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Research article

A review of synthetic and earth's resource-based slow-release fertilizers and their potential role in reducing groundwater pollution

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ABSTRACT Groundwater is an essential water source for humans. Unfortunately, some groundwater resources have been contaminated by wastes from various sectors. Fertilizer also contributes to groundwater contamination. The nutrients from fertilizer that are not absorbed by the plant move towards groundwater reservoirs and contaminate the groundwater. Slow-release fertilizer is a solution to reduce groundwater pollution while maintaining fertilizer's function as the sources of nutrients for plants. This paper discusses and reviews the relevant literatures related to groundwater, fertilizer, slow-release fertilizer, and earth's based slow-release fertilizer. The release rate of nutrient elements from different types of fertilizer is also presented. The synthetic base slow-release fertilizers and earth's resource-based slow-release fertilizers can hold nutrients longer than conventional chemical fertilizers when exposed to water. Two particular raw materials that can be used for slow-release fertilizer synthesis are hydrothermal silica and obsidian which shows the presence of amorphous silica and some other minerals in their XRD analysis result. Silica in compacted slow-release fertilizer can control the release rate of nutrient elements from fertilizer.

INTRODUCTION

Groundwater is the most crucial source of fresh water. People worldwide rely on groundwater to fulfill water supply for various needs (International Association of Hydrogeologists, 2020). Unfortunately, various activities nowadays contaminate the groundwater. It is known that household, agriculture, livestock, and industrial waste have contributed to groundwater pollution. Groundwater pollution due to the use of conventional chemical fertilizers is an issue of global concern today (Talabi and Kayode, 2019). Some nutrient elements from fertilizer that are not completely absorbed by plants migrate to the groundwater and eventually contaminate it (Swantomo et al., 2014; Wang et al., 2015). On the other hand, fertilizer is beneficial for increasing agriculture productivity and maintaining food supply globally.

The increase in population has to be balanced by an increase in food production. The effort to handle pollution caused by fertilizer through slow-release fertilizer is worth to be investigated. The application of slow-release fertilizer reduces groundwater pollution while maintaining agriculture productivity

(Bijay-Singh and Craswell, 2021). The slow-release fertilizer has the potential to maintain crop productivity while also reducing groundwater pollution; therefore, the review of groundwater pollution and the development of slow-release fertilizer is needed. This study aims to review the importance of fertilizer, the necessity of slow-release fertilizer, and the utilization of earth's material in slow-release fertilizer synthesis in order to reduce groundwater pollution without decreasing agriculture productivity.

METHODOLOGY

This study evaluated literatures relevant to groundwater, fertilizer, slow-release fertilizer, and earth's based slow-release fertilizer. In addition, an analysis of minerals for slow-release fertilizer were conducted. The analysis of minerals started with grounding and milling the samples using a jaw crusher and a ball mill. The jaw crusher blade was made of manganese steel, while the vial and ball used in the ball mill were made of stainless steel. The milled mineral was rinsed in distilled water and dried at 80 ^{0}C for 12 hours. After drying, the milled mineral was screened to obtained a 200-mesh (74 microns) powder. The powder was then irradiated with CuK-alpha radiation at a sampling pitch and a scanning speed of 0.02 degrees and 2.4 degrees/minutes, using X-Ray Diffraction (XRD) equipment (Shimadzu 7000, Japan).

GROUNDWATER POLLUTION

Groundwater is the water that occupies all the void within a geologic stratum. Based on the water saturation level in the pores, it is commonly classified into three zones; saturated zone, unsaturated zone, and aeration zone (Todd, 2005). A saturated groundwater zone is essential for water supply development, engineering work, and geological research. Groundwater utilization dates back to ancient times through vertical and horizontal wells (Voudouris et al., 2018). Nowadays, horizontal wells can still be found in Southwestern Asia and North Africa countries, with Iran having the largest number of 22,000 horizontal wells. The horizontal wells in Iran supply 75% of the water used for the households (Todd, 2005). In recent centuries, the increasing industry and other engineering projects require large-scale of groundwater. As a result, the utilization of groundwater has rapidly increased. According to Giordano (2009), 95-96% of water consumed in modern society is from groundwater, whereas the rest are obtained from surface water and soil moisture. The high dependency on groundwater requires its freshness and cleaness. Unfortunately, groundwater is vulnerable to both anthropogenic and geogenic pollution. Based on the community characteristic, the anthropogenic pollution can be classified into: residential, municipal, commercial, industrial, and agriculture (Stefanakis, 2009).

Various wastes from many sources has been regulated after the government and society realized that pollution are harmful to people. The first regulation preventing pollution was initiated in 1970, when a company, namely Hooker Chemical Company, dumped 22,000 tons of different organic chemicals wastes, including dioxin, PCBs, and pesticides, into excavated canal. The wastes eventually migrated and contaminated the area near the canal causing the relocation of 950 families from the area (Fitss, 2002). The law that regulates wastes from industries was issued after this incident.

In addition to the industries, the agriculture sector has also contributed to groundwater pollution due to excessive fertilizer application (Malki et al., 2017; Solihin et al., 2020; Malik, 2022). In contrast to industrial waste that can be easily identified as waste and therefore be regulated by environmental laws, the pollution caused by fertilizer problem cannot be quantified and regulated. Fertilizer has been known as the substance supporting the agricultural sector and never been considered as a component that could potentially harm the environment.

THE ADVANTAGE AND DISADVANTAGE OF FERTILIZER

Fertilizer plays a vital role in maintaining agriculture productivity, which is one of the essential factors in saving the population. Human population on earth has increased significantly in 69 years (1950-2019), growing from 2.5 to 7.7 billion and predicted up to 10.9 billion at the end of the century (United Nations, 2019). Indonesia's population has also increased significantly from 69.5 million in 1950 to 277.88 million in 2021 (Worldmeter, 2020). The increase in population is a challenge for the agricultural sector to provide adequate food and nutrition. Therefore, agriculture plays a leading role of in supplying food for a country's population (Food and Agriculture Organization, 2000).

The increase in crop productivity is necessary to balance the rapid growth of the world population. The best way to achieve high agricultural productivity is by using fertilizer. Food and Agriculture Organization (2000), which has assisted farmers in 40 countries for 25 years, reported that the weight of wheat of a plant with fertilizer was 60% higher than the one without fertilizer.

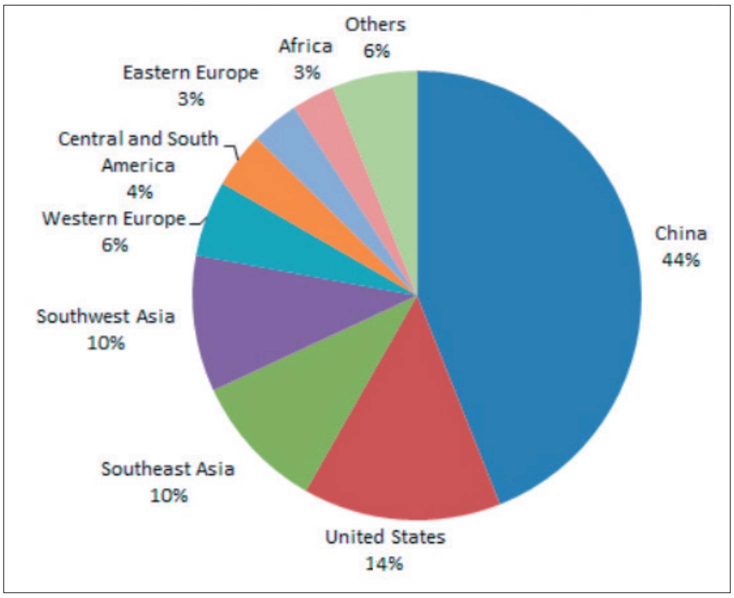


Figure 1. Fertilizer consumption in particular countries and regions (HIS Markit, 2022)

Since the productivity of agriculture is intended to provide enough food for the population, the consumption of fertilizer must be closely related to the number of people in a given region or country. Related to that concept, Figure 1 shows that China, the most populated country in the world, consumes the most fertilizer in comparison to other countries, followed by the United States of America and Asia region.

Plants can obtain nutrients from the soil that naturally contains important nutrients, such as nitrogen, potassium, phosphorus, magnesium, and calcium. However, most soil often lacks nutrients for plants, especially soil that has been repetitively used for plantation. Meanwhile, the limitation of nutrients supply will limits plant growth and reduces crop yields (Food and Agriculture Organization, 2000). Fertilizers can provide nutrients for soil lacking with nutrients to increase crop productivity (Food and Agriculture Organization, 2000). Unfortunately, the fertilizer's doses often exceed the recommended amount in an attempt of providing a rapid response to crops (Randive et al., 2021; Xiang et al., 2021). In the mean time, the excessive application of fertilizers can contaminate groundwater. It is caused by the vast difference between the rate of nutrient absorption by plants and the rate of nutrient release from fertilizer; the rate of nutrient absorption by plants is always lower than that of nutrient release from fertilizer (Solihin et al., 2020). The nutrients that are not absorbed by the plant then leach into and contaminating the groundwater. Consequently, there will be losses in both energy and financial resources due to excessive use of fertilizers, that also leads to an inefficient fertilization (Raliya et al., 2017).

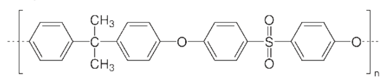
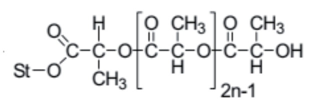
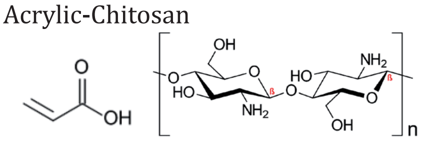
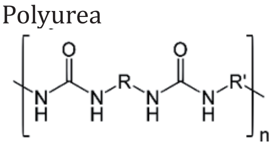
The groundwater pollution in the agricultural area has been reported by previous researchers (Pekny et al., 1989; Mulyadi et al., 2007; Mogheir et al., 2016; Taoun et al., 2017; Malki et al., 2017). According to the former findings, the leading cause of the pollution was excessive agrochemicals, especially nitrogen-based fertilizer. Additionally, it was also reported that plants only absorbed 30-40% of nitrogen, with the remaing 60-70% leaching vertically and horizontally before finally contaminating the groundwater (Mulyadi et al., 2007).

In order to resolve the issue above, an innovative fertilizer, in which the nutrient release can be slowed down and controlled, is required. Nowadays, a new type of fertilizer, known as the slow-release fertilizer, has been introduced and produced. This type of fertilizer has a unique property that allows the release of nutrient to be slowed down significantly.

THE REVIEW OF SLOW-RELEASE FERTILIZER SYNTHESIS

The first slow-release fertilizer, known as Methylene Urea (MU), was made in 1948 and commercially sold in 1955. After its first release, a series of fertilizers were produced, namely methylene di-urea (MDU), dimethylene-tri-urea (DMTU), trimethylene-tetra-urea (TMTU), etc. (Goertz, 1993). Five years after the first slow-release fertilizer was commercially sold, other types of slow-release fertilizer were synthesized, such as sulfur-coated and polymer-coated fertilizer (IFA Working Group, 2014). Following this invention, the coating method seems to be the most favorite method in the slow-release fertilizer synthesis. The coating materials used in the synthesis process were polysulfone (Tomaszewska et al., 2006), starch-g-poly (L-lactide) (Chen et al., 2008), acrylic-chitosan (Handayani et al., 2015), and polyurea (Lu et al., 2016). These coating materials seem to be effective in reducing the nutrient release rate at various levels. The molecular structure and release rate of each coating material is shown in Table 1.

Table 1. The molecular structure and release rate of nutrients from fertilizer coated by the different coating material (synthetic slow-release fertilizer)

Coating Materials	Release Rate of Nutrient
polysulfone 	10-35% of nutrient elements were released after being exposed to water for 5 hours (Tomaszewska et al., 2006)
starch-g-poly(L-lactide) 	100% of nutrient elements were released after being exposed to water for 7-26 hours (Chen et al., 2008)
Acrylic-Chitosan 	50-75% of nutrient elements were released after being used as fertilizer for 4 weeks (Handayani et al., 2015)
Polyurea 	90% of nutrient elements was released after being exposed to water for 5-55 days (Lu et al., 2016)

Conventional chemical fertilizers are usually water-soluble material; it takes short time to dissolve in water. The conventional chemical fertilizers had already released 100% of the nutrient elements into the water within 5 seconds (Gumelar et al., 2020). In contrast, the time needed for nutrient elements to release from slow-release fertilizer is much longer than conventional chemical fertilizer. Table 1 shows that synthetic base slow-release fertilizer can hold nutrient elements for several hours and days when exposed to water.

The application of slow-release fertilizers in the farm will result in a significantly longer nutrient release time because fertilizers are not sunk but poured into water. As the result, the leaching of nutrients elements is significantly decreased, and cosequently, the pollution of groundwater also can be considerably reduced.

EARTH'S RESOURCE-BASED SLOW-RELEASE FERTILIZERS

The slow-release fertilizer made through the coating method mainly consists of fertilizer and coating material. After being used, the nutrient from fertilizer is eventually reduced, leaving the coating material as waste that contaminates the farm area, especially if the coating materials are synthetic polymer. Therefore, the materials obtained from nature or earth are preferred to replace synthetic materials. Earth's materials, such as minerals, can be used as raw material to synthesize slow-release fertilizer.

Specific minerals, such as alumina (Zhang et al., 2009), hydrothermal silica (Solihin et al., 2017), and obsidian (Solihin et al., 2020) can be used as raw materials in slow-release fertilizer syntheses through various methods, such as coating, matrices, or carriers method by mixing of ordinary solid fertilizer with that mineral. Hydrothermal silica and obsidian that were used in the previous research were obtained locally from Indonesia, meanwhile alumina was obtained from Miyagi prefecture Japan. Hydrothermal silica was obtained from a geothermal power plant site located at Dieng Mountain, Bondowoso, Central Java, Indonesia (Solihin et al., 2017). The location of the hydrothermal silica in Dieng Geothermal Field is presented in Figure 2.

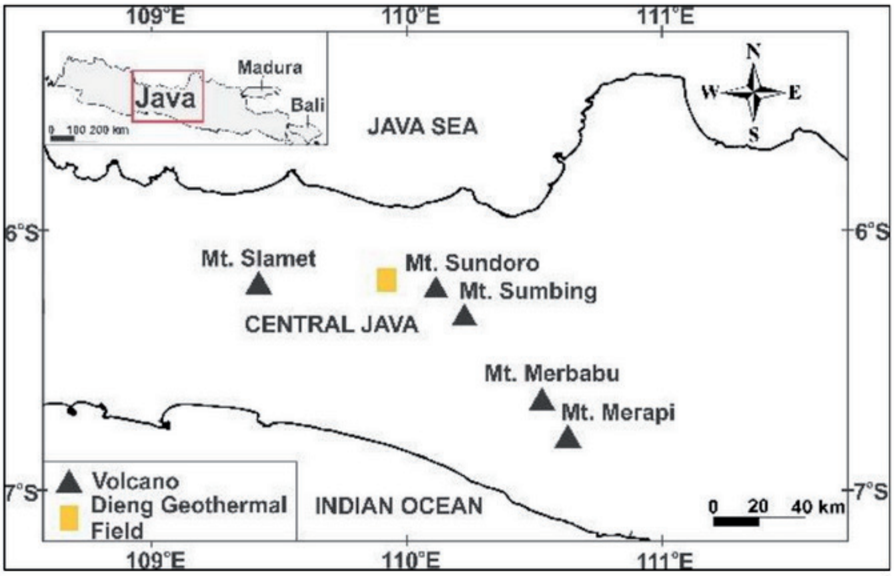


Figure 2. Location of the hydrothermal silica in Dieng Geothermal Field (Shalihin et al., 2020)

Hydrothermal silica is amorphous silica formed by erosion and decomposition of crystalline silica along with other minerals, followed by dissolution and precipitation of SiO_4^{2+} ion to form amorphous silica (Yuliyanti et al., 2015). The XRD pattern of as-received silica from the hydrothermal site shows a broad peak at 2q range of 20-30°, suggesting that the hydrothermal silica was in the amorphous form.

The peak corresponding to NaCl was also found. Furthermore, the previous study on this hydrothermal silica reported that this amorphous phase is silica (SiO_2) (Solihin et al., 2017). Thus, hydrothermal silica consists of the amorphous phase of SiO_2 and NaCl (salt). Salt can be separated from amorphous silica through washing before being used as raw materials of slow-release fertilizer synthesis.

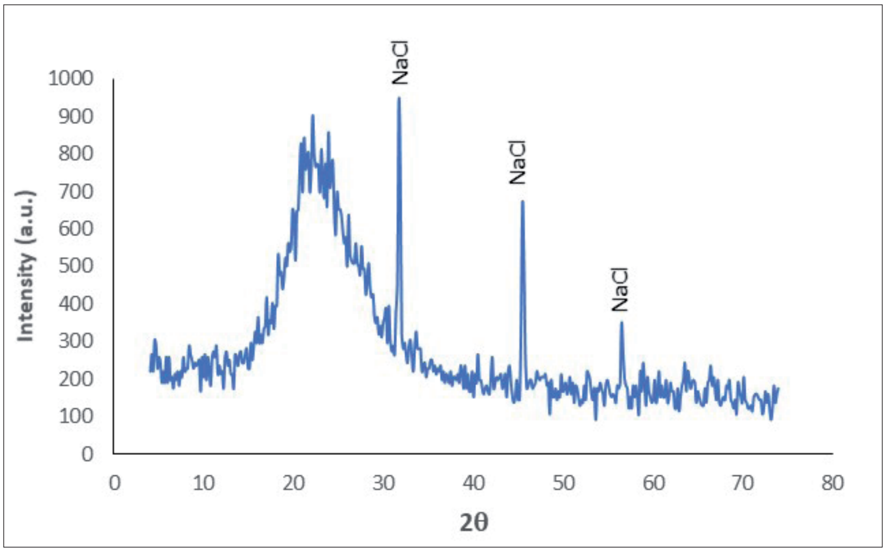


Figure 3. XRD Pattern of as-received silica from hydrothermal site

The other mineral that can be used as raw material to synthesize slow-release fertilizer is obsidian. Obsidian, also known as volcanic glass, has been used since the ancient era. The ancient people used obsidian as raw material to make knives, spears, weapons, and other tools (Fauzi et al., 2017; 2019). The utilization of obsidian to make such tools was significantly reduced after the discovery of metal. Nowadays, obsidian is only used as a decorative stone.

The archaeologists reported that obsidian-made tools were found in many places in Indonesia, Malaysia, the Philippines, and Papua New Guinea. Although these can be found in those places, the deposits of obsidian in South East Asia are located in limited areas, such as West Java (Indonesia), Pagudpud, Nagcarlan (The Philippines), and Lou Island (Papua New Guinea). The location of obsidian sources is shown in Figure 4.

Figure 4. Location archaeological sites and obsidian sources (Spriggs et al., 2011)

Obsidian was reported to be found in various locations in Indonesia, including Garut, Nagrek (West Java), Minahasa (Northern Sulawesi), Kamasa Hill (Southern Sulawesi), Banda Islands, Lahat (South Sumatera), Tulang Pandangin (Lampung), and others (Figure 4). The total reserve of obsidian in Garut was estimated to be 72 million tons (Husin and Sugiharto, 2008). Figure 5 displays the results of our XRD analysis of obsidian from Garut. The figure shows that the obsidian is composed of SiO_2, $NaAlSiO_3$, and an amorphous phase. The amorphous phase is thought to be amorphous silica or silicate.

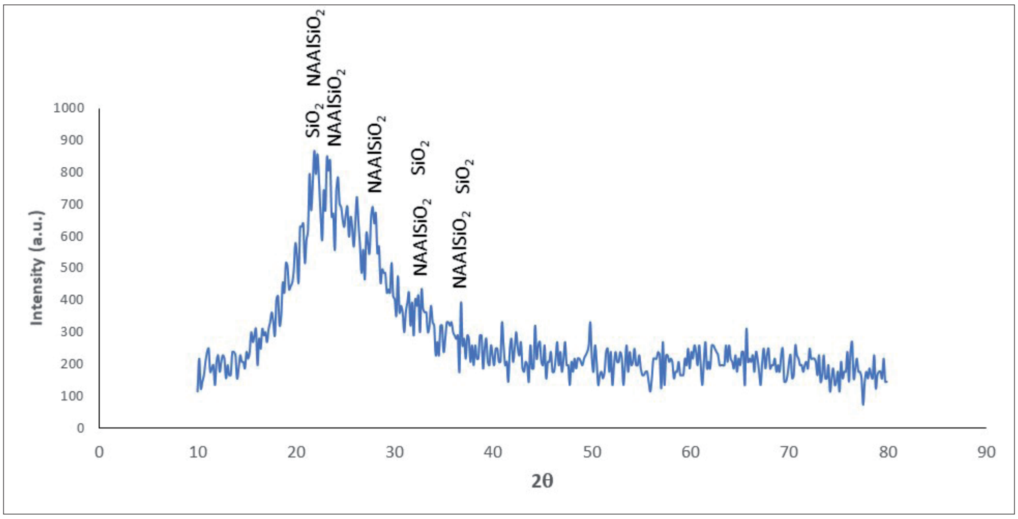


Figure 5. XRD pattern of obsidian

Silica is widely used in the synthesis of slow-release fertilizer due to its small particle size and amorphous properties, which affects the mechanism of fertilizer release to become more directed to the mechanism of the diffusion process (Kaavessina et al., 2021). These silica minerals have cavities that play a role in absorbing nutrients in their aluminosilicate structure, temporarily holding nutrients in the root area and slowly releasing them from time to time to be absorbed by plants. Additionally, they are not easily lost through washing or volatilization, making it more efficient to use as fertilizer. The mechanism of slow-release fertilizer when in contact with water or soil solution is given in Figure 6. In the discharge process, water penetrates through the coating membrane, condenses on the solid core, and then partially releases nutrients. This release occurs due to osmotic pressure, which then swells. The rupture of the coating material is followed by the breakdown of the fertilizer coating. The release mechanism depends on external stimulus, such as temperature, pH, and water permeability. Higher temperatures with greater moisture content can increase the release rate of fertilizer (Azeem et al., 2014).

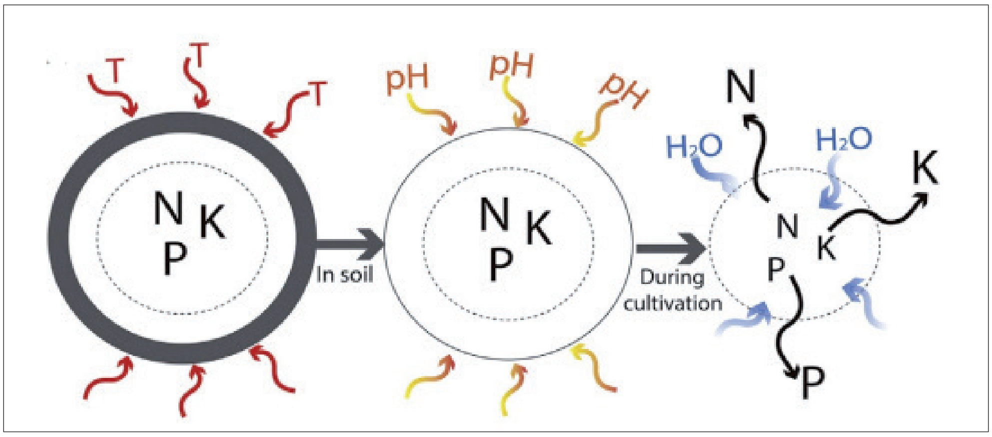


Figure 6. Diffusion mechanism of slow-release fertilizer (Calabi-Floody et al., 2018)

THE UTILIZATION OF EARTH'S RESOURCE-BASED SLOW-RELEASE FERTILIZERS TO REDUCE GROUNDWATER POLLUTION

Table 2 presents the earth materials used to synthesize the earth's resource-based slow-release fertilizers and the release rate of nutrients from each type of material. The release rate was measured by sinking the fertilizer in water and collecting the sample of water at a specific range time. Samples were then diluted and analyzed by using Atomic Absorption Spectrometry (AAS) and gravimetry analysis to determine the concentration of fertilizer elements. The performance of slow-release fertilizer was measured based on the time needed to dissolve a certain percentage of fertilizer element. Table 2 shows that the earth's resource-based slow-release fertilizers can hold nutrients much longer than conventional chemical fertilizers when exposed to the water; it takes hours and days for nutrients to be released into the water.

Table 2. The release rate of nutrients from fertilizer made by using the different types of earth's material

Earth's Material	Release Rate of Nutrient
Alumina	80-90% of nutrient elements were released after being exposed to water for 20 days (Zhang et al., 2009)
Hydrothermal Silica	100% of nutrient elements were released after being exposed to water for 5 hours (Solihin et al., 2017)
Obsidian	60-70% of nutrient elements were released after being exposed to water for 50 hours (Solihin et al., 2020)

It can be seen in Tables 1 and 2 that synthetic base slow-release and earth's resource-based slow-release fertilizers can hold nutrients much longer than conventional chemical fertilizers when exposed to the water. Although there is no standard method for determining the rate of nutrient release from slow-release fertilizers (Lawrencia et al., 2021), it was hypothesized that the earth's resource-based slow-release fertilizer is more effective than the synthetic ones because the majority of synthetic slow-release fertilizers are hydrophilic (Fertahi et al., 2021).

Since the earth's resource-based slow-release fertilizers are made from the earth's natural materials, it has no harmful substances. In addition, unlike the synthetic slow-release fertilizer, the earth's resource-based slow-release fertilizer leaves no waste after use. Therefore, the earth's resource-based slow-release fertilizers are more environmental friendly than synthetic ones.

CONCLUSION

This review paper concludes that the excessive utilization of fertilizer contaminates groundwater. Previous research revealed that the rate of nutrients released from various slow-release fertilizers synthesized was significantly low. Therefore, the utilization of slow-release fertilizer can significantly decrease groundwater contamination while maintaining agriculture productivity. Besides synthetic materials, natural minerals such as hydrothermal silica and obsidian can be used to synthesize slow-release fertilizer. The synthetic base slow-release fertilizers and earth's resource-based slow-release fertilizers can hold nutrients much longer than conventional chemical fertilizers when exposed to water. The earth's based slow-release fertilizer is environmentally friendly because it only leaves behind natural minerals rather than harmful materials. The XRD analysis of hydrothermal silica and obsidian shows amorphous silica and other minerals content. Furthermore, silica in compacted slow-release fertilizer is able to control the release rate of fertilizer elements.

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