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Evaluating resilience to sludge dry matter and particle size variations

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Robust magnetic vivianite recovery from digested sewage sludge: Evaluating resilience to sludge dry matter and particle size variations

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ABSTRACT

Phosphorus recovery via vivianite extraction from digested sludge has recently gained considerable interest. The separation of vivianite was demonstrated earlier at the pilot scale, and operational parameters were optimized. In this study, we tested the robustness of this technology by changing the sludge characteristics, such as dry matter, and via that, sludge viscosity, and vivianite particle size. It was proven that the main factor influencing recovery was the concentration of vivianite in the feed. The technology can extract vivianite even when the sludge has higher dry matter (1.8% - 3.3%) and, therefore, higher viscosity. Smaller vivianite sizes (< 10 μm) can still be recovered but at a lower rate. This made magnetic separation applicable to a wide range of wastewater treatment plants.

1. Introduction

Recovering phosphorus from sewage sludge through vivianite ($Fe_3(PO_4)_2*8H_2O$) allows a higher amount of phosphate recovered when compared to the conventional struvite precipitation method (Wijdeveld et al., 2022; Wilfert et al., 2015). In digested sewage sludge from wastewater treatment plants (WWTPs) that use iron as a coagulant, up to 90% of the total phosphorus can be in the form of vivianite (Wilfert et al., 2018). The challenge arises from the small size of vivianite particles in sludge, typically ranging from only 10 to 200 μm (Wijdeveld et al., 2022), making their separation through gravity settling difficult.

However, due to vivianite's paramagnetic characteristics, it can be recovered using devices like a high-gradient magnetic separator (HGMS). HGMS is commonly used to extract fine and weakly magnetic materials in the mining industry (Ge et al., 2017). An example of an HGMS is the SLon® vertically pulsating high-gradient magnetic separator (VPHGMS). It works by rotating a ring vertically through the liquid, attracting magnetic particles to the magnetized matrix of steel rods and then flushing them out. The pulsation prevents matrix clogging and helps purify the magnetic product, resulting in higher efficiency (Xiong et al., 1998). The SLon® is known for its effectiveness in concentrating iron minerals like hematite, magnetite, and limonite, particularly in processes such as desulphurization and dephosphorization of iron ore concentrate (Xiong et al., 1998; Zeng and Dahe, 2003).

In digested sewage sludge, a VPHGMS has been tested at pilot scale at the WWTP in Nieuwveer as an urban mining concept called Vivimag®. This study, conducted by Wijdeveld et al. (2022), focused on optimizing operational parameters such as magnetic strength, rod diameter, and pulsation frequency to improve vivianite recovery. The vivianite recovery efficiency (weight of vivianite magnetically recovered divided by the weight of vivianite in the sludge fed through the magnetic separator) could reach 80% with three recirculations. However, as the technology is scaling up, there needs to be a deeper understanding of its robustness when dealing with variations in sludge characteristics of different WWTPs.

Numerous factors in sludge affect magnetic vivianite recovery, including vivianite magnetic susceptibility, other magnetic materials, non-magnetic fraction concentration, pH, particle morphology, and sludge viscosity. According to Svoboda (2004), particle magnetic susceptibility has a minimal impact compared to particle size on the HGMS' performance. Other magnetic compounds in sludge may reduce the recovery grade. However, in digested sludge, where the Fe/P molar ratio is

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often below 1.5, vivianite predominates as the magnetic fraction, while amorphous FeS is not magnetically attracted (Prot et al., 2020; Roussel and Carliell-Marquet, 2016). In less common cases where the Fe/P ratio is higher, other iron compounds are formed, such as siderite ($FeCO_3$) or pyrite (FeS_2), which are non-magnetic (Prot et al., 2019; Wilfert et al., 2018). To the authors' knowledge, other forms of iron are not commonly found and cannot represent a significant enough fraction to disturb the purity of the recovered product. Non-magnetic fractions can influence recovery and grade, with their entrainment lowering HGMS selectivity (Hu et al., 2023). Additionally, pH changes in the carrier fluid can alter particle behavior and HGMS performance. Nevertheless, pH in anaerobically digested sludge typically remains near neutral (Canziani and Spinosa, 2019). These factors can all influence vivianite recovery efficiency with the magnetic separator, although their individual effects are often minimal, irrelevant for sludge, or impractical to alter.

On the other hand, the effects of particle size and fluid viscosity have been extensively studied in the HGMS systems, primarily other than in sludge. The two parameters are inherent to the fluid properties and are often predicted to influence the particle's trajectory toward the magnet because they can alter the forces acting on such particle, both in direction and magnitude. The force attracting paramagnetic particles is the magnetic force, enhancing selectivity and rejecting non-magnetic particles. Competing forces, primarily hydrodynamic drag caused by fluid viscosity, counteract the magnetic force. In VPHGMS, steel rods create a magnetic field gradient for enhanced particle capture. Once attached to the rods, the particles can still be detached by fluid drag force, which is especially important for small particles. In the SLon®, an extra pulse drag is induced, proportional to fluid viscosity (Peng et al., 1992).

Sludge undergoes shear-thinning, meaning reducing viscosity with increased shear stress (Baudez et al., 2011). Higher sludge dry matter (DM) correlates with increased viscosity (Cao et al., 2016; Infusino and Caloiero, 2021; Wei et al., 2018). Furthermore, DM and rheology vary with WWTP operational conditions (Wei et al., 2018). Sludge, even with the same DM, exhibits diverse viscosity due to factors like floc concentration and extracellular polymeric substance (EPS) (Papa et al., 2019; Ratkovich et al., 2013). Recently, vivianite formation in thickened sludge without anaerobic conditions has been demonstrated (Prot et al., 2022). Thickened sludge, with higher DM and larger flocs, has elevated viscosity (Civelekoglu and Kalkan, 2010). Given viscosity variations in different sludge types, exploring its impact on vivianite recovery is essential for the technology's widespread application.

Vivianite particles found in sludge are typically between 10-200 μm (Wijdeveld et al., 2022; Wilfert et al., 2018), meaning there is a significant variation in particle size. However, Wang et al. (2021) reported a vivianite size of only between 1-10 μm, attributing that to the different formation conditions at different WWTPs. Prot (2021) reported the challenge of growing bigger vivianite crystals in sewage sludge's pH and concentration conditions. This suggests limitations in the bigger sizes of vivianite. The magnetic force attracting vivianite is proportional to the cube of the particle's radius, making it the dominant factor favoring the separation. This means that, in theory, the bigger the particle, the easier it is to be attracted to the magnet and hence recovered. Moreover, the drag force's magnitude also depends on the particle size. As the magnetic recovery of vivianite is to be applied to various sludge sources where particle size variation exists, it becomes crucial to identify the favorable conditions for the magnet to operate at high efficiency and understand if there is a size limitation of the recoverable vivianite.

In the Vivimag® pilot investigation by Wijdeveld et al. (2022), the emphasis was placed on optimizing the operational parameters of the magnet to achieve enhanced recovery efficiency. However, the influence of the sludge's varying characteristics remains a critical determinant for the applicability of this technology. To examine these factors, for the first time, a comprehensive sampling campaign for vivianite recovery was conducted at a Vivimag® pilot plant in a WWTP in Schönebeck, Germany. This study addressed the alteration of sludge characteristics by manipulating sludge dry matter and reducing vivianite sizes. The focal point of our investigation was to discover the relationships between DM and, consequently, viscosity, and particle size and the recovery of vivianite through magnetic separation. This research is a unique contribution to the P recovery technology, providing novel insights on crucial relationships that improve the understanding and potential optimization of vivianite recovery in wastewater treatment processes.

2. Materials and methods

2.1. Sludge characteristics

The study was carried out at a municipal WWTP in Schönebeck, Germany, whose size is around 90,000 people equivalent with an average influent flow of 9000 m^3/ day. The WWTP has a primary settling system followed by biological treatment to remove P and N using the activated sludge process. Originally, phosphorus was mainly removed via enhanced biological phosphorus removal (EBPR). Ferric chloride ($FeCl_3$) was dosed at the inlet of the WWTP to manage odor and before the secondary clarifier to regulate discharge quality. Both the primary and secondary sludge underwent thickening before being directed to the anaerobic digester. In addition to the two sludges, 25% of the flow to the digester was from co-substrate, consisting of fat, oil, and grease from the surrounding restaurants. To promote vivianite formation in sludge, extra iron was dosed in the primary sludge pumping station for 7 months prior to this study, which raised the Fe/P molar ratio in the digested sludge from 0.5 to 1.6 at sampling time. In the meantime, the activated sludge system was still operated in EBPR mode. Some of the iron dosed reached the activated sludge system via the primary settler overflow, which may have had implications on the EBPR performance. The feed to the magnetic separator was anaerobically digested sludge with a 20-day retention time.

2.2. Pilot installation and experimental description for all experiments

The pilot installation is the same as described by Grönfors et al. (2022), with the working scheme as illustrated in Fig. 1. It is comprised of a SLON®-750 VPHGMS accompanied by supplementary equipment such as pumping and sludge storage tanks. A fraction of the sludge from the digester was diverted to a large buffer tank, which contained all the sludge used as feed throughout the entire experimental period without refilling or emptying. Each experiment used a portion of the digested sludge in the buffer tank. Subsequently, it was transported to a continuously stirred feed tank (1 m^3 capacity) and pumped through the magnetic separator. The holding capacity of the magnetic separator was around 22 liters. The concentrate contains the magnetic fraction recovered and flowed to a gravity settler to thicken. The reject from the magnet comprising the non-magnetic fraction is called the tailings and is transported to the tailings tank (1 m^3 capacity).

This section describes general operational parameters that will stay constant for all experiments. A more detailed explanation of the feed characteristics of each experimental type will be described in the subsequent sections. Before the experiments, the operational parameters of the magnet had already been optimized to maximize vivianite recovery from digested sludge in standard conditions (non-modified digested sludge as feed). The optimized parameters were similar to those in Wijdeveld et al. (2022)'s work. The same parameters were maintained during the experiments. The magnetic strength was set at 1 Tesla, the ring speed at 1 rpm, and the rod diameter at 3 mm. The SLon® can operate at the flow rate of 1 m^3/h. However, due to limitations in the available material used as feed for the experiments, we operated the magnet at 0.3 m^3/h. Each experiment was run in batch mode with no recirculation, where a batch of around 0.5 – 0.8 m^3 of sludge was loaded to the feed tank and passed continuously to the magnetic separator in a single pass. The duration of each experiment was 75 minutes. At 15-minute intervals, 500 ml samples of the feed, tailings, and concentrate were

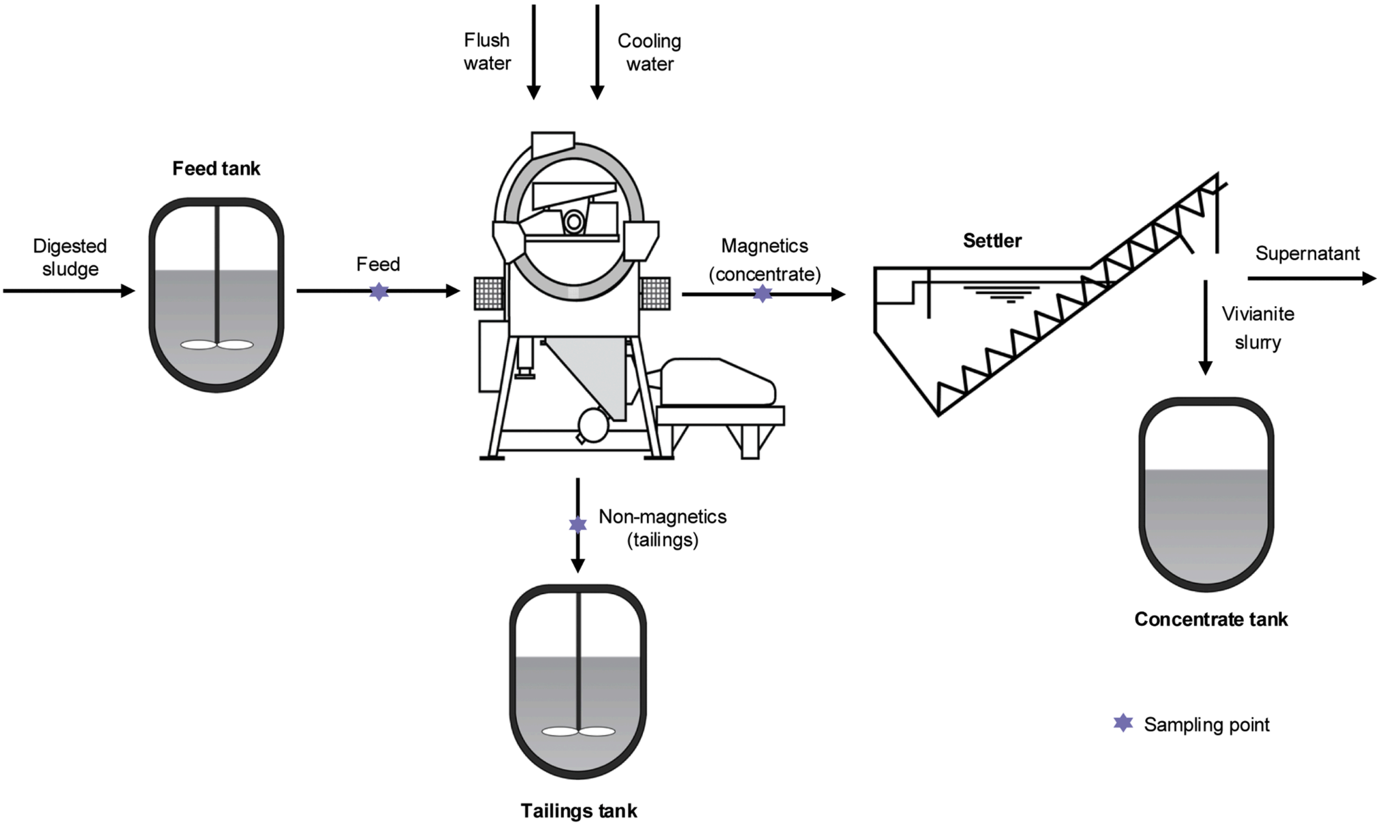

Fig. 1. Simplified scheme of the pilot-scale magnetic separator for vivianite recovery (Vivimag®) in Schönebeck WWTP, Germany.

collected, while flow rates of all three streams were measured every 30 minutes to complete the mass balance around the magnet. In total, for each experiment, a collection of 5 samples (for elemental composition determination) and measurements of 3 flow rates for each stream were recorded. Every experiment was done in duplicate to confirm its reliability.

2.3. Control experiments

Control experiments were conducted four times throughout the two-week sampling period with the pilot installation. These experiments maintained the standard conditions, with non-modified digested sludge as feed. The objective of the controls was to record the vivianite recovery under normal conditions. Additionally, they contribute to the comparative analyses with other experiments where sludge was modified and enhance the understanding of the fluctuations in recovery efficiency should there be changes in the sludge composition through time.

2.4. Sludge dry matter and viscosity experiments

As mentioned above, sludge dry matter can impact its viscosity. Therefore, the effect of viscosity was indirectly tested by manipulating DM. Digested sludge was thickened to around 4% DM for all DM experiments by gravity settling. Then, the thickened sludge was diluted to the desired DM. This is done to maintain consistency in the experiment as all sludge used here was sourced from the same thickened digested sludge with identical characteristics, varying only in dilution levels. This prevents simultaneous alterations in both particle sizes (due to selectivity for bigger particles during gravity settling) and viscosity between samples requiring thickening and those requiring diluting. The dilution was done with process water from the WWTP, which is filtered effluent, to not significantly alter the sludge's ionic strength or pH. Three different sludge batches were thickened on three different days to obtain enough feed material. To test whether the thickening can alter the outcome, the control was also diluted in an experiment called "Diluted control".

2.5. Particle size experiments

Concentrate, consisting of 86% vivianite, was collected from the settler after the magnetic separator. Due to a difference in settling rate, the vivianite and organics separate in the settler, increasing the vivianite purity. This settled vivianite was ground in a food blender to break down the vivianite particles. A picture of the ground vivianite can be found in the Supplementary Information (Figure S1). The original vivianite's D50 was 57.3 μm, and that of the ground vivianite was 31.8 μm (Table S2). This ground vivianite was added to digested sludge, and this mixture was passed through the magnet. With this, vivianite would originate from the digested sludge and extra-dosed ground vivianite. Therefore, the effect of having ground vivianite might not be significantly highlighted. Nevertheless, this would ensure that the original sludge did not undergo drastic changes (in viscosity, for example). This is because the amount of ground vivianite added (around 13 kg of added ground vivianite slurry in 0.5 m^3 digested sludge) was not big enough to change sludge viscosity. The unground vivianite was added to the digested sludge as feed in the same concentration as ground vivianite. This test aimed to evaluate whether the rods reached saturation with enhanced concentration of feed vivianite.

Ground vivianite was added to vivianite-depleted tailings and passed through the magnet to observe particle size's influence more explicitly and increase the proportion of smaller-sized vivianite. The vivianite-depleted tailings was achieved by passing the tailings to the magnet twice more to eliminate as much vivianite existing in the sludge as possible so that the majority of vivianite would come from the added ground vivianite. While this approach can effectively highlight the impact of small vivianite, the recirculation process led to the dilution of the tailings with rinsing water (estimated 1.5 – 1.6 times), risking altering sludge viscosity.

The recovered vivianite concentrate was used because they are certainly recoverable vivianite particles from the magnet. By adding the ground vivianite as feed, if the recovery percentage remains unchanged, it would indicate that the smaller size of vivianite does not influence the recovery. Through observing the effects of both experiments, comprehensive conclusions can be drawn.

2.6. *Summary of the experiments*

All experiments are summarized in the table below. Each experiment was named with the format “Date” + “Experiment”.

For example, an experiment where non-modified digested sludge was used as feed on the 24th of October will be named “2410 control”.

2.7. *Recovery definition*

Since the concentrate was deposited on the horizontal concentrate collecting pipe, it was impossible to collect them all, so the recovery could not be evaluated directly from the concentrate data but was deducted from the difference between the feed and tailings. After verification of flows and concentrations of each element in the feed, tailings, and concentrate, it was concluded that the difference between what was found in the feed and what was measured in the tailings would be in the concentrate. The recovery of vivianite is therefore calculated as:

$$R = \frac{Feed - Tailings}{Feed} * 100\%,$$

Wherein, both feed and tailings denote the kilograms of vivianite present in the respective samples.

2.8. *Analyses*

2.8.1. *Elemental composition*

All samples were digested with an Ethos Easy digester from Milestone using an SK-15 High-Pressure rotor. Approximately 2 g of wet feed and tailings were used, while for concentrate, 10 g of sample was used. The samples were dispensed into Teflon vessels with 10 ml ultrapure HNO_3 69% addition. The mixture was heated for 15 minutes at 200°C, maintained at the same temperature for another 15 minutes, and cooled down for about an hour.

Post-digestion, the samples were diluted and analyzed with Inductively Coupled Plasma (Perkin Elmer, type Optima 5300 DV) equipped with an Optical Emission Spectroscope (ICP-OES). The autosampler used was the Perkin Elmer type ESI-SC-4 DX, and the software was Perkin Elmer WinLab32. The standard solution was 10 mg/L Yttrium, and the rinsing solution was 2% HNO_3.

Table 1

Summary and description of the experiments with the Vivimag® pilot in Schönebeck WWTP, Germany

Experiment name	Description	Study topic
Control	Non-modified digested sludge from the WWTP	Baseline
Diluted control	Digested sludge diluted with process water	Dry matter/ viscosity
DM *x*%	4% thickened sludge, then diluted to *x*% dry matter	
DS viv	Digested sludge with the addition of recovered unground vivianite*	Particle size
DS gv	Digested sludge with the addition of ground vivianite*	
Tailings gv	Tailings was passed through the magnet two more times with the addition of ground vivianite*	

* The vivianite added is the recovered concentrate collected from the pilot before, which contains 86% of pure vivianite.

2.8.2. *Sludge viscosity measurement*

The measurement of sludge viscosity was done with an Anton Paar MCR302 rheometer. Around 15 ml of feed sludge was added to the rheometer, which consisted of two concentric cylinders (a rotating measuring bob and a stationary cup) as a rotational Couette geometry. The CC27 system comprised a bob of 26.656 mm and a cup of 28.920 mm in diameter. The measurements were done at 20°C. Before the measurement, each sample underwent pre-shearing at the shear rate of 1000 min^{-1} for 90 seconds to reach homogeneity. After 30 seconds of pause, the viscosity was assessed across shearing rates ranging from 0.01 s^{-1} to 1000 s^{-1}, with a progressively decreasing time interval from 10s to 1s, following a logarithmic variation as previously demonstrated by Wei et al. (2018).

2.8.3. *Particle size analysis*

The particle size distribution of the recovered concentrate from the experiment was measured using laser diffraction with a hydro wet dispersion unit (120 ml), wherein a few ml of the wet concentrate was dropped. The equipment was the Malvern Mastersizer3000.

2.8.4. *Vivianite concentration determination*

The vivianite was determined using Mössbauer spectroscopy coupled with elemental analysis from ICP-OES after the samples had dried at 35°C and been ground. For concentrate samples, dilution with carbon powder was needed to reach a maximum of 17.5 mg Fe/ cm^2. The measurements were conducted at 300K. The ^{57}Fe Mössbauer transmission spectra were acquired using the conventional constant-acceleration spectrometer with a ^{57}Co(Rh) source. An α-Fe foil was used for the velocity calibration, and the Mosswin 4.0 program was used for spectra fitting (Klencsár, 1997).

The Mössbauer findings for the concentrate revealed that all the iron in the concentrate was vivianite. Additionally, the Fe/P molar ratio was 1.3, slightly lower than the ideal vivianite ratio of 1.5. So far, no other P-containing minerals in sludge have exhibited magnetic properties than vivianite. Thus, the most likely reason for the lower Fe/P ratio found in the concentrate is the presence of other divalent cations like Mg, Ca, and Mn replacing Fe in the vivianite structure (Rothe et al., 2016). Further purification of the obtained concentrate to 93% pure (calculated based on P concentration) revealed the same Fe/P ratio of 1.3, suggesting that only an insignificant amount or very low amount of P could come from the organic P entangled in the concentrate. Therefore, using P concentration leads to more accurate quantification of vivianite in the concentrate.

It was necessary to account for impurities to quantify vivianite in the feed sludge. Due to impurities, the iron grade in our vivianite decreases to an average of 29.2% compared to pure vivianite (33.4%). For each experiment, the average value of the iron grade in the concentrate was calculated and used to adjust the vivianite concentration calculated by Mössbauer spectroscopy for the feed. Further details on vivianite quantification are provided in Section S3.

2.8.5. *Reporting of results*

In every experiment, five samples were gathered for elemental analysis. Their results will be presented in the section below without averaging or including error bars but with raw data. Replicates within the same experiment will be visually identified with identical marker shapes and patterns.

3. Results and Discussion

Phosphate recovery via vivianite recovery from sewage sludge was tested using the magnetic separator at pilot scale. The anaerobically digested sludge has a Fe/P molar ratio of 1.6 originating from a WWTP using EBPR. The digested sludge is derived from primary and secondary thickened sludge, with approximately 25% co-substrate. The study initially examined the correlation between vivianite concentration in the feed and its recovery, subsequently investigating the influence of different sludge DM and smaller vivianite sizes.

3.1. *The influence of vivianite concentration in the feed*

When non-modified digested sludge was fed to the magnetic separator, there is a positive correlation between the concentration of vivianite in the feed (in g vivianite/ kg DM), or feed grade, and the percentage of vivianite recovered (Fig. 2). The recovery rate saw a significant increase, rising from only 35% to about 60% with the rise of feed grade from 180 to 400 g vivianite/kg DM. This trend is still valid for the case where unground vivianite was added to the digested sludge. Once the vivianite concentration surpasses 300 g/kg DM, the recovery starts to plateau.

A possible hypothesis for the increasing recovery percentage could be the magnetic flocculation of vivianite. This phenomenon is widely recognized, involving the magnetization of magnetic particles in the presence of a magnetic field, causing them to behave like mini magnets. Therefore, these particles can attract each other, forming chains or rings of agglomerates, thereby increasing the overall particle size, facilitating a more efficient settling or separation process (Garcia-Martinez et al., 2010).

Magnetic flocculation was frequently employed to recover magnetic particles ranging from ultrafine to colloidal sizes. Numerous studies have investigated the flocculation of paramagnetic particles, with examples including studies examining Fe_2O_3 (Svoboda, 1981; Tsouris and Scott, 1995), $Mn_2P_2O_7$, MnO, Mn_2O_3 (Janssen et al., 2000; Parker et al., 1982). Irrespective of the particle size, it was consistently observed that efficient magnetic flocculation occurs when the magnetic field strength is sufficiently high (Garcia-Martinez et al., 2010).

Previous studies mainly focused on smaller particles, particularly 0.2 – 5 μm colloids, employing enhanced magnetic fields (1 – 12T) (Janssen et al., 2000; Parker et al., 1982). Nevertheless, this also applies to larger particles, like our vivianite (up to 100 μm). Svoboda (1981) proposed a model suggesting a lower magnetic field (0.4 – 1.6 T) for inducing flocculation in larger particles. Tsouris and Scott (1995) and van Kleef et al. (1983) indicated that larger particles enhance flocculation due to increased magnetic force and collision efficiency, and flocculation could transpire across a broad size spectrum without an upper limit. Hencl and Svoboda (1979) observed agglomeration of 30 μm paramagnetic siderite under a magnetic field much lower than 1 T (our operating magnetic field). This suggests that while colloidal particles require a high magnetic field, coarser particles can efficiently agglomerate with a considerably lower field. It is important to note that these studies were conducted in purer systems, whereas sludge contains additional components and significantly higher organic matter content.

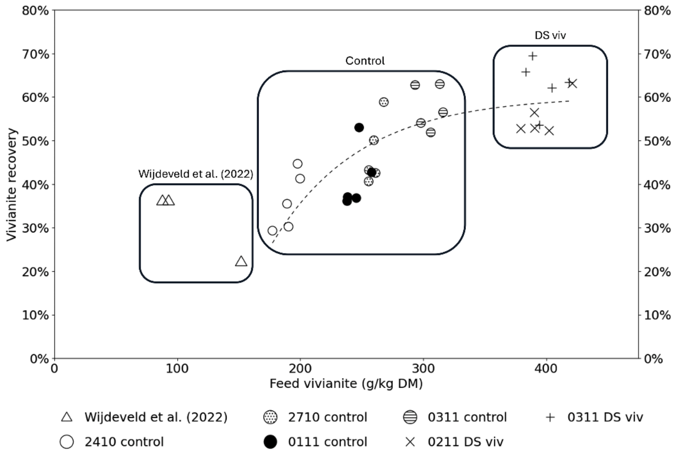

Fig. 2. The vivianite recovery as a function of the feed grade of the control (digested sludge as feed, represented as dots) and when unground vivianite was added to digested sludge as feed (represented as crosses). The triangles are the results of Wijdeveld et al. (2022) at the same operating parameters except flow rate (0.5 m^3/h for them and 0.3 m^3/h for us). Trendline and R^2 (0.66) do not include Wijdeveld et al. (2022)'s results. The recovery increases with feed grade but starts to plateau at 300 g vivianite/ kg DM.

The proposed hypothesis can also explain the higher recovery percentage with increased vivianite feed grade. A crucial factor influencing flocculation is the efficiency of particle collisions, wherein a higher concentration of particles results in increased collisions and, consequently, improved agglomeration (Zhou et al., 2019). Applying this to our case, the increased presence of vivianite particles leads to more frequent collisions, facilitating the flocculation process, resulting in larger particles that are more easily separable.

The plateau in recovery observed when the concentration of vivianite exceeds 300 g/kg DM may be due to several factors. First, the magnet may be mechanically unable to collect all the vivianite in the sludge, a limitation that could be addressed by repeatedly passing the sludge through the separator. Additionally, flocculation efficiency is also constrained by hydrodynamic interactions and variations in particle sizes (Tsouris and Scott, 1995).

3.1.1. *Comparison to the previous Vivimag® pilot study*

Wijdeveld et al. (2022) conducted pilot-scale tests on vivianite separation using digested sludge from Nieuwveer, the Netherlands. This section compares our results with theirs, focusing on recovery with a single pass through the magnet, under identical magnetic field strength, rod diameter, and pulsation frequency, with the only difference being the flow rates (0.3 m^3/h for us, 0.5 m^3/h for them).

A higher feed grade at Nieuwveer led to lower recovery rates, contrary to our findings (88 g vivianite/kg DM at 36% recovery and 152 g vivianite/ kg DM at 22% recovery) (Fig. 2). Differences in sludge sources and, thus, characteristics may contribute to this discrepancy. For example, the sludge from the Nieuwveer study was mainly CPR, while in the present case it was a mix of EBPR and iron-coagulated primary sludge. Regardless of where iron was dosed in the treatment plant, vivianite was formed efficiently in the digester in both cases. It is unknown whether the physical characteristics between EPBR and CPR sludge, such as dry matter, EPS content, etc. can contribute to the differences in recovery between the two studies. Moreover, Wijdeveld et al. (2022) conducted a longer recovery test during a period of rising iron dosage. Their initial two data points, sampled three days apart in January 2019, had lower vivianite levels in the feed. It is hypothesized that at this point, most vivianite in the digested sludge was already formed due to prior Fe dosing. These vivianite particles had the opportunity to mature and grow larger, leading to a higher recovery rate (36% in Fig. 2). As the iron dosing increased, the vivianite concentration in the feed also rose (Prot et al., 2020). However, it could be that they may not have had sufficient time to mature, resulting in smaller sizes that were not effectively recovered by the magnet. This phenomenon was observed when the samples were taken two months after the first two data points, with the recovery rate dropping to only 22%. In contrast, our two-week test had consistent vivianite quality and size, making direct comparisons with their study less valid. Overall, our vivianite purity consistently falls within the 70-85% range, aligning closely with Wijdeveld et al. (2022)'s.

3.2. *The influence of sludge viscosity through changing dry matter*

Fig. 3 compares recovery efficiency between the controls and experiments where the controls were diluted with process water. With dilution, the feed grade remains constant, but the dry matter of the feed sludge was altered, thus changing sludge viscosity (Figure S4). The diluted control experiments still align with the recovery trend observed

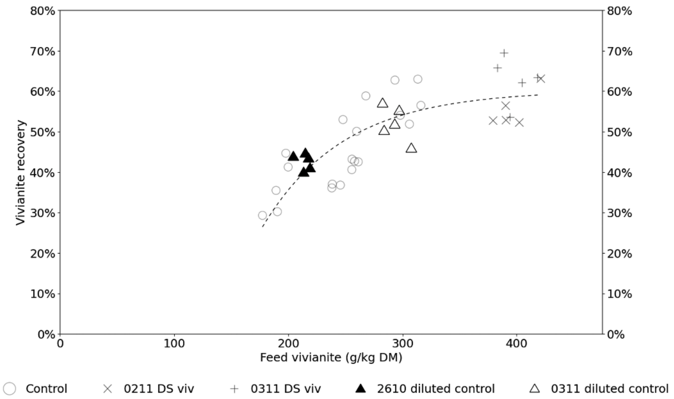

Fig. 3. Vivianite recovery as a function of feed grade in the control (digested sludge as feed, represented as dots) and the diluted control experiments (diluted digested sludge as feed, represented as triangles). R^2 value of the fitted trendline is 0.62. The recovery still increases with feed grade, and dilution did not change the trend.

in the control. This suggests that the dominating factor influencing the recovery was the feed vivianite content and sludge dry matter, and viscosity altered only by changing dry matter, does not play a significant role in the recovery efficiency.

Altering the DM of the feed sludge through different dilutions of the thickened 4% sludge also did not affect the recovery, as depicted in Fig. 4, where the variation among replicates is larger than that of different DM contents. These findings imply that the sludge dry matter does not significantly impact the magnet's performance, and other parameters might substantially influence the recovery.

Given that the feed is sourced from the same thickened sludge but with different dilutions, variations in the feed grade should be minimal, especially for experiments using sludge from the same thickened batch. Fig. 5 shows that most samples ranged from 225-240 g vivianite/kg DM without a significant change in recovery (averaging 50%), while sample 3110 DM 3.2% exhibited unexplainably higher feed grade. Although the feed grade of this sample was not higher than that of the control samples, the recovery was slightly lower but remained within the same range as other samples with varying dry matter contents. While such fluctuations are expected in field samples, when considered alongside Figs. 3 and 4, the data still suggests that feed grade remains the primary influencing factor.

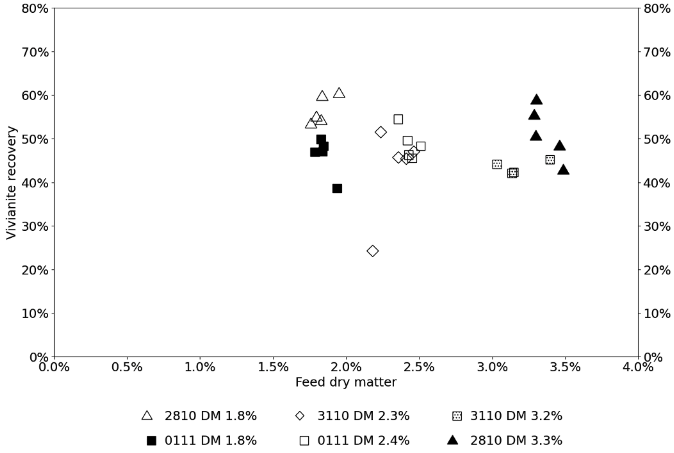

Fig. 4. Vivianite recovery as a function of dry matter in the feed sludge. The same symbols represent sludge that originated from the same thickened sludge batch. The change in DM does not influence recovery.

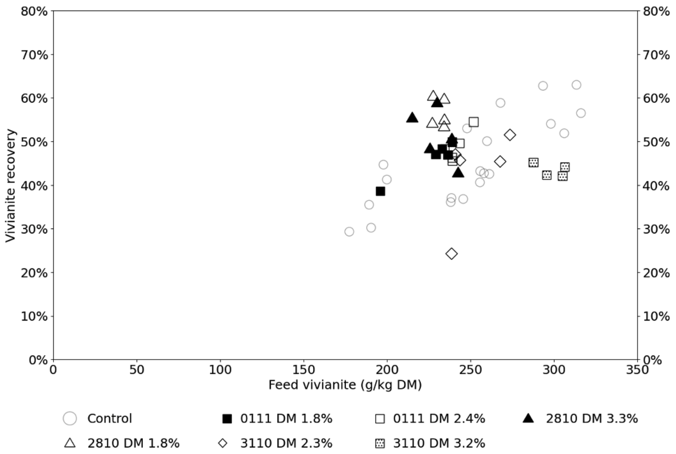

Fig. 5. Vivianite recovery as a function of feed grade for dry matter experiments. The same symbols represent sludge that originated from the same thickened sludge batch. The primary determinant of the recovery is still feed grade.

Through the modification of sludge DM, the viscosity of the sludge was changed, wherein an increase in DM corresponds to an elevated viscosity (Figure S4). This observation aligns with findings from Wei et al. (2018). It can be inferred that alterations in viscosity resulting from changes in sludge DM through dilution do not have a substantial impact on vivianite recovery (Fig. 4).

As previously discussed, the prediction of magnetic separator performance depends on the dominance of magnetic force over competing forces (Oberteuffer, 1974), with drag force caused by viscosity lowering the recovery. In experiments testing the impact of increased viscosity on HGMS performance, Dobby and Finch (1977) and Hayashi et al. (2010) observed negligible effects, with an 8% decrease in recovery for both. Increasing viscosity leads to poorer nanoparticle separation (Roth et al., 2015), it slows particle movement for small particle sizes (100-1000 nm) (Wittmann et al., 2021). While these studies indicate an influence of high viscosity on recovery efficiency, the impact is not overly substantial and mostly applicable to nano-sized particles. Given the higher viscosity, shear dependence, and other distinct characteristics of sludge, these findings may have limited applicability to our case. Moreover, our HGMS includes a pulsating mechanism, which enhances the likelihood of particle-rod collisions, diminishing the significance of the drag force that hinders particle movement toward the rods.

The magnetic versus competing forces model may not fully represent all variables in the multiwire magnetic separator. Deep bed filtration models are considered more accurate, emphasizing particle-rod collisions (Svoboda, 1986). Hu et al. (2023) found higher drag force on suspended particles than on those attached to rods, with fluid pulsation exerting a stronger drag force than the fluid itself (Peng et al., 1992). This highlights the significance of prioritizing the fluid pulsation's drag force for efficient particle detachment from the rods. While the precise shear rate within our magnetic separator cannot be determined, it is anticipated that the high pulsation frequency substantially reduces sludge viscosity because of its shear-thinning behavior, consequently reducing the pulsation drag itself. It could also be that the magnetic force exerted by the rods is strong enough to keep the particles attached to the rods regardless of pulsation and that the vivianite in the sludge is large enough so that the drag force acting on them is not significant.

Importantly, apart from being altered by dry matter, sludge viscosity can also be modified through methods such as polymer dosing or variations in sludge EPS content. However, these aspects are beyond the scope of this study. Our experiments suggest that variations in viscosity due to changes in DM do not significantly affect vivianite recovery. Further research should explore alternative methods for modifying sludge viscosity. Additionally, a broader range of dry matter content variation than the 1.8% to 3.3% used in this study would better highlight the impact of viscosity changes resulting from dry matter alterations.

3.3. The influence of vivianite particle size

3.3.1. When ground vivianite was added to digested sludge

The diverse range of vivianite particle sizes in sludge, coupled with the influence of size on magnetic force, makes understanding how particle size impacts recovery important. Fig. 6 illustrates the comparison of recovery efficiencies when digested sludge was dosed with ground and unground vivianite.

The inclusion of ground vivianite showed no significant deviation from the established recovery trend, starting to plateau beyond a feed grade of 300 g vivianite/kg DM. The recovery did not differ from when unground vivianite was added, where variations in recovery among the replicates were larger than the difference in recovery between adding ground or unground vivianite to sludge. This suggests that particle size may not strongly influence recovery. The primary factor influencing recovery continues to be the feed grade.

The intention behind adding unground vivianite to the sludge was to assess whether the steel rods were saturated with vivianite. As depicted in Fig. 6, this does not appear to be the case, as the recovery percentage continues to rise while the feed grade doubles compared to the controls. If the rods were saturated, a sudden decline in recovery would be evident. Should a sludge exhibit an elevated phosphorus concentration, such as 45 g P/kg DM, with 90% of the P present as vivianite and with excessive iron dosing to maximize vivianite concentration (Fe/P molar ratio of > 1.5) the vivianite concentration would be approximately 330 g/kg DM. This is possible in Scandinavian countries where more iron is dosed in sludge, such as in the study of Wilfert et al. (2018), where Fe/P was 2.5. The concentration of 330 g vivianite/kg DM remains lower than in our study, suggesting that the magnetic separator is adept at managing relatively high concentrations of vivianite in the feed. Nonetheless, it is essential to highlight that the operational flow rate in our study was 0.3 m^3/h, representing only 33% of the magnet's capacity. Under higher flow conditions (1 m^3/h as the magnet's capacity), there is still a possibility that the rods might reach saturation levels, particularly at elevated vivianite concentrations.

Fig. 7 displays the particle size distribution of the recovered vivianite from experiments involving the addition of ground and unground vivianite into the digested sludge. There is no noticeable difference in the size distribution among these experiments, with the D50 values for the particle size of the recovered concentrate only fluctuating slightly between 41.8 and 43.7 µm (Table S2).

This similarity may be attributed to the limited impact of the dosed ground vivianite, given its relatively low quantity. For instance, the "DS viv" experiments show that only 30-34% of the vivianite in the feed originates from the added vivianite, while those for "DS gv" experiments are 39-46% (Table S5). Additionally, the significant difference in particle sizes between unground and ground vivianite is apparent primarily for particles below 30 µm, constituting around 53% of the total particles for ground vivianite and only 29% for unground vivianite (Fig. 7). Therefore, the concentration of particles < 30 µm added from the ground vivianite is relatively small (20 – 24%) compared to the total amount of vivianite already available in the sludge and the fraction > 30 µm in the added ground vivianite, rendering the effect not noticeable and difficult to observe in this context.

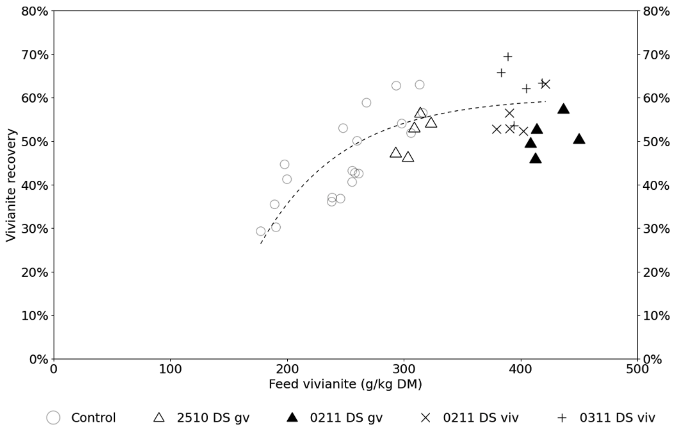

Fig. 6. Vivianite recovery as a function of feed grade when ground vivianite was added to digested sludge (in triangles), compared to when unground vivianite was added (in crosses). R^2 value of the trendline is 0.53. Adding ground vivianite did not change the established trend of increasing recovery with higher feed grade.

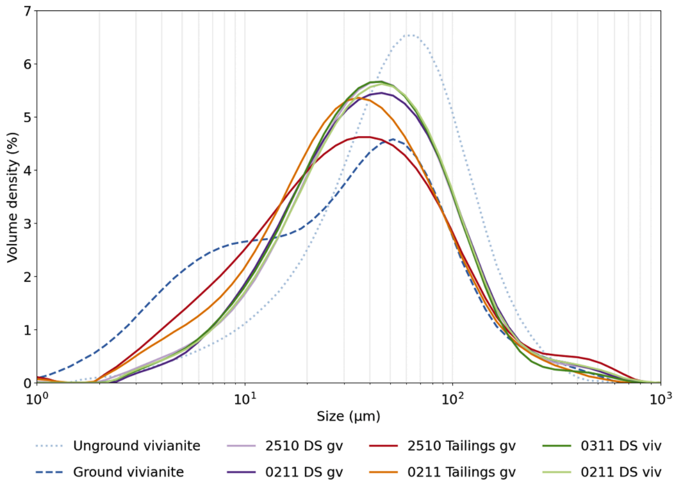

Fig. 7. Particle size distribution of recovered vivianite of the particle size experiments, and the size of the added ground (dash line) and unground vivianite (dotted line). When ground vivianite was added to digested sludge, the recovered vivianite size did not differ from when unground vivianite was added. When ground vivianite was added to the tailings (red and orange curves), the size of the recovered concentrate was smaller.

Since only, on average, 80% of the recovered concentrate comprises pure vivianite, the presence of other materials, such as organics, in the size distribution cannot be quantified, potentially introducing interference with the results. Consequently, based on this data and the recovery percentage, it was impossible to observe a significant impact of small particle size on the recovery when ground vivianite was added to digested sludge. However, given the test conditions, we cannot dismiss the possibility of an effect.

3.3.2. When ground vivianite was added to tailings

For a more precise examination of the impact of introducing ground vivianite without pre-existing vivianite in the background sludge, the ground vivianite was added in vivianite-depleted tailings. The added ground vivianite accounts for a large proportion, ranging from 67% - 73% of all vivianite in the feed (Table S5). This establishes it as the predominant fraction of vivianite, strongly decreasing the background vivianite previously present in the sludge.

Fig. 8 illustrates the comparison of recovery when ground vivianite was added to the vivianite-depleted tailings. Among the duplicates for the "Tailings gv" experiments, the results from 02/11 showed similar recovery values (53.5% on average) observed in other experiments. However, the experiment conducted on 25/10 exhibited notably higher recovery at approximately 70%.

The main difference between the two "Tailings gv" experiments lies in the variation of feed dry matter, shifting from 1.35% to 1.97% for the experiments done on 25/10 and 02/11, respectively. A possible explanation for the high recovery in the 25/10 samples could be the lower viscosity of the sludge at 1.35% DM. As previously mentioned, drag force has a significant impact on smaller particle sizes. Higher viscosity can reduce the recovery in a sludge rich in small particles. In the dry matter study (Fig. 4), the vivianite was unground and therefore larger in

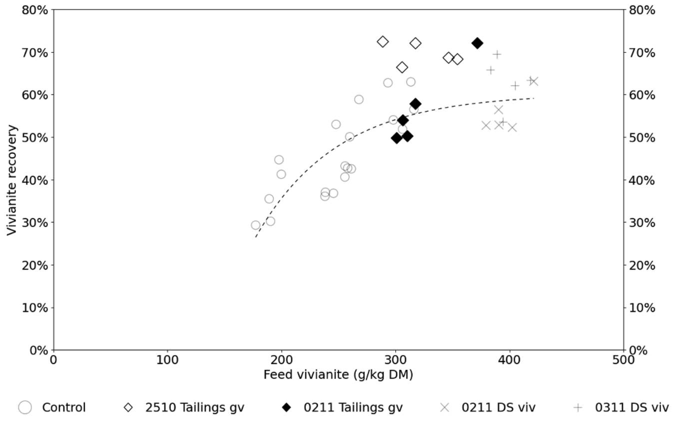

Fig. 8. Vivianite recovery as a function of feed grade when ground vivianite was added to vivianite-depleted tailings (in diamond), compared to the controls (digested sludge as feed). There is a difference in the recovery efficiencies in the duplicate (empty and full diamonds).

size, viscosity had little effect. In such case, the recovery remained unchanged with changing dry matter and viscosity. However, when the particles are ground and smaller ("Tailings gv" experiments), the influence of viscosity is more pronounced, resulting in lower recovery for samples with higher viscosity.

Fig. 7 shows the particle size distribution of the recovered concentrate from the "Tailings gv" experiments. The curves for the "Tailings gv" experiments are shifted toward smaller sizes compared to when ground and unground vivianite was added to the digested sludge. In the 25/10 trial, where a slightly higher percentage of ground vivianite was added, a greater quantity of smaller particles was recovered compared to the 02/11 experiment. Specifically, the percentage of particles < 10 μm for these experiments is 19.4% and 15.9% on 25/10 and 02/11, respectively, whereas this number ranges only around 10.5% for experiments using digested sludge as a base. This suggests the ability of the magnet to recover smaller particle sizes.

3.3.2.1. The recovery rate for each particle size. We conducted a calculation to delve deeper into the impact of recovering smaller particle sizes. The experiment chosen for this analysis is "2510 Tailings gv" because the highest vivianite content in the feed originated from the dosed ground vivianite (73%), ensuring that the effect is observed most prominently. Thus, the particle size distribution of the vivianite in the feed, though cannot be 100% the same, will still have a similar shape to that of the ground vivianite in Fig. 7. Especially, the presence of small vivianite particles below 30 μm should be apparent. This analysis serves as a guide to the trend in the recovery rate for each particle size. By inputting the size of the ground vivianite, we sought to match the corresponding size observed in the recovered vivianite of "2510 Tailings gv."

Arbitrary recovery rates were used for each particle size, and they were selected to match the observed recovered vivianite particle size distribution of the "2510 Tailings gv" experiment as closely as possible. To achieve an interpretable number, we applied normalization to the calculated results for the recovery rate. A value of 1 indicates the normal recovery rate (all particles have the same recovery), values above 1 suggest a relative increase, while values below 1 indicate a lower likelihood of recovery for those particle sizes. To compute the final recovered volume density for the given particle size, the recovery rate was multiplied by the starting volume density:

$$V_f(s) = V_i(s) \cdot R(s)$$

where V_f is the calculated volume density, s is the particle size, V_i is the initial volume density of the ground vivianite, and R is the recovery rate.

The trend of the calculated recovery rate indicates variations in the recovery of different particle sizes (Fig. 9). Particles below 10 μm are less likely to be recovered, while recovery is highly favorable for particles between 10-50 μm. Subsequently, the recovery rate returns to normal and remains constant for larger particles. The analysis of this scenario reveals a preference for recovering medium-sized particles (10-50 μm) over smaller and larger sizes. The prevailing consensus in existing literature supports the fact that greater separation efficiency is associated with larger particle sizes (Arol and Aydogan, 2004; Dobby and Finch, 1977; Maxwell et al., 1981; Xian et al., 2022; Zheng et al., 2017). The diminished recovery of fine magnetic particles is often attributed to the increased drag force acting on them, coupled with the weaker magnetic force keeping them attached to the rods. The improved recovery of medium-sized particles and a slightly lower preference for large particles, as observed in our study, appears not to be commonly documented in the literature. In many cases, larger particles are thought to theoretically exhibit lower recovery rates because the gravitational force causing the particles to settle can surpass the magnetic force attracting them to the rods. However, this is less probable in our study, as the constant up-and-down movement of the fluid through pulsation ensures that the particles remain in suspension.

One possibility is that medium-sized particles align well with the SLon®'s operational design. Additionally, given the 70-86% purity of both the ground vivianite and recovered vivianite, it is also plausible that particles larger than 200 μm may be the entrained organics, considering the absence of such sizes of particles observed under the microscope thus far. The optimal size for recovery is distinct for each case, dependent on factors such as particle shape, magnetic susceptibility, impurities, and the components and viscosity of the carrying fluid.

It is important to note that the recovered vivianite contains particles of all sizes, indicating that the magnet, while exhibiting a preference for larger particles, can still recover smaller sizes. Otherwise, a higher concentration of these particles due to the dosing of ground vivianite would result in a significant reduction in recovery, which was not observed from Fig. 8. It can be said that, overall, the impact of particle size on the recovery is not significant. Beyond the 10 μm threshold, which is the typical vivianite size range in sludge, the magnet encounters no difficulty in recovering vivianite particles.

4. Conclusions

This study highlights the promising potential of vivianite recovery as a robust and versatile pathway for phosphorus reclamation from digested sewage sludge. The principal determinant of recovery efficiency is the vivianite content in the sludge, with higher concentrations correlating with increased recovery, while beyond 300 g vivianite/kg

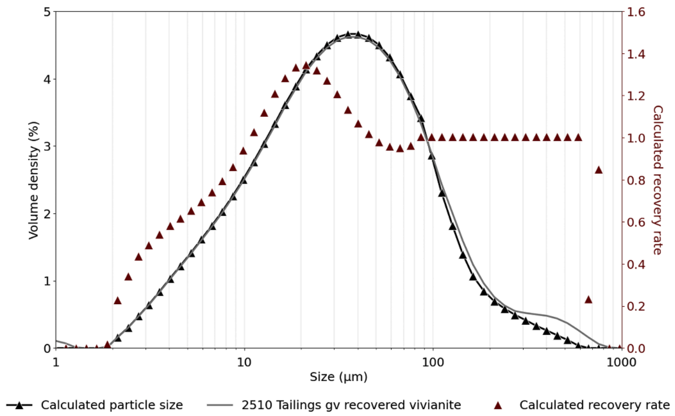

Fig. 9. The calculated recovery rate of each particle size. Vivianite particles $<$ 10 μm are still recoverable but at lower recovery rate.

DM, a recovery plateau is reached. The sludge viscosity variation due to changing sludge dry matter content does not significantly impact recovery. Smaller particle sizes are captured, though less likely to be recovered for particles under 10 μm. The effect of viscosity becomes pronounced for these very small particle sizes. Overall, the technology demonstrates high adaptability and flexibility to changes in sludge, making it well-suited for various conditions across different wastewater treatment plants.

CRediT authorship contribution statement

H. Nguyen: Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **T. Prot:** Supervision, Methodology, Formal analysis, Conceptualization. **W. Wijdeveld:** Methodology. **L. Korving:** Supervision, Methodology, Formal analysis, Conceptualization. **A.I. Dugulan:** Formal analysis. **E. Brück:** Supervision. **A. Haarala:** Data curation. **M.C.M. van Loosdrecht:** Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2024.122407.

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