

Review

The Use of Blast Furnace Slag for Removal of Phosphorus from Wastewater in Sweden—A Review

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Abstract: Research on Phosphorus (P) removal capacity by blast furnace slags (BFS) has been undertaken in Sweden for the last decade. Both laboratory experiments and field trials have been carried out. While laboratory investigations revealed that BFS has a high P-sorption capacity (95–100%), P removal in field trials was much lower, ranging from 40 to 53%. In addition, a number of problems have been observed in BFS field testing including clogging, sulfuric odor and environmental (regulatory) concerns about possible leaching of heavy metals from the slag. In spite of these problems, and questioning by the environmental regulatory authorities, research continues to provide evidence that BFS can be regarded as a suitable filter media, and attempts have also been undertaken in order to further improve the P-removal capacity of this adsorbing material.

Keywords: ageing of slag; chemical improvement; clogging; sulfuric release; wastewater

1. Introduction

In Sweden, as well as in many other countries, excess amounts of phosphorus (P) in water bodies are regarded as a serious threat to the aquatic environment. A large portion of the P released into our water bodies can be derived from single households with poor wastewater treatment in the country-side [1]. To prevent P input to surface water bodies or the ground-water, small-scale and low-cost technologies have been sought for. Following extensive research on P sorbing materials by Johansson [2–5], during the last decade, a number of researchers in Sweden continued to investigate the use of filter substrates that can be included in treatment facilities such as constructed wetlands (CW)

or other on-site filtration systems [6-10]. These researchers investigated a number of different filter materials (both natural and man-made materials) available in Sweden, with regard to their capacities to remove P from a variety of wastewaters. Among these materials, blast furnace slag (BFS) has attracted attention by several researchers, not only in Sweden but also elsewhere in the world [11-17].

The Swedish results, mainly based on laboratory investigations in which removal rates ranging from 95–100% have been observed. Problems with the use of BFS have been observed in field investigations. Three field trials of varying sizes have also been described in the literature, see [18-20]. The results from these field trials have not been as successful with regard to P-removal as the results obtained in laboratory investigations. Removal rates from the field investigations have been reported to range between 40–53%. Some of these problems were not associated only with the use of BFS but also with the design of the treatment system [19].

Discrepancies between P retention capacity estimates from laboratory investigations and the actual treatment performance in the field have been observed in other studies worldwide [21]. For over a decade, Drizo *et al.* [22] have been advocating that estimating the maximum P adsorption capacity of the filter media using standard 24 h batch experiment and Langmuir and Freundlich isotherms leads to unrealistic estimates of the materials P adsorption capacities. Yet, researchers continue to employ this method, which results in frequent failures in the field [22].

The aims of this paper are (i) to present a brief review of the Swedish research on BFS with special regard to P-removal; (ii) to present and discuss problems that occurred when using the slag material in field trials and (iii) to present possible measures that can be taken in order to improve the use of slag material for P-removal.

2. Materials and Methods

2.1. Materials

BFS is an industrial by-product that is generated in the production of iron at steel plants. Similarly to other parts of the world, the major use of steel slag aggregates in Sweden is for construction of roads and buildings. There are two steelworks in Sweden, one in Oxelösund, approximately 100 km south of Stockholm and one in Luleå, approximately 800 km north of Stockholm. Both are run and operated by Svenskt Stål AB (SSAB). Both steelworks produce BFS materials: on a yearly basis 500,000 tones of slag is produced by SSAB in Oxelösund; the corresponding figure for the steelworks in Luleå is roughly 250,000 tones. The chemical composition of the slag produced in Oxelösund and the slag produced in Luleå are quite similar (Table 1).

Table 1. Example of the chemical composition of the crystalline blast furnace slag materials (0–8 mm) produced in Oxelösund and Luleå [23].

| Chemical Components | Oxelösund | Luleå |
|--------------------------------|-----------|-------|
| SiO ₂ | 34% | 33% |
| CaO | 30% | 32.5% |
| MgO | 16.5% | 16.5% |
| Al ₂ O ₃ | 13% | 12.5% |
| TiO ₂ | 2.3% | 2.3% |
| S | 1.0% | 1.1% |
| MnO | 0.6% | 0.3% |
| FeO | 0.5% | 0.2% |

The pH values of the Swedish BFS range between 10.2–10.3 for crystalline slag, and 10.3–10.6 for amorphous slag [3-5].

2.2. Methods

Swedish researchers have investigated P-removal in batch and column studies in the laboratory. A few investigations on a larger scale, e.g., pilot tests and field trials have also been carried out. The reader should consult original papers for more detailed information on a particular experimental set up.

2.2.1. Laboratory Batch Tests

In Sweden, batch tests, e.g., agitation of filter material and P-solution or wastewater (with a pre-determined ratio) in flasks, on BFS slag were first carried out by Johansson [3]. In these tests, laboratory made P-solutions were used in various concentrations (ranging from 5 to 25 mg PO₄-P/L) mimicking the P-concentration in a real-life municipal sewage wastewater. Various amounts of slag materials, e.g., amorphous and crystalline slag in different particle sizes from the steelworks in Oxelösund were tested. Other researchers [7] have used different slag materials, particles sizes and ratio between amount of slag and P-solution. Hedström and Rastas Amofah [24] also carried out batch tests using real-life wastewater, mechanically and biologically pre-treated in a wastewater treatment plant. Also, Nehrenheim *et al.* [18] used real residential wastewater, only mechanically pre-treated, from a single household.

2.2.2. Column Test

Based on the initial results from the batch tests, long term column investigations were carried out by Johansson [4]. Four columns were filled with crystalline and amorphous slag with different particle sizes (ranging from 0–4 mm); the columns were fed with P-solution containing 10 mg P/L over a 13 months period. Samples were taken several times a week and analyzed for dissolved phosphorus (PO₄-P). The aim of the study was to investigate the long-time performance of P-removal by two different BFS materials. Following these experiments, Gustafsson *et al.* [25] evaluated the performance of BFS and several other materials, e.g., Filtra P, Polonite® and natural wollastonite with regard to P-removal using a column system also fed with a laboratory made P solution, containing

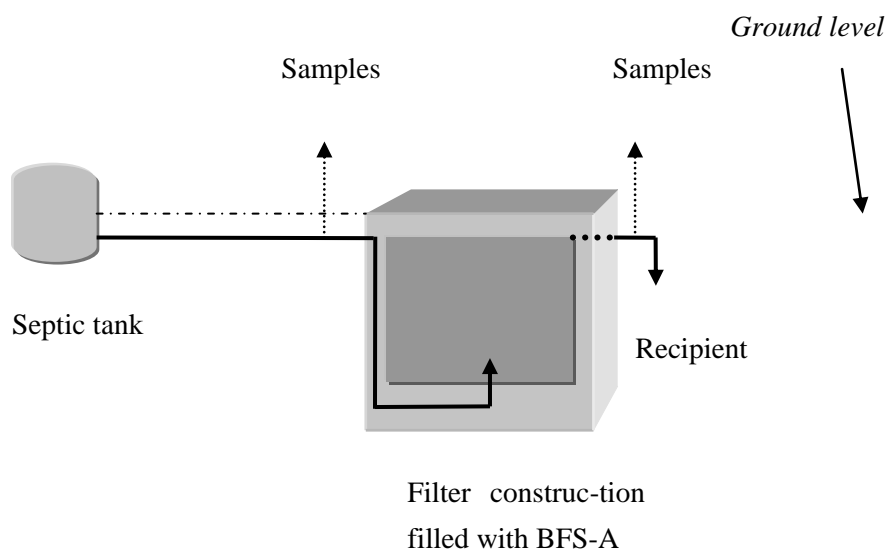
5 mg P/L. Column experiments using real-life wastewater have also been carried out by others; see for instance [26,27].

2.2.3. Field Trials

Slag filter as a septic tank P removal upgrade for a single household:

Nehrenheim *et al.* [18] used domestic wastewater effluent to investigate BFS for P removal in a pilot scale field application. Wastewater (P-concentration average 10 mg P/L) from a single household in the Södertälje area south of Stockholm was treated in a septic tank before being pumped to an underground one cubic meter filter box filled with 1.6 tones of amorphous slag, diameter size 0.25–4 mm, see Figure 1. The estimated flow through the filter box was 500 L/day, resulting in a residence time of ~21 h/day; the system was monitored for 70 days.

Figure 1. Experimental set-up of filter box system for P-removal [18].



Steel slag as filtration material in Constructed Wetland treating residential wastewater

Rastas Amofah and Hanæus [19] used two filter materials, e.g., BFS and Norwegian Filtralite, in a small constructed wetland (CW) system of approximately 65 m² outside Luleå in the northern part of Sweden. The CW system was fed with treated residential wastewater from a village. The average concentration of tot-P was 6.6 mg/L. Firstly, the pre-treated wastewater passed a willow bed (volume 26 m³, covering surface area of 32 m²) before being transferred to the filter beds. The slag filter bed had a height of 0.9 m, an area of 2.4 m², thus a volume of 2.2 m³. The Filtralite bed had the same size. The wetland system was fed with 600–900 L wastewater/day during the first four months of operation, thereafter the flow decreased to 300–600 L/day.

Steel slag filter as filtration material in Wastewater Treatment System

In 2002, the Swedish Road Administration (now the Swedish Transport Administration) constructed a service area, Ångersjön, along the European highway E4. The area is a wildlife area

where public toilets and a small café are located. The wastewater streams from the toilets and the café are urine-separated; the urine is transferred to a tank for future use in agriculture. The remaining wastewater is transferred to a septic tank and later on to a treatment facility consisting of a lime gravel horizontal filter followed by a vertical pre-filter (LECA) intended for removal of particles, BOD (biological oxygen demand) and pathogens [20]. This filter promotes sorption of phosphorus and killing of pathogens. After having passed these filters, the wastewater is transferred to a well from which it is pumped to two parallel P-filters, one containing BFS, the other Filtralite-P. The P-filters are constructed as troughs with a bottom area of 16 m² and a surface area of 64 m². After treatment in the P-filters, the wastewater is discharged into a small stream.

3. Results

3.1. Laboratory Batch Tests

The results from the Swedish investigations showed that BFS materials remove P from both laboratory made P-solutions and real wastewaters to various extents. In results derived from laboratory batch experiments, P adsorption capacity was estimated to range between 25 and 100% when using laboratory made P-solutions. The corresponding adsorption when using real-life wastewater ranged between 35 and 70% (Table 2).

Table 2. P retention capacity estimated from laboratory batch experiments using laboratory made P-solutions and real-life domestic wastewater.

| Slag material | Particle size (mm) | P-source | | P-conc. (mg/L) | P-removal (%) | Reference |
|--------------------------------------|--------------------|------------|------------|----------------|---------------|-----------|
| | | P-solution | Wastewater | | | |
| BFS-A ¹ (O ²) | 0–0.125 | x | | 5–25 | ~40–100 | [4] |
| BFS-A (O) | 0.25–4 | x | | 5–25 | ~25–60 | [4] |
| BFS-C ³ (O) | 0–0.125 | x | | 5–25 | ~60–95 | [4] |
| BFS-C (O) | 0.25–4 | x | | 5–25 | ~70–95 | [4] |
| BFS-C (L ⁴) | 0.5–2 (fresh) | x | | 0–20 | ~100 | [24] |
| BFS-C (L) | 0.5–2 (weathered) | x | | 0–20 | ~40–85 | [24] |
| BFS-C (L) | 1–5.6 (weathered) | x | | 0–20 | ~40–85 | [24] |
| BFS-C (L) | 0.5–2 (fresh) | | x | | ~55–70 | [24] |
| BFS-C (L) | 0.5–2 (fresh) | | x | | ~35–45 | [24] |

¹ A = Amorphous, ² O = Oxelösund, ³ C = Crystalline, ⁴ L = Luleå

These results are in agreement with results obtained by international researchers; see for instance Vohla *et al.* [28] for an overview.

3.2. Laboratory Column Tests

In long-term column tests using laboratory made P-solutions, removal rates as high as 95–100% have been observed [4]. There were small differences in P-removal between crystalline and amorphous slag. At the end of the experiment, no signs of break-through (e.g., when the concentration in the influent is equal to the concentration in the effluent) could be detected and Johansson concluded that

the slag materials were not saturated (Table 3). Gustafsson *et al.* [25] reported an average P-removal of 86%, while there were hardly any differences between columns fed with P-solution or columns fed with real-life wastewater in the experiment carried out by Nehrenheim *et al.* [18]. The results reported by Renman *et al.* [27] also varied.

Table 3. P removal in longer term column studies fed by laboratory made solution and real wastewater.

| Slag material | Particle size | P-source | | P-conc. (mg/L) | Operation time (weeks) | P-removal (%) | Reference |
|--------------------------------------|---------------|------------|------------|----------------|------------------------|---------------|-----------|
| | | P-solution | Wastewater | | | | |
| BFS-A ¹ (O ²) | 0.25–4 | x | | 10 | 56 | ~95–100 | [4] |
| BFS-C ³ (O) | 0.25–4 | x | | 10 | 56 | ~95–100 | [4] |
| BFS-A (O) | 0–4 | x | | 5 | 68 | 86 (average) | [25] |
| BFS-A (O) | 0.25–4 | x | | 10 | 1 | 97–100 | [18] |
| BFS-C (O) | 0.25–4 | | x | 20–30 | 1 | 97–100 | [18] |
| BFS-A | 0.25–4 | | x | ~0.4 | 10 | >98 | [26] |

¹ A = Amorphous, ² O = Oxel ösund, ³ C = Crystalline

High removal of P has also been reported from column experiments carried out outside Sweden; see for instance Vohla *et al.* [28] for an overview.

3.3. Field Testing of Blast Furnace Slag

Results observed in pilot and field trials also varied. In the pilot experiment described by Nehrenheim *et al.* [18], less than 40% of the added P expressed as PO₄-P was removed by the slag, while less than 20% of the added P was removed in the wetland experiment described by Rastas Amofah and Hanæus [19]. Wastewater had a more complex composition than the P-solution, contributing to a lower P-removal. A wastewater containing a high concentration of suspended material can easily clog the filter media. Nehrenheim *et al.* [18] also suggested that channeling, e.g., waterflow through parts of the filterbed rather than the whole bed, thus reducing possibilities for P-sorption to occur, might play a crucial role for lower treatment efficiency in field applications. In addition, weather conditions such as snow melt and temperature might also have an impact on the results obtained under field conditions [24].

Compared to results from international field trials, the Swedish results are somewhat low, see Table 4. In a Turkish investigation, Korkusuz *et al.* [15] investigated blast furnace slag in a CW system and observed that total-P was removed to 45%. The corresponding figure reported by Shilton *et al.* [30] was 77% over a period of five years. In this case, melter slag was used in a full-scale treatment plant in New Zealand. It is however difficult to compare these results since different researchers have performed their investigations in different ways, a fact that has been stressed by Drizo *et al.* [22] and Johansson Westholm [30].

Table 4. Results from field trials using blast furnace slag materials.

| Field site | Slag material | Particle size (mm) | Waste-water | P-conc. (mg PO ₄ -P/L) | P-load (m ³ /day) | Operation time (weeks) | P-removal (%) | pH In/Out | Reference |
|------------|---------------|--------------------|---|-----------------------------------|------------------------------|------------------------|---------------|--------------------|-----------|
| Södertälje | BFS-C (O) | 0.25–4 | Domestic wastewater pre-treated in a septic tank | 20–30 | 0.5 | 16 | 37.5 | 7–8/8 | 18 |
| Luleå | BFS- (O) | 4–7 | Primarily pre-treated domestic wastewater from a village | ~5 | 0.5 | 64 | <20 | 7.4/10.7 | 19 |
| Ångersjön | BFS- (O) | 2–4 | Pre-treated wastewater (urine separated) in a septic tank and in a vertical lime filter | 1–11 | ~3 | ~150– | 57 | 7.3–7.8/ 7.8–11 | 20 |

4. Discussion

4.1. Problems and Possibilities

Swedish researchers have demonstrated that BFS has potential as a filter media for removal of phosphorus [2-10]. The use of BFS as a filter media for P removal has not been without questions, however. It is a well-known fact that steel slag material contains heavy metals, thus potential leaching of metals from BFS into the environment ought to be investigated. The slag materials have also proved to clog in field trials, indicating that particle sizes larger than 4 mm should be used; in addition, leaching of sulfuric components have been observed in one study [19].

4.1.1. Environmental Hazards

As steel slag aggregates represent an industrial by-product from steel slag production, where iron ore and scrap metal are mixed together at high temperatures to produce steel, one might fear that the slag might leach heavy metals into the environment. A study on metal leaching from blast furnace slag materials has been published in Sweden [31]. This investigation showed that although BFS do leach metals, this occurs to a very low degree. The leaching from the slag was found to be in the same range as the corresponding leaching from natural gravel. Similarly, the Landcare Research Institute from New Zealand, and National Steel Slag Association in United States, performed comprehensive research on metal leaching from eight different steel slag materials [32-35]. These studies also showed that leaching from steel slag is not detrimental to environment or human health.

Although steel slag material is currently used as fertilizer and/or as a liming agent within the agriculture in Sweden and other parts of the world, governmental agencies are reluctant to accept its use in water pollution control. For example, in Sweden, steel slag-based fertilizer is marketed as ecological sound according to KRAV, a Swedish member of International Federation of Organic Agriculture Movements (IFOAM). In the United States, the use of steel slag material as a soil amendment RECLIME® has been approved in 14 States [35]. Yet, according to the Vermont and New York Departments of Environment and Water Quality Solid Waste Management Section, beneficial use permits and approvals must be obtained from the governmental officials prior to any further field testing of steel slag for water pollution control [36].

4.1.2. Field Performance Issues

It is not unusual that clogging appears in filter beds due to the settling of particles in the wastewater, especially at the entrance of the filter [24]. These authors observed, in a pilot scale experiment, that clogging occurred, and suggested an efficient pre-treatment of wastewater before it enters a slag filter. Clogging of the filter material was also observed by Nehrenheim *et al.* [18]. In their field trial, the pre-treatment of the wastewater in a settling tank was probably insufficient and is therefore the most plausible reason for the clogging and thus the termination of the pilot trial. Alternatively, one can use larger particle sizes (4–7 mm, or larger), but in this case the specific surface area will be smaller and offer less sorption sites, thus reducing the P-removal.

Blast furnace slag (BFS) is also known to contain sulfuric compounds, see Table 1. Under oxidizing conditions, these compounds can be leached and this phenomenon was reported by Hedström and Rastas [23] in a pilot-scale experiment. Rastas Amofah and Hanæus [19] also observed this phenomena in their wastewater treatment facility in which real-life wastewater was fed to a BFS filter bed. In this treatment facility, sulfuric odor was also a problem, especially in the beginning of the experiment. Hedström and Rastas [24] concluded that the release of sulfuric compounds must seriously be taken into consideration if blast furnace slag is going to be used as filter media in wastewater treatment applications since the sulfuric compounds might harm the groundwater quality negatively. Since this problem has been observed by a few researchers, further studies on this issue are therefore needed in order to find out whether BFS is a suitable or un-suitable filter material with regard to its content of sulfuric compounds.

4.1.3. Improving Efficiency

Even though Swedish researchers have reported various, but mainly positive, results with regard to P-removal by BFS, the material could be improved in order to further optimize the P-removal capacity. Attempts to improve the P-removal capacity of different filter media have been reported in the international literature. Brogowski and Renman [37] suggested opoka, siliceous bedrock, heated in a way that transformed CaCO_3 into CaO , which is a more advantageous form of calcium to which P can be attached. Compared to BFS materials that have been produced in other parts of the world and investigated by researchers outside Sweden, e.g., in Japan [38,39], Australia [11] and Germany [13] the Swedish slag is rich in MgO and Al_2O_3 , but have much lower CaO content [5]. If the CaO content in the Swedish produced BFS could be higher through some kind of treatment, larger amounts of P might be removed by the BFS.

Chemical pre-treatment would be another way to improve a materials capacity to remove P. Swedish researchers are currently investigating whether hydrothermal treatment of BFS can enhance the P-removal capacity. The hydrothermal method is based on the formation of tobermorite, a mineral which is rare in the nature. Initial tests have shown that this converted slag material has higher P-sorption capacity and in addition, also a prolonged lifetime compared to untreated slag [40]. According to G. Renman [40], further studies have to be performed to confirm these results.

Another important observation made by Swedish researchers is that the age of the steel slag used in field applications can affect their P-removal capacities. For example, Hedström and Rastas [24] observed that freshly quenched slag material demonstrated higher P-removal capacity than did older slag that had been stored outdoors for a certain period of time. Similar results were observed in a study where blast furnace sludge was tested for its capacity to remove metals from multi-metal contaminated water [41,42]. The processes that are responsible for the deterioration of the P-removal capacity of BFS are not known. Hedström and Rastas [24] suggested that the BFS is depleted of calcium (Ca) when being exposed to weather conditions such as rainfall or snow melting and suggested that ageing of slag material is a factor that has to be taken into consideration when being considered as filter media. Kalmykova *et al.* [42] also concluded that the age of the blast furnace sludge, should be considered in order to optimize the removal of metals.

5. Summary and Conclusions

From the results observed and reported by Swedish researchers on the use of BFS for P removal from wastewaters, it can be concluded that slag materials produced in Sweden remove P to various extents depending on laboratory or field conditions. High removal rates have been reported from laboratory investigations while lower removal capacities have been observed in field trials. Problems that have arisen during the experiments are related to hydraulics, e.g., clogging effects, and possible leachate of metals and sulfur. Possible measures for improvement of P-removal can be either testing of larger particle sizes or pre-treatment of the slag material prior to its use in on-site treatment system applications and selection of fresh slag material.

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