IOP Conference Series: Materials Science and Engineering



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**Abstract.** Limestone is originally generated from sedimentary rocks that are mainly made from $CaCO_3$, the latter in turn is made from calcites and aragonites. This chemical composition of limestone makes it a suitable material for water/wasters treatment as these elements are identified for their capability to adsorb several pollutants. Although, limestone is environmentally sustainable material and is broadly applied in water filtration, it is has a moderate affinity for heavy metals. Thus, due to this serious drawback, limestone becomes less attracting for researchers. Therefore, this study is aiming at producing an environmentally sustainable filtration system by mixing limestone and activated carbon, and applies it to remove heavy metal (iron) from synthetic wastewaters. The ability of the new filtration media, which was made of 50% activated carbon and 50% limestone (1:1 ratio), to remove iron from 10 mg/L synthetic iron solution. The latter was created using ferrous sulphate heptahydrate and deionised water. The removal of iron was optimised for the influence of the absorbent dosage (AD) (500-1000 g), detention time (DT) (10-120 minutes) and pH of solution (4-10). The outcomes of the experiments evidenced the capacity of the new filter to efficiently remediate wastewater from iron. Where, 1000 g of this filter needed only 95 minutes, at pH of 6.0, to completely remove iron ions from the synthetic solution.

## 1. Introduction

The release of substantial amounts of hazardous waste, particularly liquid and solid material, into the environment, has escalated a world-wide worry mainly in recent years [1-6]. Due to the inadequate amount of freshwater in the Earth's planet, discharging of untreated or poorly treated wastewaters are categorised as the greatest threat to human existence and civilization [7-11], and also to the aquatic life [12-14]. Currently, human beings are facing a grave concern because of the pollution of freshwaters, and this concern is growing on a daily basis because of the growth of industrial, farming and urbanising activities that consumes huge amounts of water and discharge highly contaminated effluents [3, 10], and because of climate change that maximises water consumption [15-18]. Moreover, the poor treatment of these effluents, especially in developing countries, plays substantial roles in contaminating the sources of freshwaters [19-21]. Therefore, these activities maintain the continuous pollution of the limited sources of freshwater with various hazardous contaminants [22-26], and consequently some harmful contaminants accumulated to hazardous levels in the water environment, resulting in serious health risks on the consumers through drinking the polluted water or through food chain [27, 28]. Amongst these many contaminants, heavy metals are considered to be one of the most hazardous contaminants in wastewaters and freshwaters [3, 29, 30]. This isn't only due to their negative impacts on the eco-system, but also because of their fatal impacts on the health of the consumers, such as human, even at relatively small intakes [29, 31]. In addition, most of these pollutants are not biodegradable and could be straightforwardly accumulated in the bodies of different species of aquatic life. For instance, some heavy metals like mercury and arsenic are reported to be quite toxic to the consumers when exceeding the allowable limit, usually on a microgram scale, and they can cause some genetic and rare diseases beside the fatal illnesses like cancer [32]. Additionally, they can easily reach a toxic level inside the bodies of different aquatic creatures, and hence, to the human being through the food chain [29]. Hence, elimination of these pollutants them from freshwaters and wastewaters represents a matter great importance.

The effective treatment of wastewaters and freshwaters and controlling the quality of drinking water are key factors in mitigating the freshwater shortage [15, 17, 29]. Therefore, the development of a sustainable and consistent method of treating wastewater is of world-wide attention. Adsorption processes are currently considered to be sustainable technologies to be used in treating wastewater for a number of reasons [22]. Firstly, they are effective in separation of different types of contaminants in wastewater. Secondly, the possibility of using natural materials that makes the adsorption process easier and cost-effective. Last but not least, the process considered to be more environmentally friendly than other methods due to the opportunity of recycling the adsorption materials [22, 33-38].

This study is therefore aiming at the use of a new hybrid filter, which utilizes equal amounts of activated carbons and limestone, as an effective and economic method that could be applied to treat synthetic wastewater containing heavy metals (iron as a model pollutant). Limestone was used as environmentally sustainable material, but with low adsorption capacity for heavy metals. While the activated carbon, which is usually expensive and requires chemicals to activate, was used here due to its high capacity for heavy metal adsorption. In summary, the suggested filter balances the sustainability and efficiency goals.

## 2. Methodology

### *2.1. Simulated samples*

In the present study, synthetic wastewater models were used to test iron removal using the new hybrid filter. At the start of the tests, a 1500 mg of $FeSO_4.7H_2O$ powder was placed in a 1.0L container and then a litre of deionised water was decanted into this container producing a 300 mg/L iron stock solution [29]. Due to the possibility of bacterial growth in the concentrated solution; this concentrated solution was stored refrigerated (≤2 Celsius). On the day of experiments, a proper volume of this stock solution was taken and diluted in deionised water to produce 10 mg/L iron solution. The later was used to run the filtration experiments.

The tests were conducted in a batch flow conditions considering the effects of AD, DT and initial pH of the diluted samples. The effects of each one of the mentioned parameters using 3 diluted samples. Due to the limited laboratory time, the temperature influence was not considered in this investigation, thus all tests were conducted at a temperature of 20.0ºC. Hydrochloric diluted acid and sodium hydroxides were applied, in this research, to adjust the pH of samples [29].

### *2.2. Adsorption experiments*

The new hybrid filter was installed at the industrial laboratory in Liverpool John Moores University. As stated before, this new filter was manufactured by mixing equal amounts of activated carbons and limestone (1:1 ratios). In addition, water/solid ratio was adjusted to 0.55 as recommended in past studies [22]. The first studied parameter was the pH; the experiments were run at pH of 4, 5, 6, 7, 8, 9 and 10 for a constant DT of 60 minutes and AD of 750g. The new filtration material was placed in 2 liters vessel; then 1000 mL of iron solution was decanted into the vessel. To prevent any external contamination, this vessel was closed and left in the laboratory for the required DT.

Every 20 minutes, iron removal was measured by taken a suitable volume (few millilitres) of the being filtered solution from the new filter. The taken samples were filtered at No.2 Whatman paper filters then at 0.45μm Sigma-Aldrich filters to remove any undesirable materials and contaminants [29]. Measurement of the residual iron concentration was conducted using typical cuvette test specifically for

iron (LCK 320 and 521) and a spectrophotometer device. To ensure the results are precise, the test was repeated twice.
The effect of DT was then examined by treating the iron solution for durations from 10 to 120 minutes. The test was conducted at the optimum pH level from the prior tests with the same iron concentration. The removal of iron was determined according the sated method in pH experiments.
Lastly, impact of the AD was examined by conducting experiments with the optimum solution pH and DT but with a different AD (500, 750 and 1000 g).
Following these groups of tests, the total removal of iron from synthetic solution using the new filter was calculated by equation (1) [29].

$$Iron\ removal\ \% = \frac{Initail\ concentration - Final\ concentration}{Initail\ concentration} \times 100\% \quad (1)$$

## 3. Results and discussion

### *3.1. Influence of pH*

Figure 1 shows the change in the remediation of the synthetic solution from iron, using the new filter, with the change in the initial pH. These tests were performed at a pH of 4 to 10 with constant DT of 60 minutes and AD of 750g per 1000 mL of the solution. The resultes showed that as the initial pH increased from 4 to 6, iron concentration decreased by about 58% and 62%, respectively. However, this trend did not continue as iron removal clearly starts to decendinding after pH of 7. For instant, the iron concentration dropped by about 60% as the initial pH value increased to 10. It could be conculded that the optimum value of initial pH to get the best removal of heavy metals (iron) using the new filter is ranging between 5 and 7. It means the new filter removes this type of pollutants efficiently is in favoure to a nueltral pH. This could be explaind by the fact that at low pH values the concentration of hydrogen ions ($H^+$) while higher ones will increase the concentration of hydroxide ions (OH-). These two speciouse compete with iron ions on the surface of the filter material for the adsorption sites [39]. For the new filter, the pH of 6.0 is therefore the optimum initial pH value.

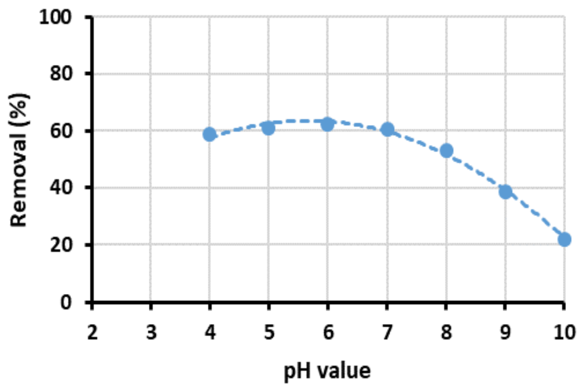


**Figure 1.** Relationship between iron removal and pH of solution.

### *3.2. Influence of DT*

The influence of different DT, 10 to 120 minutes, was examined on iron removal using the new filter, keeping the initial pH and AD fixed at 6.0 and 750 g, respectively. The results presented in figure 2 below clearly show that the pollutant (iron) removal efficiency, dramatically, ascended to more than 80% before reaching 100 minutes of treatment. This dramatic increase is likely due to the amount of available adsorption sites on the filtration media surfaces. However, the efficiency has followed a flat linear pattern after that. This can be explained by the fact that the surfaces of the particles of the new filtration material (activated carbons and limestone) were almost saturated with the iron (or what is known in the literature as the equilibrium mode) after 100 minutes of treatment [22]. According to these results, the best DT for iron removal using the new filter (activated carbon-limestone filter) is 100 minutes.

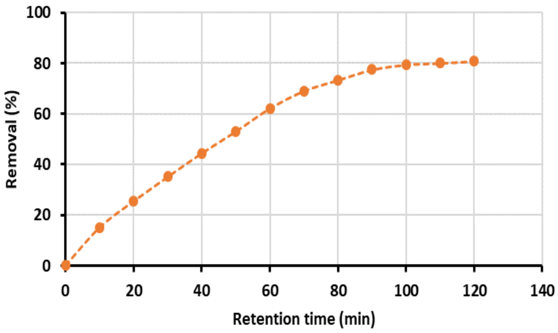


**Figure 2.** Effect of DT on iron removal by the new filter.

*3.3. Influence of AD*

The final step in this study was about the influence of the AD on the ability of the new hybride filter to absorb iron from solution. In this step, three different weights (500, 750 and 1000 g) of the new filteration materials (activated carbon and limeston) were used to treat the iron solution, at the same time the initial pH and DT were fixed at 6 and 100 minutes, respectivelly. Figure 3 demonstrates the effects of each one of the mentioned weights on the remediation of solution from iron. The resultes show similar removal trends for each one of the studied AD, but with different uptakes. Generally, the adsorption of iron was increased with the increase of the used AD, which suggests a direct relationship between the increase in AD and the increase in iron removal. Additionally, it can be clearly obsereved from this figure that using a AD of 1000 g achieved iron removal of 100% in 95 minutes, while the other ADs achieved less removals, about 80% and 60% for 750g and 500g, respectivally. This is can be explained by the fact that the number of vacant active sites increases with the increase of AD, which improves the adsorption of iron in turn, and vice versa [40]. According to the outcomes of the conducted results, the new hybrid filter could compeletely remove heavy metlas (iron as a model pollutant) from solution at the following conditions: initial pH of 6.0, DT of 95 minutes and AD of 1000 g.

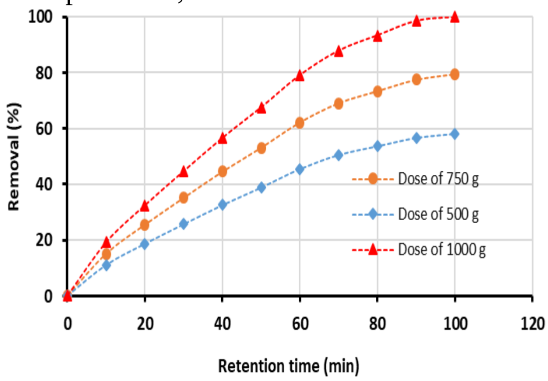


**Figure 3.** Effects of AD on iron removal using the new hybrid filter.

In summary, the new hybrid filter could be used as efficient, environmentally sustainable and cost-effective manner for remediation of solutions from heavy metals. Additionally, the authors suggest that sensing technology, which has been widely used in various applications [41-45], could be used to track the saturation of the media of the new filter, which ensures more accurate and efficient performance of the new filter.

## 4. Conclusions

The findings of the present research could be an incidence about the suitability of the new hybrid filter, which utilises activated carbon and limestone, to remediate solutions from heavy metals at a reasonable cost and detention time. Additionally, the new hybrid filter could meet the requirements of sustainability and treatment efficiency.

In terms of performance, the outcomes indicated that the new filter could work efficiently when the pH of solution is neutral, AD of 1000 g and DT of 95 minutes.

Finally, for future studies, the new filter could be utilised for remediation of freshwater or wastewaters from other pollutants or a group of pollutants. Additionally, some studies should be allocated to check the effects of solution temperature on the performance of the new hybrid filter.

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