled. Hence, the determination of the permeability coefficient is important to properly simulate nutrient release and investigate the releasing mechanism.

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List of symbols

| | | | |
|----------------|--|------------|--|
| P_s | urea permeability coefficient of the film, m ² /s | l | thickness of the film, mm |
| v_1 | initial dissolution rate, % | $m(t)$ | urea mass that diffused from the film into the deionised water, μg |
| $v_{\Delta t}$ | differential dissolution rate, % | A | area of the film, mm ² |
| v_t | accumulated nutrient release rate, % | K | slope of the linear relation between time and the urea mass that diffused into the deionised water, $\mu\text{g/s}$ |
| w | total nitrogen content of the coated fertilizer, % | R | correlation coefficient |
| w_1 | dissolute nitrogen content after the first day, % | $M(t)$ | released core nutrient content from a single coated controlled-release fertilizer particle at time t , μg |
| w_t | accumulated nutrient dissolution rate after t days, % | M_0 | core nutrient content, μg |
| t | time, d | r | radius of the core urea particle, mm |
| J_s | permeation flux | ρ_s | density of urea particles, kg/m ³ |
| ΔF | driving force of diffusion process | t' | nutrient release lag, d |
| C_{up} | concentration of saturated urea solution tank, kg/m ³ | t^* | recession time of the nutrient, d |
| C_{down} | concentration of deionised water tank, kg/m ³ | Δt | release time of the coated fertilizer, d |
| C_{sat} | saturated urea concentration, kg/m ³ | | |

The permeability of the coating film to gas or pure liquid is determined by a pervaporation test device [13]. There have been limited studies on film permeability coefficients of polymer-coated fertilizers. Zhang et al. (1994) used a lab-fabricated leaching device [14] to calculate the permeability coefficients of two controlled-release fertilizers (with release times of approximately 15 and 30 days) by measuring the total nutrient released from the coated fertilizers into an eluent per unit time. To obtain a controlled-release film with a large area for the direct measurement of permeability coefficients, Lan et al. (2011) simulated coating conditions and fabricated a controlled-release film on a roller surface covered with urea [15]. Watanabe et al. (2009) measured the permeability coefficient of polyurethane using an in vitro membrane permeation system [16]. However, these studies indirectly measured the permeability coefficient of controlled-release films. Mathematical models and computer simulations can also determine the film permeability coefficient of small-molecule solutes [17]. However, molecular modelling cannot account for the effects of film structure. Studies have shown that the permeability of a controlled-release coating on fertilizers is significantly affected by its porosity [18–20]. Therefore, a direct determination of the permeability coefficient of a controlled-release fertilizer film is required. Additionally, there are three routes of nutrient release through film [21]: Route 1 - If the controlled-release film is a dense film, the nutrient molecules are released through the film by dissolution-diffusion; Route 2 - If the controlled-release film is a porous film, the nutrient molecules are released through micropores based on a fluid flow mechanism; and Route 3 - If the controlled-release film is a loose, porous film, the nutrient molecules are released through the film based on a combination of the two mechanisms of Routes 2 and 3. The nutrient release pathway through the PE-controlled release film determines the nutrient release mechanism and directly influences the nutrient release model. In previous studies, nutrient release models were based on that of Route 1, which assumed a homogeneous dense film [9–11]. However, this hypothesis conflicts with current study results, which seem to indicate porous structures in the controlled-release film. Therefore, an investigation of the nutrient release pathway mechanism is critical to preparing an accurate nutrient release model.

This study focused on two areas. First, a device was designed to directly measure the permeability coefficient of PE controlled-release film coated on urea. The reliability of the method was assessed by studying the correlation between the measured and theoretical values of the permeability coefficient. Second, the

permeability coefficients of urea through PE dense film produced by different processes were measured. The nutrient release pathway of the PE-coated fertilizer was assessed by comparing the urea permeability coefficients of the PE controlled-release film and the PE film and by analysing the compactness of the PE controlled-release film.

2. Materials and methods

2.1. Experiment materials

The test sample was PE-coated urea with a 42% nitrogen content. The coating was performed using a fluidised bed-coating process at the Novel Fertilizer Research Group of the College of Resources and Environmental Science, China Agricultural University. Urea, *para*-dimethylaminobenzaldehyde, diacetyl oxime (analytically pure), and antipyrine (chemically pure) were obtained from Sinopharm Chemical Reagent Co. Ltd. PE-blown film and PE-cast film were prepared by conventional methods and were obtained from Shanghai Packaging Materials Co. Ltd.

2.2. Instruments

The following instruments were used for sample preparation and measurement: a UV spectrophotometer (UV-2201, Germany SHIMADZU); an incubator (PRX-450D, Ningbo Haishu Seif Experimental Instrument Factory); and a micrometre calliper (217–111, Suzhou Suce Electronic Technology Co. Ltd.).

2.3. Experimental device

The urea permeability coefficient measuring apparatus of the PE film is shown in Fig. 1. The apparatus consisted of two parts: a saturated urea solution tank and a deionised water tank. The saturated urea solution tank consisted of a urea loading inlet and a deionised water loading inlet. The deionised water tank included a deionised water loading inlet and a sampling inlet with a cap. A mounting hole at the bottom of deionised water tank was included for a PE tube. On the top of PE tube, a hemispherical film was placed to release the controlled-release fertilizer. The hemispherical controlled-release film was prepared as follows: a sphere was cut in half on a fertilizer cutting platform and repeatedly rinsed in distilled water. A hemispherical controlled-release film was obtained after drying in an incubator at 40 °C for 24 h. The hemispherical controlled-release film was sealed to one end of the PE

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