



Influence of the particle size of carbonate-siliceous rock on the efficiency of phosphorous removal from domestic wastewater



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ABSTRACT

The aim of the study was to determine the influence of the particle size of carbonate-silica rock (opoka) used in rock filters on the efficiency of phosphorus removal from domestic wastewater. The investigations were carried out in a laboratory using a model consisting of three vertical flow filters with carbonate-silica rock of different particle sizes ($G_1 = 1-2$ mm; $G_2 = 2-5$ mm; $G_3 = 5-10$ mm). The tested rock was subjected to decarbonising at 900°C and consisted primarily of 51.7% SiO_2 , 23.2% CaO , and 7.6% Al_2O_3 . In the first three weeks of the study (1–3), the hydraulic load of each filter was $Q_1 = 0.72$ l/day and the hydraulic residence time was $\text{HRT}_1 = 24$ h; in the next three weeks (4–6) $Q_2 = 1.08$ l/day and $\text{HRT}_2 = 16$ h, and during the last three weeks (7–9) $Q_3 = 1.44$ l/day and $\text{HRT}_3 = 12$ h. A significant influence of the size of rock particles and the hydraulic load (hydraulic residence time) on the efficiency of total phosphorus removal and on phosphorus concentration in the wastewater discharged from the system was observed ($\alpha = 0.05$). Statistically, the best removal of phosphorus from wastewater – an average of 97%, was found in the substrate with the smallest particle size (rock G_1 , 1–2 mm) at the lowest hydraulic load of 0.72 l/day and at a hydraulic residence time of 24 h. The lowest phosphorus removal efficiency was observed in the filter containing rock G_3 with a particle size of 5–10 mm (mean <60%). The average concentration of total phosphorus in wastewater flowing out from filter G_1 was 0.23 mg/l, which was much below the limit values specified by EU regulations. The overall phosphorus load removed during the study period (nine weeks) in the filter with fraction G_1 was 0.38 g/kg of rock, in the filter with G_2 –0.30 g/kg of rock, in the filter G_3 –0.28 g/kg of rock. The load of phosphorus removed during this period not characterized the full sorption capacity of the rock. The study showed that the rock subjected to decarbonising at 900°C could be successfully used to remove phosphorus from domestic wastewater, especially in areas where phosphorus removal requirements are very high ($P_{\text{tot}} < 2$ mg/l).

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1. Introduction

The problem of removal of biogenic compounds from wastewater is still unresolved, in particular with regard to small and local treatment plants. Around the world, researchers are looking for innovative ways of eliminating biogenic elements (nitrogen and phosphorous) from wastewater with a view to reducing the process

of eutrophication of surface waters. The main element responsible for the fertility of fresh water is phosphorus. It may come from various sources: (1) natural – organic compounds of animal and plant origin, and (2) anthropogenic – agriculture (aerial sources) and different types of insufficiently treated wastewater (point sources). Among point sources, the most important ones are: industrial wastewater, e.g. discharged from plants producing fertilizers and cleaning agents based on detergents, municipal wastewater, wastewater from pig farming and domestic wastewater. Total phosphorus concentrations in raw domestic wastewater

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are in the range from a dozen to several dozen mg/l (Metcalf and Eddy, 2003).

Discharge of improperly treated wastewater may cause many problems in the recipient body. The most important of those problems include oxygen depletion due to mineralization of organic matter and oxidation of ammonium nitrogen (nitrification) (Józwiakowski et al., 2017). When the concentration of the two biogenic compounds in the discharged wastewater is too high, oxygen depletion accelerates due to the intensive growth and death (decomposition) of algae. An increase in phosphorus concentration in surface waters up to over 15 µg/l may lead to an intense growth of algae (Yang et al., 2008). It is estimated that large amounts of biogenic compounds contained in sewage can increase secondary oxygen consumption more than five-fold, compared with primary consumption associated with the disposal of organic matter contained in wastewater. Therefore, it is crucial to reduce the content of biogenic compounds in the effluent discharged from treatment plants to receivers (Mikosz and Mucha, 2014; Gajewska et al., 2015; Józwiakowski et al., 2015).

Phosphorus concentration in the effluent can be reduced using biological and chemical purification methods (Clark et al., 1997; Wei and Zhi, 2002; Chiou and Yang, 2008, 2008; Wei et al., 2013). Biological removal consists in creating optimal conditions in a sewage treatment plant for the growth of microorganisms capable of collecting an excess of phosphorus, and then removing the accumulated phosphorus with excess sludge (Morse et al., 1998). Chemical processes rely, instead, on the use of coagulants, e.g. coagulants of iron and aluminium. The removal of phosphate with reactive media as sorption filters has been more and more frequently used over the recent years in small wastewater treatment plants (Renman and Renman, 2010; Bus and Karczmarczyk, 2014; Nastawny et al., 2015; Jucherski et al., 2016). In this method, wastewater filtering through a substrate bed remains in contact with a reactive material, which facilitates chemical precipitation and sorption of phosphorus compounds (Eveborn, 2013). The biological method usually guarantees the elimination of required amounts of phosphorus/reduction of phosphorus to a required level, but is only effective when used in large (over 100 000PE) wastewater treatment plants (WWTP). The use of aluminum or iron coagulants is cumbersome and relatively expensive, especially for small and medium wastewater treatment plants. The main disadvantage of chemical precipitation of phosphorus is production of chemical sludge which needs to be disposed of, increasing the cost of WWTP operation. Given these difficulties, there is a need to study materials capable of absorbing large amounts of phosphorus, both natural and man-made, such as rock, granulated blast furnace slag, fly ash, gravel, or brick covered with iron (Vohla et al., 2011).

Studies on the use of carbonate–silica rock (opoka) to remove phosphorus from wastewater have been carried out for many years now (Brogowski and Gworek, 1996; Brogowski and Renman, 2004; Cucarella et al., 2007; Józwiakowski, 2006, 2012; Renman and Renman, 2010; Karczmarczyk and Bus, 2014; Bus and Karczmarczyk, 2014). This rock material is highly reactive to phosphorus because it contains large amounts of calcium and silicon. The content of these components varies in different types of opoka from 14 to 56% CaO and from 5 to 75% SiO₂ (Kozłowski, 1986; Brogowski and Renman, 2004; Bus and Karczmarczyk, 2014). Carbonate–silica rock is characterised by more than 50% porosity (Brogowski and Gworek, 1996). This material is of organic origin and consists mainly of small organic debris with some addition of silica. Thus opoka is assumed to be an intermediate form between rocks containing carbonate and those containing silica (Pinińska, 2008; Brogowski and Renman, 2004). The composition and properties of the rock favour chemical sorption of phosphorus. The good sorption properties of the material could be enhanced in an alkaline environment since in such conditions phosphorus forms

Table 1

Relationship between sorption capacity of carbonate–silica rock fired at 900 °C and Ca content (Cucarella et al., 2007).

Element	Rock 1	Rock 2	Rock 3
Ca [g/kg]	220.79	364.39	419.75
Sorption capacity [mg P/g]	79.37	136.99	181.82

chemical bonds with calcium to give calcium phosphates (Reddy and D'Angelo, 1997; McGechan and Lewis, 2002).

So far, it has been established that thermal treatment increases the sorption capacity of rocks. A study by Brogowski and Renman (2004) shows that a natural carbonate–silica rock is characterized by a sorption capacity of 19.6 g P/kg, but after firing at 250 °C the capacity increases to 60.5 g P/kg, and at 1000 °C–119.6 g P/kg. Additionally, experiments conducted by Cucarella et al. (2007) and Bus and Karczmarczyk (2014) demonstrate that sorption capacity of rocks is also closely related to their calcium content (Table 1).

For a few years now, a rock excavated in the village of Bełżec in Poland (50°3'04" N, 23°26'18" E), heated at a temperature of about 900 °C, with a particle size of 2.0–6.0 mm, bearing the trade name Polonite®, has been used, primarily in Sweden, as a sorbent for the removal of phosphorus from wastewater (Bus and Karczmarczyk, 2014).

The reports cited above have shown that using this rock as a substrate in phosphorus-removal filters gives very good results. Up till now, however, only one grain size of Polonite® (2.0–6.0 mm) has been shown to have a high (over 90%) phosphorous removal capacity. Researchers believe that the main mechanism of phosphorous removal is chemical sorption, and if so the process is strongly dependent on the availability of calcium for binding to P (Karczmarczyk, 2000, 2003; Karczmarczyk and Mosiej, 2003; Karczmarczyk et al., 2003; Brogowski and Renman, 2004; Józwiakowski, 2006; Renman, 2008; Cucarella, 2009; Nilson, 2012). Unfortunately, there are few studies regarding the effect of the composition of rocks and their particle size on phosphorus removal. To investigate this problem, we decided to conduct experiments with different particle sizes (different surfaces) of carbonate–silica rock and different hydraulic retention times. Both of these parameters were hypothesized to have an influence on the efficiency of phosphorus removal from domestic wastewater.

2. Material and method

The experiments were conducted using a laboratory model consisting of three vertical-flow wastewater filters filled with opoka as a substrate (Fig. 1). The rock was obtained from a mine in the town of Chrzanów, located in south-eastern Poland (50°46'26"N, 22°36'19"E) and was characterized by a different composition in comparison to those described by Karczmarczyk (2000, 2003); Karczmarczyk and Mosiej (2003), Karczmarczyk et al. (2003), Brogowski and Renman (2004), Józwiakowski (2006), Renman et al. (2008) and Renman (2008). The substrate rock was dried, crushed and sorted, to obtain three types of samples differing in particle size: G₁, 1–2 mm; G₂, 2–5 mm; and G₃, 5–10 mm (Fig. 2).

Then, the three particle-size fractions of rock were heated in a muffle furnace at 900 °C, as treatment at this temperature had been reported to ensure the highest sorption capacity and thus the most effective removal of phosphorous. Prepared in this way, the granular substrate, sorted by grain size, was packed into three filters with a volume of 1.4 l each (Fig. 1). The chemical composition of the rock, estimated using a ground mixture of samples is shown in Table 2. The effect of the size of substrate on its composition was not investigated. Studies performed using an X-ray spectrometer (fluorescence spectrometry, MINIPAL 4 from PANALYTICAL) showed that the test rock consisted primarily of 51.7% SiO₂, 23.2%

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