ISBN-978-989-9121-11-9
30th BARCELONA International Conference on "Agriculture, Biology, Environment & Natural Sciences" (ABENS-22)
Barcelona (Spain) Sept. 20-22, 2022

Effects of Physico-chemical Parameters and Acid Rain Water on the Mobility of PAHs from Coal Tar

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Abstract**:** *In this study coal tar from the coking process was exposed to acid rain water at various solid/liquid ratios, temperatures and time. It was found that a ratio of 5/95 suitable for maximum mobilization of PAHs; although there was no consistent trend of the effect temperature, it was however, evident that better mobilization of PAHs was achieved above room temperature (25 ℃); the maximum mobilization of PAHs was achieved at 45 ℃. Naphthalene was the most abundant PAH in the leachate while Benzo(a) pyrene which is one of the most toxic PAH was easily mobilized in more dilute solution. It is therefore proven in this study that the extent of mobilization of PAHs from coal tar in receiving water and hence the impact on the environment will depend on the physico-chemical properties of the water; high temperature and dilution of coal tar are most likely to promote the mobilization of PAHs and their dispersion in the environment*

Keywords: *Coal tar, PAHs, mobilization, acid rain water, physico-chemical conditions*

1. Introduction

Natural security and amendment of ecological issues are significant inquiries for a compelling change of life quality and for feasible improvement [1, 2, 3, 4]. Shortage of fresh water has been an overall worry for a long time [5, 6, 7, 8, 9, 10, 11, 12]. It has been reported in the literature (UN Environmental program report) that by 2019, populations will live submerged or focused on conditions. The Department of Water Affairs and Forestry [13] in South Africa reported that the country is a semi-dry nation having a normal precipitation, which is 52% of the world normal. In the course of the last few decades, expanding mechanical exercises have brought about an expansion in the release of organic contaminations into the water condition [14, 15, 16]. Consistently 230 000 t of PAHs achieved the marine condition and are pervasively disseminated around the world [17, 18]. The investigation of these compounds is of great scientific interest, since most of them demonstrate high cancer-causing and mutagenic activities [19, 20, 21, 22] and are also known to affect the skin as well as the immunological and hepatic systems [23].

Pyrolysis of fossil fuels leading to atmospheric deposition has been reported as the main source of anthropogenic PAHs in the environment [24, 25, 26, 27]. Several processes including gasification of coal for fuel and chemical production, coal coking for iron production process as well as underground coal for syngas and energy production are likely to produce coal tars which are likely to contribute to PAHs release in the environment when not properly controlled. The PAHs concentration originating from human activities is very high compared to the PAHs concentration due to natural processes [28]. In this study the potential release of PAHs from coal tar exposed to acid rain is investigated considering the impact of physico-chemical parameters.

2. Methodology

2.1. Chemicals

- Nitric acid (HNO_3) 55% purity
- Sulphuric acid (H_2SO_4) 98% purity

2.2. Sample collection

Coal tar generated from the coking process at Arcellor Mittal South Africa, Vanderbijlpark branch, was collected in 5 L container, transported in the laboratory and stored in a dark area at ambient room temperature (approximately 25℃).

2.3. Preparation of synthetic acid rain

Rainwater was prepared to simulate the acidic rain in the Mpumalanga area. The pH of the rainwater in Mpumalanga is below 4.3 due to the high release of Sulphur dioxide in the area. S. Rain was made by mixing 1 L of deionized water with 0.911 µL of H_2SO_4 and 2.86 µL of HNO_3 to achieve a pH of 4.3 according to our previous protocol [29].

2.4. Leaching experiment

Optimum solid liquid ratio

This study aimed to test the leachability of PAHs from coal tar into water sources. Before investigating other factors that can enhance the release of PAHs from coal tar, it is of importance to first determine how the leaching solution volume affects the process. An optimum ratio of coal tar to leaching lixiviant obtained was used for the rest of the experiments. To determine an optimum ratio, the ratio of coal tar to leaching solution was varied in this way: first, a ratio of 5/95 weight per cent (wt.%) was used, and then 10/90 wt.% and finally 15/85 wt.%. 5 g, 10 g and 15 g of tar were weighed and transferred into three different 250 mL sealable jar containing the corresponding volume of the prepared synthetic acid rain. The experiments were carried out in duplicate. All the samples were placed in the platform shaker for 12 hours. The shaker speed and temperature were set at 250 rpm and 55°C, respectively. The ratio that resulted in a higher amount of PAHs in the leachate was taken as the optimum ratio. For all the experiments, the maximum leaching time was set to 6 hours.

Fig.1 Mixture of coal and acid rain in batch system

Effect of time and temperature on coal tar leaching

A batch test utilizing AMD, S. AMD, S. Rain, River and pure water as leaching solutions was used in this study. Coal tar and leaching solution were added into a 250 mL sealed jar. 5 g of coal tar was weighed in a 250 mL sealable jar, and 95 mL of lixiviant was added into the jar. The sealed jars were placed in a platform shaker at 250 rpm. Jars in the shaker were allowed to shake for different times. The shaking times of jars were varied in this way: 30 minutes, 1 hour and half, 3 hours and 6 hours. The resultant leachate was collected at each time interval. This step was carried out while also changing the temperature of the Platform shaker. The experiment was run at 25°C, 35°C, 45°C and 55°C. Each experiment was carried out in duplicate. The control samples consisted of water as leaching solution. They were prepared by weighing 5 g of coal tar in a jar and adding 95 mL of pure distilled water. All the samples were kept in the fridge at a temperature of 0-6°C before analysis.

2.5. Data analysis

The collected leachate from both batch and column leaching was analyzed for PAH content at UIS organic laboratory using UISOL-T-020 method.

3. Results and discussion

Effect of time and temperature on the leaching of PAHs from coal tar

The leaching of coal tar in the prepared synthetic acid rain at various temperature and time resulted in the release of PAHs into solution (Fig 2). It was however observed that there was no defined trend of the release of PAHs over time and with the increase of temperature. Although the concentration of PAHs at 25℃ increased with time, the opposite was observed at 35℃; while at 45 and 55℃ there was inconsistency with regard to PAHs release over time. This could be due to the fact PAHs could bind on the side of the bottle which was cover with oily coal tar. With regard to the effect of temperature an increase of PAHs release with temperature was almost evident at 90, 180 and 360 min. Overall it could be observed that the highest concentration of PAHs was released after 30 min at 45℃.

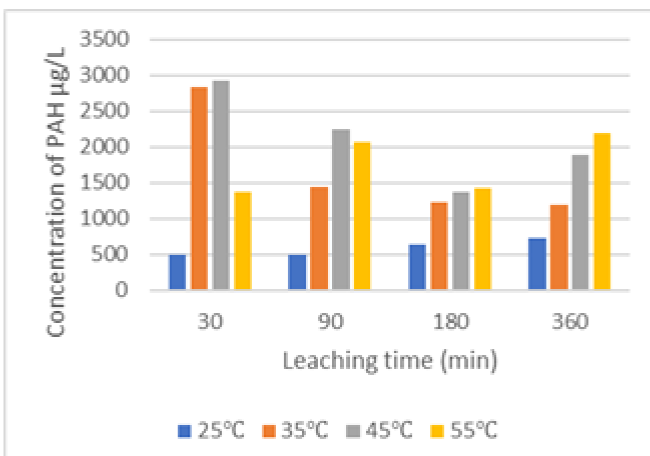

Fig. 2 Concentrations of PAHs released from coal tar under the effect of time and temperature.

Effect of temperature on the type of PAH released from coal tar

The toxicity of PAHs varying between PAHs, it was important to determine the type of PAHs released at each temperature. It was observed (Figure 3) that most of the PAHs were released at 25℃ and Dibenz(a, h) anthracene was exclusively released at this temperature. Benzo(g, h, i) perylene and indeno(123-cd) pyrene were both released only 25 and 35℃; it is likely that these three PAHs are thermolabile and therefore degrade easily at higher temperature. On the other hand, all the other PAHs considered in our analysis were released at all the four temperatures.

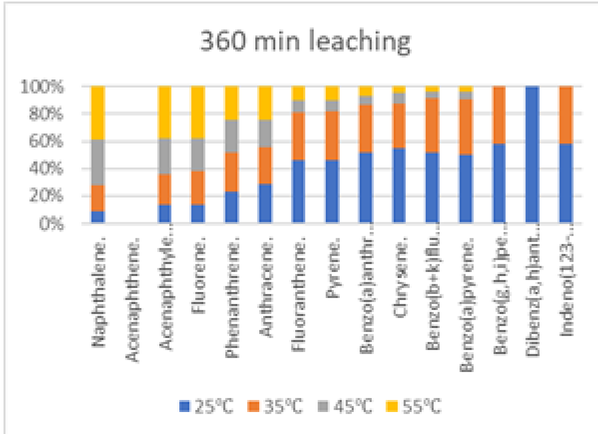

Fig. 3 Proportion of PAH released from coal tar at various temperatures

Effect of solid liquid ration on the release of PAHs from coal tar

Effect of solid to the liquid ratio on the mobility of PAHs is a vital parameter to assess, given the variation of such parameter in the environmental scenario. Figure 4 displays the leachability behaviour of PAHs when the solid to the liquid ratio is varied. From the observation on Figure 4, the leaching volume does influence the mobility of PAHs. The maximum leaching is achieved in the presence of a larger volume of liquid. This behaviour is observed with all the PAHs. This implies that in the environment higher dilution of the coal tar in the receiving water will promote the risk of contamination with PAHs. Similar results were found in the study done by [30]. Naphthalene is found to be easily released from the coal tar compared to other PAHs.

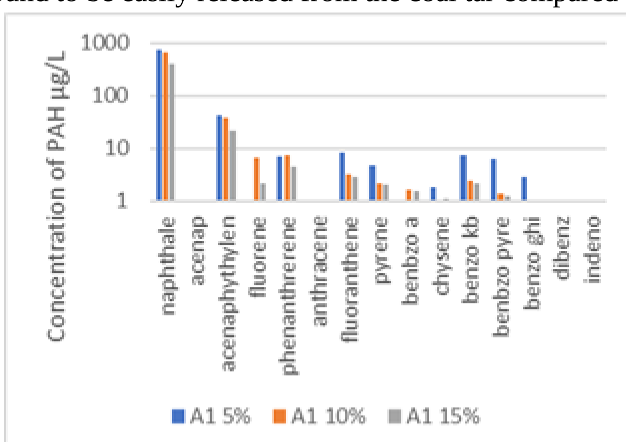

Fig 4 Concentration of PAHs released from coal tar at various solid liquid ratio

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4. Conclusion

The aim of this study was to assess the potential of PAHs release from coal tar in acid rain at varied temperature, time and solid liquid ratio. It was observed that the leaching time did not consistently affect the release of PAHs from coal tar, while the PAHs release increased with an increase of temperature; however, some of the PAHs where likely to be thermolabile as they were not identified in the leachates at higher temperature. The room temperature was observed to be most conducive for the leaching of PAHs. The effect of solid ratio was found to play a role in the release of PAHs from coal; in fact higher dilution of coal tar was conducive for PAHs release. Further study is required to better understand the leaching kinetic of PAHs from coal tar.

5. Acknowledgements

The authors acknowledge the financial support from the Water Research Commission. Project No: K5/2974//3.

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