A Review on Industrial By-products as Materials to Coat Compound Fertilizer

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Abstract
Fertilizers play the most important role in agriculture. The uses of compound fertilizers are common to supply nutrients needed by plant. However, with conventional application, nutrients are easily lost due to various factors. Coated compound fertilizers are started to take place in fertilizer management. Different coating materials and coating techniques are explored to hopefully provide better production and fertilizers efficiency. The main purpose of this paper is to explore technologies and sources of coating materials of compound fertilizers and how do they work.

Keywords: industrial by-products, compound fertilizers, coated compound fertilizers

1. Introduction

Nutrients are necessity for crops to grow and these nutrients are usually available in soil. However, some nutrients may not be enough or not readily available in soil when needed. Therefore, the use of fertilizers is necessary to ensure the nutrients are available for the crops (Finch and Lane, 2014). Nutrients required cab be divided into two groups viz macro and micro elements (Finch and Lane, 2014).

In Malaysia, there is more than 90 percent of fertilizers use for agriculture activity (FAO, 2009). Fertilizers use can be straight fertilizers which only have one of the major elements (usually macro) or compound fertilizers which have more than one element (Finch and Lane, 2014). Usually, the main fertilizers that commonly used in agriculture are urea, ammonium sulphate, calcium ammonium nitrate, phosphate rock, super phosphates, ammonium phosphate, potassium chloride, potassium sulphate and NPK, NP and PK compound fertilizers (FAO, 2009). The use of fertilizers has been increase significantly caused by the rapid growth of the crop production, especially the plantation crops such as oil palm, rubber and cocoa (Goh and Hardter, 2003).

Compound fertilizers like NPK are popular especially in plantations which are used to reduce application frequency, energy and labour cost which are mostly in inorganic form (Yahya et al., 2018; Satriawan and Fitri, 2019). Information such as dosage and type of fertilizer is crucial to help the farmers use them effectively (Tarmizi and Mohd Tayeb, 2006).

Even though fertilizers are used, nutrients applied are usually cannot be uptake by plants and in the end lost to the environment. There was about 40 – 70% of nitrogen, 80 – 90% of phosphorus, and 50 – 70% of potassium is lost with normal application (Wu and Liu, 2008). There are a few ways to reduce this problem. According to Savci (2012), soil analysis should be carried out to choose the right fertilizers. The fertilizers should also be applied at the right time to reduce the loss of energy and finance (Savci, 2012). According to Russel and Williams (1977), fertilizers technology should be able to reduced total energy requirements.

Another way to improve the efficiency for fertilizer application is using slow- or controlled-release fertilizers. These types of fertilizers are usually produced by adding materials that are physically mixed or coat to reduce nutrient release (Jarosiewicz and Tomaszewska, 2003). Coated fertilizers are surrounded by a barrier that prevents the nutrients from releasing rapidly into the environment (Sartain, 2002). Usually, coating materials were chosen based on the ability to modify nutrient release characteristics of fertilizer and they can be organic or inorganic materials. However, some of coating materials have the ability to reduce nutrient release and provide other beneficial element for plant at the same time such as sulphur coated and humic-calcium coated fertilizers (Trenkel, 2010; Sulakhudin et. al., 2010). Some additional materials, such as a combination of soluble plant nutrients, a gel and a thickener were used to form a hydrophilic polymer. This polymer allow water to penetrate in the fertilizer particles and nutrients diffused out gradually (Shavit et al., 1997).

Compound fertilizers can be produced by accretion and agglomeration process (IFDC, 1998). As for coating compound fertilizers, agglomeration process is usually used to attach particles to a surface with or without binders (Pietsch, 2008). A mechanical agitation is usually involved in this process (Salman et al., 2006). Nowadays, coating with by-products from various industries has been studied since these materials have nutrients that are beneficial to crops such as waste materials of slaughter houses, by-products of biodiesel and fertilizers production (Treinyte et al., 2018) or sludge (Welzenbach et al., 2009) and coal combustion by-products (Taulbee et al., 2007).

This paper aims to provide an overview of the coated compound fertilizers with by-products as coating materials. Literature abstracts and full text articles from books, journals, reports, and any relevant publications as well as electronic searches such as Google Scholar, Elsevier, Scopus, and Springer, as well as from other relevant websites, are analysed and included in this review.

1.1 Fertilizers

Plants need nutrients to grow and most of the nutrients came from the soil. When plants take up nutrients, those nutrients are actually removed from the soil with each harvest. These nutrients need to be replenished to ensure there will be enough nutrients for the next planting season. Therefore, fertilizers are needed in order to overcome this matter (Hebebrand, 2015).

Definition of fertilizers are; any materials contain organic or natural, inorganic or synthetic which were applied on or mixed with soil to supply plants with at least 5% of one or more elements to promote normal growth (Nielsson, 1987; Hebebrand, 2015). The fertilizers are available in solid, liquid, and gaseous form (for example, anhydrous ammonia) which can promote plant available nutrients level in soil and enhance plant growth (Savoy, 2010).

The first fertilizer used were recorded about 8000 years ago where European farmers began to use manure as slow release fertilizer to crops since it breaks down slowly and watered their crops to improve yield. It was found that charred cereal and pulse seeds which were taken from 13 Neolithic sites in Europe contain high level of nitrogen-15 (Bogaard et al., 2013).

There are two major type of fertilizers; organic and inorganic fertilizers. The organic fertilizers contain a carbon hydrogen linkage while inorganic fertilizers are usually manufactured fertilizers which contain specific element at certain desired level (Funk, 2014).

1.2 Compound Fertilizers

Compound fertilizers are actually mixture of compounds consist certain percentage of N, P, and K. These types of fertilizers become popular due to availability, easy to manage and low maintenances due to less application for several nutrients compared to straight fertilizers which may need to be applied several times (Homenauth, 2013). There are two major granule formation mechanisms i.e. accretion and agglomeration process.

Accretion process refers to a fluid material is applied to a solid particle layer by layer until desired size, giving the final granule an 'onion-skin'-like structure. Usually, this process used to produce DAPs, MAPs, TSPs, and some nitro-phosphate compounds (IFDC, 1998).

Agglomeration process refers to initial granule formation where solids particles are assembled and attached with agglomerates with a combination of cementing and mechanical interlocking. This process resulting various size, shape, solubility, strength, and surface texture of granules produced. Most granular NPK produced by agglomeration process (IFDC, 1998).

1.3 Controlled Release Fertilizers

Controlled release fertilizers are fertilizer granules concealed within 'barrier' or 'capsule' to control nutrient release to improve nutrients supply to crops and minimise pollution (Ukessays, 2013). The controlled release fertilizers provide nutrients at slower rate to crops. Therefore, crops will be able to uptake nutrient slowly through time without waste and improve fertilizer use efficiency (Trinh & KuShaari, 2016; Subbarao et al., 2013). Controlled release fertilizers are dependent on the thickness of formulated coatings to extend the nutrients release leading to an increased control over the rate and pattern of release (England et al., 2012).

In Official Publication 50 (1997) published by American Plant Food Control Officials (AAPFCO) stated that there was no official differentiation between slow-release and controlled-release fertilizers (Trenkel, 2010). Slow-released or controlled-released fertilizer is a type of fertilizer which has the ability of delaying nutrient release after application. This ability may occur by variation of mechanisms include semi-permeable coatings, sealant, or chemical materials to controlled water solubility (Trenkel, 2010). According to Shaviv (2005) as cited by Trenkel (2010), there are three types of slow-released and controlled-release fertilizer viz. organic-N low-solubility compounds, inorganic low-solubility compounds, and fertilizers in which a physical barrier controls the release.

Organic-N low-solubility compounds fertilizer can be divided into biological compounds and chemically decomposing compounds. The biological compounds are usually based on urea-aldehyde concentrated products such as urea-triazone, crotonylidene diurea, and urea-formaldehyde. The example of chemically decomposing compounds product is isobutylidene-diurea (Trenkel, 2010).

Inorganic low-solubility compounds fertilizer includes fertilizers product such as metal ammonium phosphates, for example magnesium ammonium phosphate, and partially acidulated phosphate rock. In addition, the biologically and microbially decomposed nitrogen products are commonly referred to in the trade as slow release fertilizers while encapsulated or occluded coated or products as controlled-release fertilizers (Trenkel, 2010).

Physical barrier controls fertilizer can be divided into two which are; fertilizer covered with matrices and fertilizer covered with coat. The matrices made by soluble active material is spread is spread in certain range and limits the dissolution of the fertilizer. The materials used to prepare matrices can be divided into hydrophobic materials, for example polyefines, and gel-forming polymers also known as hydrogel which can reduce the solubility of fertilizer by swelling capability (Trenkel, 2010).

Fertilizer covered with coat is also known as coated fertilizer. The coating materials of coated fertilizer are usually made from hydrophobic polymers that prevent them from releasing rapidly into the environment (Sartain, 2002; Trenkel, 2010). The coated fertilizers have several advantages. Some coated products offer a cheaper ways to exploit slow-release characteristics, for example sulphur-coated fertilizer. Sulphur-coated technology was developed since 1960s by National Fertilizer Development and sulphur was chosen due to its value as nutrient for plant and its low cost (Sartain, 2002).

The coated fertilizers are also may offer certain release characteristics depending on certain conditions (Sartain, 2002), for example, polymer/sulphur-coated fertilizers (PSCF). The nutrient release of PSCF is a combination of diffusion and capillary action due to its hard polymer outer layer. Water must diffuse through the polymer coating where the rates of diffusion are depending on the thickness and composition of the polymer. Then, at the sulphur/polymer interface, the water infiltrates through capillary action and dissolves the fertilizer core. After that, the dissolved nutrient of fertilizer penetrates out through the coating (Sartain, 2002). Generally, the use of coated fertilizer is more popular compared to matrices. There are two types of coated fertilizer viz. fertilizer with organic polymer-coatings and fertilizer coated with inorganic material or mineral-based coatings (Trenkel, 2010; Ali and Danafar, 2015).

1.4 Coated Fertilizers

1.4.1 What are Coated Fertilizers?

Coated fertilizers are actually a type of controlled release fertilizers where soluble fertilizers such as urea or NPK were coated with other materials which are whether semipermeable, semipermeable, or water insoluble after granulation, prilling or crystallization. These materials acted as physical barrier to control nutrient release through the coating by diffusion (Ali and Danafar, 2015). There is wide variety of materials used to coat granular fertilizers; from natural to synthetic. Basically, these coating materials can be divided into two main categories; non-organic and organic coatings (Trenkel, 2010; Ali and Danafar, 2015).

Non-organic materials that commonly used are sulphur, vermiculite or cement, silicate, and phosphates compounds such as sulphur, silicates, and phosphates such as attapulgite and phosphogypsum (Wu and Liu, 2008; Chien et al 2009; Hua et al., 2009; Ni et al., 2010; Ali and Danafar, 2015). Organic coatings consist of natural and synthetic polymers to coat fertilizers such as polyesters, polyethylene, polyoefin, resins, pine lignin, polyacrylamide, ethyl-cellulose, latex, wax, rubber, guar gum, fatty acid salts (for example castereate in Multicote® which is stearic acid and calcium hydroxide), and many more (Wu and Liu, 2008; Chien et al., 2009; Hua et al., 2009; Ni et al., 2010; Ali and Danafar, 2015).

The production of coated fertilizers and the coating technologies are varied, depending on coating materials and types of coating process used (Ali and Danafar, 2015). Basically, there are two types of polymer coatings; thermoset resin and thermoplastic polymer (Jacobs, 2004; Lu et al., 2013).

There is lack of study on thermoset resin as coating materials (Lu et al., 2013). However, there is commercialized fertilizer with thermoset resin coating namely Osmocote® (Scott-Sierra, Marysville, OH). A granular fertilizer which is soluble coated with a thermoset copolymer of dicyclopentadiene and a glycerol ester (linseed oil) dissolved in an aliphatic hydrocarbon solvent and the coating is applied in layers where thickness of coating determines the nutrient release pattern. The Osmocote® durability range from 3 to 16 months (Jacobs, 2004). Other examples of thermoset resins are epoxy resin and unsaturated polyester resin where these resins were sprayed directly onto the fertilizer granules. Lu et al. (2013) used these resins to study the coating structure and nutrient release of fertilizers.

Thermoplastic polymers or resins are highly impermeable to water. Therefore these types of resins are widely used to coat fertilizer granules (Jacobs, 2004). Examples of thermoplastics resins are poly(vinylacetal), polypropylene, low density polythene, and ethylene or carbon monoxide copolymer (Ge, 2002). Commercialized fertilizer with thermoplastic polymers is Nutricote® (Chisso-Asahi Fertilizer Company, Ltd, Tokyo, Japan). The Nutricote® uses polyolefin, poly[vinylidene chloride], and copolymers as coating materials. Other than that, ethylene-vinyl acetate and surfactants are also added as controlling agents to achieve desired diffusion characteristic. The Nutricote® is designed to control the effect of temperature on nutrient release pattern by spreading mineral fillers into the coating (Jacobs, 2004).

1.4.2 Types of Coated Fertilizers

1.4.2.1 Coated Fertilizers with Non-Organic Materials

The most common coating material for coated fertilizer especially urea is sulfur. Sulfur is a secondary plant nutrient, low cost material and can slowly degrades by microbial, chemical and physical processes (Trenkel, 1997). Sulfur-coated urea was developed through research conducted by Tennessee Valley Authority (TVA) in 1960s (Young (1974), Tennessee Valley Authority (1978), and Landels et al., 1981 as cited by Hays, 1987). Until now, sulfur is used in the coating formulation along with other material and sulfur coated fertilizer has been used as a bench mark for analyses of coated fertilizer (Ibrahim et al., 2014).

Sulfur coating appears to have low wettability and adhesion to the urea substrate due to its inherently increased surface tension (Tsai, 1986). However, the sulfur-only coating is not an effective sealant and requires additional conditioning materials that become vital for its application to fertilizer granules which poses economic constraints. Sulfur shells left in the soil are not immediately integrated. Therefore, an excessive amount of sulfur may build up and react with water to acidify the soil (Detrick, 1997).

Sulfur-coated is prepared by coating preheated urea cores with liquefied sulfur which was melted at about 156ºC and wax (as sealant) is sprayed to conceal cracks or gaps on the sulfur coating surface. Finally, a conditioner layer (usually attapulgite) is applied. Usually, sulfur-coated urea contains about 31 to 38% of nitrogen (Allen et al. (1971), MaClellan and Scheib (1973), Oertli (1974) & Jarrell and Boersma (1980) as cited by Shaviv, 2001; Ali and Danafar, 2015). Usually, durability of sulfur-coated urea is about 2 to 4 months (McNabb and Heser, 1997).

Polymer sulfur-coated urea (PSCU) was developed to enhance the quality of sulfur-coated urea fertilizer where sulfur as the first layer and polymeric material as the second layer to improve attrition resistance of the fertilizer. Therefore, the nutrients release rates can be improved. (Ali and Danafar, 2015). The PSCU was significantly reduced nitrous oxide and ammonia volatilazation loss (Svedin et al., 2015).

1.4.2.2 Coated Fertilizers with Organic Material

A wide range of organic polymers have been investigated as coating material for fertilizer such as polyolefin, polyurethane, polyethylene, polyesters, lignin, lignocelluloses, and many more. Other than to minimize the nutrients losses from fertilizer, organic materials are chosen because they are biodegradable, non-toxic, and offers stable coating without unintentional effects on the environment (Junejo et al., 2011; Ali and Danafar, 2015). There are two major types of organic polymers namely hydrogels and superabsorbents (Ali and Danafar, 2015).

Hydrogel polymers have been used in biomedicine, bioengineering, agriculture, and many other fields. It has water-swollen materials with a three-dimensional structure. Nutrient release from fertilizer with hydrogel-based coating is controlled by wetting and swelling of the hydrophilic polymer and nutrient dissolution rate (Shavit, 2003; Berger, 2004; Ali and Danafar, 2015). There are two methods to use hydrogel coating. The first method is by allowing dry gel to swell in compound solution until swelling equilibrium is achieved and after that, the final product is dried and ready to be used. The second method is by loading the compound in the gel matrix by adding it to the reaction mixture and polymerized in situ where the compound is entrapped in the gel matrix (Ward and Peppas, 2001).

Superabsorbents polymer is highly water-swellable which has three-dimensional polymeric networks. Therefore, large volume of water can be absorbed and retained in it. Superabsorbents polymer has been used in various fields such as agriculture, horticulture, wastewater treatment and et cetera. The usage of superabsorbents with fertilizer showed an effective way in utilization efficiency of water and fertilizers (Wu and Liu, 2008; Ali and Danafar, 2015). Liu et al. (2006) found that the use of slow release and superabsorbent nitrogen fertilizer (synthesized through the copolymerization of acrylic acid and maleic anhydride in the presence of urea) could absorb, preserve soil moisture and has slow nutrient release properties. They found that water evaporation of the soil with this fertilizer were lower (between 24.0 to 54.8%) compared to the soil without it (between 28 to 60.0%).

1.5 Coated Compound Fertilizers

Polymer-coated compound fertilizers are the latest sophisticated fertilizers where multi-nutrients core surrounded by polymer coating. Nutrients release of these types of fertilizers is controlled by thickness and chemical composition of coating (Thomas et al., 2009).

There were studies conducted on compound fertilizers coated with polymers. Tomaszewska et al. (2002) used polysulfone, cellulose acetate and polyacrylonitrile to coat commercial granular NPK 6-20-30 fertilizer using phase inversion technique to study the physical and chemical characteristics of these and products. The phase inversion technique (wet method) is polymer is precipitated (a solid is created from a solution) from its solution which is mixed with a non-solvent material and form a coating on solid particle. This technique was used to encapsulate fertilizers to create controlled release characteristics (Chanda and Roy, 2006).

Tzika et al. (2003) used an aqueous latex (a film-forming material with 55.12% of dry solid content and dynamic viscosity ≈ 14cP at 20ºC) of a vinylidene-chloride-based copolymer to coat a commercial fertilizer consisted of a mixture of $(NH_4)_2SO_4$, $NH_4H_2PO_4$, NH_4NO_3 and $CaSO_4$ that contained 20% N and 20% P in w/w. This fertilizer was coated using Wurster fluidized bed to investigate the morphology and release characteristics of the coated fertilizer.

Tomaszewska and Jarosiewicz (2004) studied the possibility of using enhanced degradability coating using polysulfone-starch mixture. In their study, granular NPK 6-20-30 fertilizer was coated with the mixture using phase inversion technique. Wu and Liu (2008) used chitosan, poly(acrylic acid-co-acrylamide) superabsorbent polymer to coat industrial grade of NPK using rotary drum to investigate nutrient release and water retention properties.

There are some polymer-coated fertilizer which already been commercialized and had been used in forest and conservation nurseries in North America. There are; Osmocote®, Apex®, Difussion®, and Multicote®. The Osmocote® (Scott-Sierra, Marysville, OH) is one of the early inventions of polymer-coated controlled release fertilizer. It has polymeric resin (dicyclopentadiene, glycerol ester, and aliphatic hydrocarbon solvent) which is applied in a few layers. The Apex® (J.R. Simplot, Boise, ID) has polyurethane membrane coating applied over the core using a continuous coating drum. Difussion® fertilizer produced by Green Valley Agricultural, Caledonia, MI, was produced in many nutrient formulations and customized for different temperature zones. Multicote® (Sun Gro Horticulture, Bellevue, WA) were produced by special release-controlling agents mixed with thermoplastic resin coatings to enhance longevity of fertilizers Landis and Dumroese (2009).

2. Materials and Methods for Coated Compound Fertilizers Production

2.1 Coating Process

The coating process involves the covering of particulate materials including seeds, agglomerates, pellets and powders with a surrounding layer of a coating agent (or coating material) (Saleh and Guigon, 2006). Agglomeration is the key in mechanism of coating growth of bound powder around fertilizer pellets. Agglomeration is a process of attaching of particles to one another or to a solid surface and this process is needed to coat fertilizer. Usually, binders are needed to complete this process but sometimes the particles themselves can stick together with a special condition (Pietsch, 2008). The function of binder is to assist in the agglomeration process and there are available in many types, from starches, to waxes or even plastics. Binders are chosen depend on the desire characteristics, cost, and availability. Some of characteristics which binders should have are; help the actual agglomeration process, improve crushing, and green strength of the end products (Carlson and Kozicki, 2016). Examples of binders used sodium alginate, sodium silicate, kaolin clay.

Sodium alginate is a natural polysaccharide product which is extracted from kelps or brown seaweed. It is white to yellowish-white powder and almost odourless. Sodium alginate will turn into gel when calcium ions presence (Clegg, 2014). In agriculture, there were studies where sodium alginate was used as a binder for slow release fertilizer. Biochar encapsulated within polymeric alginate beads was able to reduce phosphorus loses by leaching (Domingues et al., 2014). Hydrogel nanocomposite based on sodium alginate-g-poly (acrylic acid-co-acrylamide) with the presence of zeolite clinoptilolite has good water adsorption capacity and slow release fertilizer property (Rashidzadeh et al., 2014). Alginate-based fertilizer can preserve nitrogen for a longer time compared to commercial fertilizer (Kay, 2004).

Sodium silicate is produced by combining silicon oxide with sodium carbonate at the temperature 1100-1200°C. Sodium silicate is also known as 'waterglass' when in liquid form and looks like 'glass' when in solid form. Dissolved and hydrous powder of sodium silicate is widely used for agglomeration application. It is used as a cement accelerator since it reacts quickly with calcium (McDonald and Thompson, 2003). Sodium silicate can be useful in agriculture. The production of perennial ryegrass (*Lolium perenne*) which was grown in an acidic allophanic soil was increase up to 5% when the combination of poly-carboxylic acids (AlpHaTM) and sodium polysilicate applied (Bishop et al., 2012).

Kaolin was origin from China and it is named after a hill called Kao-ling, the mining area. Kaolin clay or kaolinite is a soft clay mineral. It is actually hydrated aluminium silicate ($Al_2(Si_2O_5)(OH)_4$) and mainly white in colour. It is widely used in ceramics, cosmetics, paint, paper making and other products (Encyclopædia Britannica, 2015). There studies where kaolin clay was used as a material for slow release fertilizer. Urea-intercalated with kaolin clay granules performed as slow release fertilizer where it reduced the urea release ratio (Mahdavi et al., 2014). Higher grain yield and yield components of rice were recorded when encapsulated urea-kaolinite controlled release fertilizer applied (Roshanravan et al., 2014).

Basically, coating process is conducted to achieve one or more objectives such as to improve appearance, to stabilize the particle surface, to prevent caking during storage and transportation, to delay and/or control the release of active agents involved in core particles, and so on. Generally, there two types of coating technologies; mechanical agitation and pneumatic solid mixing (Salman et al., 2006).

The mechanical agitation is actually a shaking process with a machine. The most common mechanical agitation coating technologies are drums (Figure 1) and pans coater (Figure 2). Usually, granular fertilizers with or without coating material will be placed in drum or pan coater. After that, water or coating solution will be sprayed onto the granules (Salman et al., 2006).

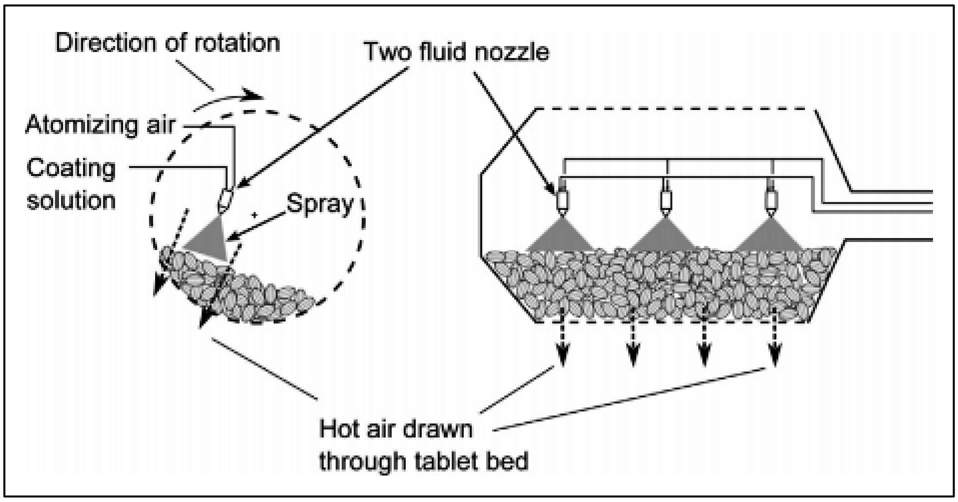

Figure 1: Typical drum coater (Toschkoff and Khinast, 2013)

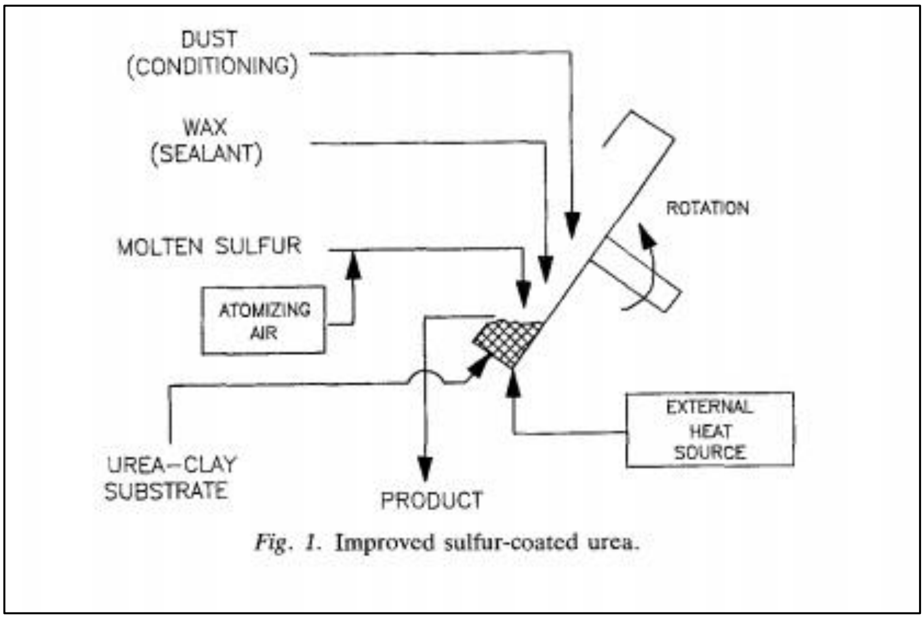

Figure 2: Typical pan coater (Gullett, et al., 1991)

The pneumatic solid mixing is actually a process containing or operated by air or gas under pressure. The most common coater in this category is fluidized-bed and there are three common type of fluidized-bed (top spray, bottom spray, and tangential spray (Figure 3). In fluid bed, fertilizer particles will be placed in a close chamber and hot air will be blown from below. After that, coating liquid will be sprayed onto the particles (Salman et al., 2006).

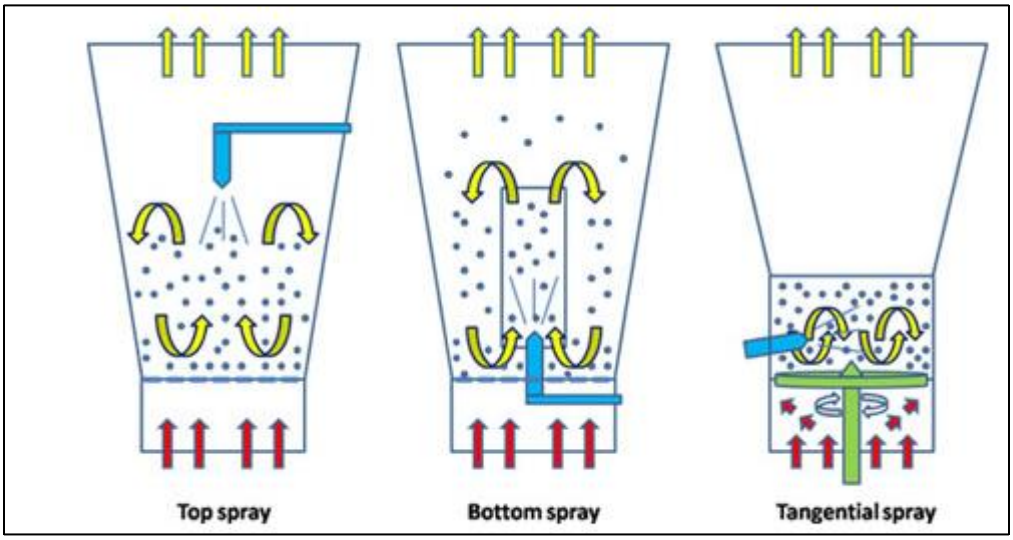

Figure 3: Typical fluidized-bed coating (Colorcon, 2015)

2.2 Combination of Industrial By-products and other Materials to Coat Compound Fertilizer

Industrial by-products are the unwanted materials generated by industrial activities. Although these materials are considered as waste, some of them have certain element that might be useful for other activities including agriculture. Therefore, these by-products can be recycled for other purposes (Agardy et al., 2002).

Industrial by-products disposal has become a serious issue for every municipal government throughout the nation. As available landfill space decreases and the cost of siting and building new landfill increases, local authorities are struggling to develop alternative means of meeting the waste disposal challenge. Landfilling is the most widely used method for solid waste disposal in developing countries. Landfills have created various environmental problems such as emissions and leachate. The public has become more aware of landfill issues such as increasing concern on the groundwater contamination, potential release of toxic gases and odour (Saheri et al., 2009).

Recycling of industrial by-products as potential materials to coat fertilizer is an excellent alternative to reduce landfilling solid waste disposal. Characteristics of industrial by-products sludge make them a potential coating material for compound fertilizer (Welzenbach et al., 2009). Example of industrial by-product is red gypsum, a by-product from the extraction of titanium. It is semi-solid mud form and reddish brown in colour, produced during white titanium dioxide production. It mainly comprises of 70% of calcium sulphate, 30% of ferum oxide and small amount of aluminium oxide (Chai et al., 2003).

Red gypsum usually was disposed of outside the titanium dioxide plant. In Malaysia, red gypsum was produced about 340,000 tonnes annually (Kamarudin and Zakaria, 2007).

Studies have been conducted in utilizing red gypsum in agriculture. With the high rates of application (more than 2.5%), red gypsum may has significant influence to the soil pH (Nur Hanani et al., 2009). Soil amendment with high ferum industrial by-product such as red gypsum could be used of soils co-contaminated with heavy metals and anionic metalloids (Lombi et al., 2004). Red gypsum was also can be co-utilized with sewage sludge with recommended combination rate of 2.5% red gypsum and 5% sewage sludge for corn cultivation. However, the requirement of P and K of corn need to be observed due to high pH of red gypsum (Fauziah et al., 2011).

Another industrial by-product that has potential as coating material is steelmaking slag. Steelmaking slag is a by-product produced during iron and crude steel production and the steel industry. In Malaysia, about 8 million tons of steelmaking is being produced annually (SEAISI, 2012) and 10-15% of by-product is being generated also known as slag (Reddy et al., 2006). Currently in Malaysia, this steelmaking slag is being disposed in landfill and no utilization made (Lion Group, 2010). In order to utilize steelmaking slag to reduce landfill, it was suggested that this industrial by-product need to be used in different fields of application, for example in agriculture based on its properties (World Steel, 2008). Steelmaking slag contains calcium, silicon, magnesium, mangan, ferum, phosphorus and other elements which made it suitable to be used as fertilizer (Table 1).

Table 1: Typical compositions of steelmaking slag (Ito, 2015)

Compositions	CaO	SiO_2	MgO	MnO	Fe	Al_2O_3	P_2O_5
%	45.8	11.0	6.5	5.3	17.4	1.9	1.7

A number of studies about the use of steelmaking slag in agriculture had been conducted since it had fertilizer effects. Steelmaking slag has a liming effect which may increase crop yield (Younger, 1994). It also has other nutrients such as Mg (6.5%), Ca (24%), Fe (26%), Mn (1.2%), and Zn (0.1%) which can improve soil fertility. However, the amount of nutrients can vary from one batch of steelmaking slag to another (Mitchell, 2004).

Paper mill sludge also has the potential to be a coating material for fertilizer. Paper mill sludge was generated from the effluent treatment during paper production which was up to 23.4% per unit of produced paper (Miner, 1991). In Malaysia, there was about 1,000,000 metric tonnes of recycled paper mill sludge produced annually from 67 paper mills (Rosazlin et al., 2010).The benefits of land application of paper mill sludge as fertilizer was reported in numerous studies where it increased tree growth, crop nutrition, and improved soil physical conditions (Bellamy et al., 1995; Jackson et al., 2000). It is a source organic matter and contains several essential plant elements that may contain beneficial nutrients including nitrogen, phosphorus, potassium, calcium and magnesium (Feldkirchner et al., 2003).

Table 2: Physico-chemical characteristics of paper mill sludges of Muda Paper Mills Sdn. Bhd. (Rosazlin et al., 2015)

No.	Parameters	
1.	Moisture (%)	69.32
2.	pH	6.45
3.	EC, mS/cm	1.12
4.	Nitrogen, %	1.29
5.	Carbon, %	24.39
6.	C/N ratio	18.91
7.	Organic matter, %	41.95
8.	CEC, $cmol_{(+)}\ kg^{-1}$	25.00
9.	Phosphorous, %	0.07
10.	Potassium, %	0.13
11.	Calcium, %	0.54
12.	Magnesium, %	0.73
13.	Sodium, %	0.69
14.	Aluminium, %	1.65

Paper mill sludge can be used as a fertilizer but the uptake and heavy metals residue in soil should be monitored to avoid potential pollution (Rosazlin et al., 2015). The early growth of both forest tree (*Khaya senegalensis*) and medicinal shrub (*Orthosiphon stamineus*) were increased when treated with composted paper mill sludge and raw paper mill sludge (Rosazlin et al., 2015).

There is lack of study on powdered industrial by-products as coating material for fertilizer. Some of industrial by products such as stone dressing dust, fuel ash, leather dust, and coconut fiber were used as trial materials to prevent caking by coating them to dry mixtures compound fertilizer. These materials were finely ground (dust) and fed into rotating drum filled with fertilizer particles. The idea was to interpose inert material between particles as anti-caking agent in a very fine state of subdivision by rolled the fertilizer particle in the dust. The dust may be applied alone or mixed with binder which may be sprayed before or after dusting (Lowrison 1989).

Taulbee et al. (2007) used four types of coal combustion by-products (CCBs) to coat ammonium nitrate (AN) fertilizer. There were fly-ash C, fly-ash F, FGD (flue gas desulfurization) ash, and AFBC (Atmospheric Fluidized Bed Combustion (AFBC) ash. In this study, there were two methods used to coat AN prills; disk pressure and drum rolling with two different speed (70 and 106 rpm) at three rolling time (5, 10, and 20 minutes). There were three techniques used; CCBs only, CCBs with water addition, and CCBs with secondary binder agents (starch, guar, gum, bentonite, and sodium silicate).

Taulbee et al. (2007) reported that the drum rolling method produced better coated AN compared to disk pelletization. This study revealed that more durable coatings were produced at higher drum roller speed (106 rpm) and at shortest time (5 minutes). When water was applied, coating efficiency of CCBs improved except for AFBC and the addition of small amount of bentonite improved the particle size distribution of FAC sample.

The particle size of powder material must be small enough (often < 1mm) to allow the adherence of the coating layer on the substrate is guaranteed by Van der Waals forces or by electrostatic forces. Dry coating process is used for coating of powder paints or some mineral powders to improve their flowability (Saleh and Guigon, 2006).

2.3 Mechanism of Controlled Released with Industrial By-products

According to Liu (2008) and Shaviv (2005) as cited by Azeem et al. (2014), a nutrient release mechanism for coated fertilizer is known as multistage diffusion model. When water penetrates into coated fertilizers, the water will condense on the core and caused osmotic pressure due to partial nutrient dissolution. Therefore, the fertilizer will swell and causes mechanism of nutrient release; failure mechanism and diffusion mechanism.

The failure mechanism, also known as catastrophic release occurs when this osmotic pressure exceeds threshold membrane resistance, the coating will crack and nutrients inside core will suddenly be released. This mechanism usually happens on weaker or more fragile coatings such as sulphur or modified sulphur. The diffusion mechanism occurs when the coating is able to endure the osmotic pressure (within the fertilizer) and nutrients release slowly via diffusion caused by concentration or pressure gradient (outside the fertilizer), or both. This mechanism usually happens on polymer coatings (Azeem et al., 2014). There are many other factors affect controlled release mechanism such as ambient temperature, ambient moisture, types of controlled release fertilizers and many more (Rose, 2002).

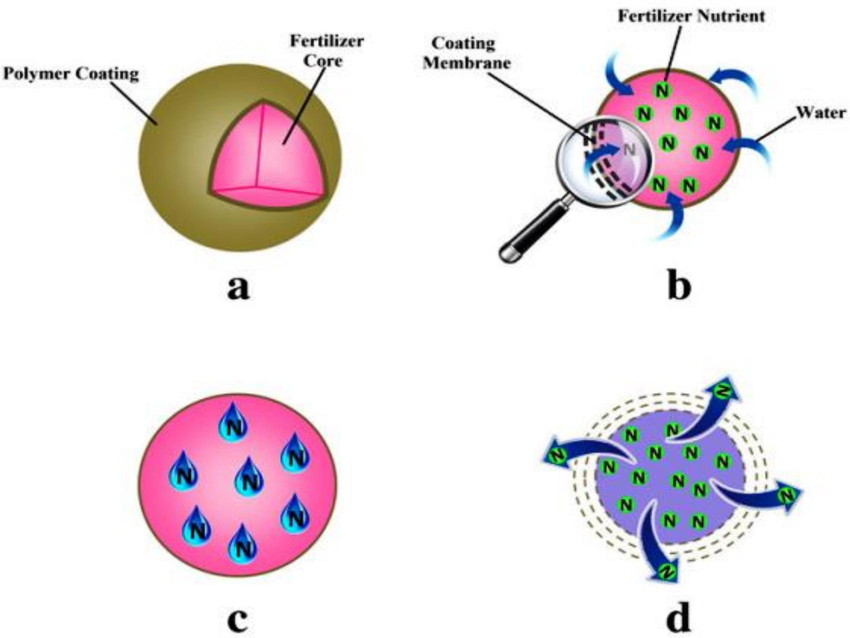

Figure 4: Diffusion mechanism of coated fertilizer

(a) Fertilizer core and coating (b) Water enters the fertilizer (c) Nutrient dissolution and osmotic pressure develops, (d) Nutrient releases through swollen coating membrane

(Azeem et al., 2014)

Generally, the nutrient release pattern of controlled release fertilizers is affected by factors such as quality of coating (surface and thickness), coating process, and type of coating (Hanafi et al., 2000; Tzika et al., 2003). Cracks on the surface and thin coating cause in high nutrient release rates. Therefore, smooth surface and thick coating result in controlled nutrient release (Hanafi et al., 2000; Chien et al., 2009).

2.4 Advantages and Disadvantages

There are advantages and disadvantages of controlled release fertilizers when using industrial by-products as coating materials. One of the advantages of using controlled release fertilizer is lower cost of fertilizer management. Fertilizer management includes frequent of application and labour. When lesser fertilizers applied, the use of labour is also been reduced.

The other advantage is preventing nutrient loss. Nutrients solubility from controlled release fertilizers is slower due to its coating. Therefore, plants are able to uptake the nutrients in time and reduce waste. It is also able to prevent plants tissues from burning due to 'unnecessary' extra application of fertilizers and improve germination rates (Rose, 2002).

One of the disadvantages of coated fertilizers is; they are expensive which made some of these fertilizers cannot be commercialize. This is due to high production cost and raw materials (example; bentonite) (Halt and Kawatra, 2013). Other than that, there are some of the coating materials are not biodegradable and toxic to the environment (example; cement) (Azeem et al., 2014; Carlson and Kozicki, 2016).

The other disadvantages are; the release pattern of coated fertilizers is uncertain especially in field application. Some of the coated fertilizers may change the pH of soil, sensitive to ambient moisture, temperature and bioactivity of soil. Other than that, coated fertilizers are also need to be stored carefully and storage facilities may need to be modified to prevent pre-mature nutrient release (Rose, 2002).

3. Conclusion

The coating of compound fertilizer is needed to avoid nutrient loss. Several of coating materials has been used to produce coated compound fertilizer. However, the use of industrial by-products is still lacking and more study need to be done since industrial by-products might have heavy metals which can pollute environment. The choice of binders and coating method also play an important role depending on coating materials used to produce good quality of coated compound fertilizers.

4. References

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