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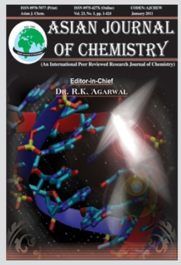

Analysis of Petroleum Leaching Behaviour from Cement-Solidified Oily Sludge

QI-PENG LIU[1] and YONG-LI YAN[2,*]

[1]Civil & Environment Engineering School, University of Science & Technology Beijing, Beijing 100083, P.R. China
[2]College of Chemistry & Chemical Engineering, Xi'an Shiyou University, Xi'an 710065, P.R. China

*Corresponding author: Tel: +86 29 88382706; E-mail: yylhill@163.com

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Stabilization/solidification is a process widely applied for the immobilization of inorganic constituents of hazardous wastes. But knowledge of the exact leaching behaviour of contaminants is limited. In this study, the effects of parameters including time, temperature and the cement/sludge ratio on the diffusion behaviour of petroleum after the solidification of oily sludge were investigated by using extraction and UV spectrophotometer. These experimental results indicated that the quantity of petroleum diffusion varies from 1.18 at 24 h to 3.48 mg/L at 72 h. The amplitude of petroleum diffusion within 72 h is the most distinct and thereafter, the variation becomes slower. The diffusion quantity enhances from 2.01-2.77 mg/L with the increment of temperature under the range of 298-308 K. The trend displays flat when the temperature is beyond 308 K. The amount of petroleum diffusion reduces from 3.82 to 2.80 mg/L with the increasing of the cement/sludge ratio under the range of 1.0-1.8. The diffusion quantity reaches 2.69 mg/L at 120 h after adding reinforcing agent of melamine resin, which is lower than without reinforcing agent. The diffusion quantity could arrive at 5 mg/L through 120 h, which meets the first order dispatching demands requested. These results could be used to assess the extent of leaching and understand the leaching mechanism.

Key Words: Leaching behaviour, Petroleum, Oily sludge, Solidification.

INTRODUCTION

Cement-based stabilization/solidification (S/S) is a widely applied and well-established technique for the immobilization of inorganic hazardous constituents. Many industrial wastes are well treated with this cost-effective technology. However, solidification technology has run into difficulties when trying to solidify organic wastes[1,2]. In a well solidified product, hazardous contaminants are chemically immobilized in the cement hydration products or physically entrapped by encapsulation. The resulting solidified specimen has improved structural integrity and physical characteristics.

The solidification technology is commonly used as the final treatment step for treating hazardous wastes before they are land disposed[3-6]. It involves adding one or more solidifying agents to a waste to convert it into a monolithic solid with structural integrity. Through solidification, the waste is chemically stabilized and physically modified into a low permeability solid matrix. The leachability of the waste is therefore reduced. The resulting product is also easier to handle and for transportation[6].

Among the various types of binders used for solidification, cement-based systems are the most commonly used systems because of their low cost and versatility[7-11]. Cement-based techniques use hydraulic cement as the major solidifying reagent. Solidification relies on the reaction of the cement with the aqueous phase of the waste or with the added water. Additives such as fly ash, blast furnace slag and sodium silicate are often used to replace part of the cement either to lower the cost or to improve product performance.

However, these progresses were designed to manage heavy metals in the waste, not organics. Organics are present to some degree in many predominantly inorganic waste materials and their leaching can lead to serious public health or environmental problems. Studies involving the use of solidification processes for organic wastes have reported little successes in immobilizing the organics constituents. Consequently, there is a great need to develop effective and economic solidification systems that can reliably treat hazardous wastes containing organics. Little information is available on the mechanisms involved with the release or leaching of organics from the stabilized waste[12-14]. A better understanding of the leaching behaviour of organics would enable us to predict their fate under "real-world" disposal conditions.

The present work focuses on the leaching behaviour of hydrocarbons from stabilized/solidified oily sludge. In this study, synthetic cement-based waste forms with oily sludge generated from the petroleum production and processing activities, were prepared and subjected to a toxicity characteristic leaching procedure (TCLP) to study the effect of parameters such as time, temperature, cement/sludge ratio and different additives on petroleum leaching.

EXPERIMENTAL

Sludge samples were obtained from Yanchang petroleum production plant. The additives used were cement, sand, clay, fly ash and lime and the accelerators used were sodium silicate, bentonite. UV-Vis spectrophotometry (Shimadzu, UV-160 μ) was used to determine the concentration of petroleum in the leachate.

Mixture preparation: The sludge was mixed with additives in different properties followed by the addition of water to form homogenous mixtures. The prepared mixtures were poured into standard moulds of size 111 × 111 × 111 mm. The solidified blocks were removed from moulds after 2 days and cured for 28 days under the wet condition to set.

Leaching tests of solidified samples: Leaching tests were carried out with each solidified sample by toxicity characteristic leaching procedure (TCLP). The TCLP leachate was analyzed for petroleum. The leaching experiments were performed by soaking the blocks with pure water. The jars were then capped and tumbled for 18 h. After 18 h, the leachate was decanted and measured for the contaminants of interest and also for calcium. Special care was taken (because of the volatile organics in the waste) to ensure that the TCLP jar was air tightly and had a zero headspace to prevent any volatilization during the tumbling operation.

RESULTS AND DISCUSSION

The leaching behaviour of petroleum from the solidification matrix appeared to depend on several factors. These include liquid/solid (L/S) ratio, curing period, concentration and presence of other organic constituents. These effects of parameters including time, temperature, cement/sludge ratio and additives are discussed in detail.

Effect of time: The leaching behaviour as a function of time was plotted in Fig. 1. The quantity of petroleum diffusion displays an upward trend gradually with the dissipation of extraction time. The concentration of petroleum leaching increases from 1.18 at 24 h to 3.48 mg/L at 72 h. The amplitude of petroleum diffusion within 72 h is the most distinct and thereafter, the variation becomes slower. This is mainly because the amount of leaching is the function of the dissipated time. The longer the dissipated time for samples, the more is the amount of petroleum diffusion into the aqueous solution. As the diffusion process carried out at a specific degree, the gradient concentration of petroleum between the samples and aqueous solution becomes progressively smaller and the diffusion rate of petroleum into the aqueous bulk reduces. Consequently, the variation of petroleum diffusion exhibits flatter than previously.

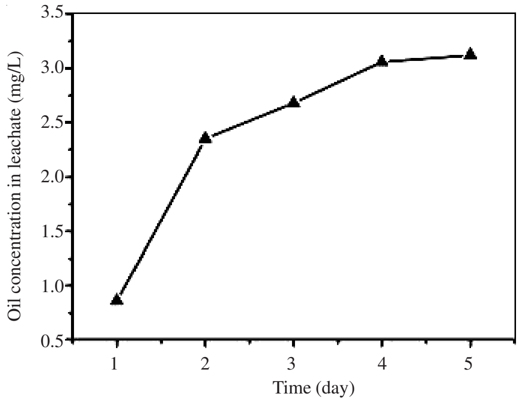

Fig. 1. Relationship of the concentration of petroleum in leachate and time

Effect of temperature: Fig. 2 showed the effect of temperature on leaching behaviour of petroleum under the condition of the range of temperature 298-313 K, cement/sludge ratio 1.6 and time 72 h. The higher the temperature, the larger is the quantity of petroleum diffusion into the aqueous solution. The diffusion quantity enhances from 2.01 to 2.77 mg/L with the increment of temperature under the range of 298-308 K. The trend displays flat when the temperature is beyond 308 K. These influences are mainly due to the interaction of molecules under the range of 298-308 K. As the temperature elevated, the movement of molecules becomes more rigorous. Consequently, the quantity of diffusion is to be quick. On the other hand, the viscosity of petroleum in the samples gets thinner as the result of elevated temperature. As the temperature lies between 308-313 K, the change amplitude of diffusion rate and viscosity is smaller than the range of 298-308 K, so that the amount of petroleum diffusion enhances more mild with temperature being beyond 308 K.

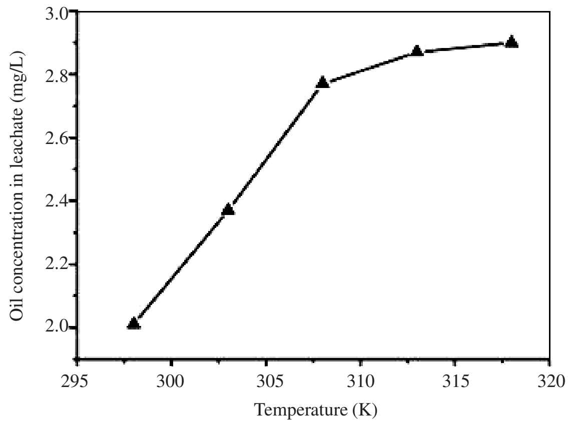

Fig. 2. Effect of temperature on leaching of petroleum

Effect of cement/sludge ratio: The results of cement/sludge ratio on the leaching behaviour are depicted in Fig. 3. As can be seen from Fig. 3, the larger the cement/sludge ratio, the smaller is the quantity of petroleum leaching. The quantity of petroleum diffusion reduced from 3.82 to 2.80 mg/L with the increasing of the cement/sludge ratio under the range of 1.0-1.8. A possibility for this result is that with the increase of content of cement, the microstructure of the hydration of cement becomes more tightly, which resulted in the permeability of petroleum in solidification blocks decreased dramatically.

Effect of additives: The effect of reinforcing agents on leaching behaviour under the temperature of 298 K, cement/sludge ratio of 1.80 and the time of 120 h are shown in Fig. 4. As can be seen from Fig. 4, the concentration of petroleum

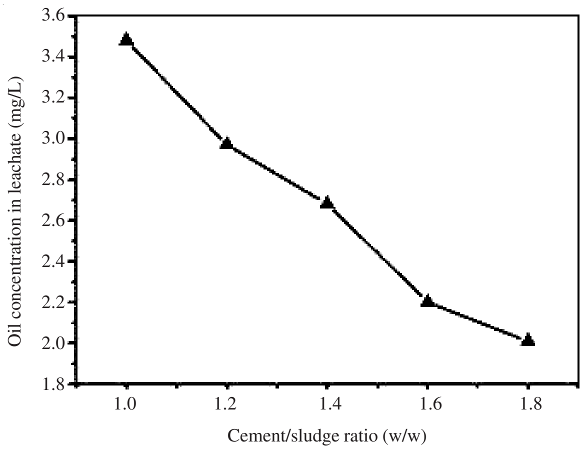

Fig. 3. Influence of cement/sludge ratio on leaching of petroleum

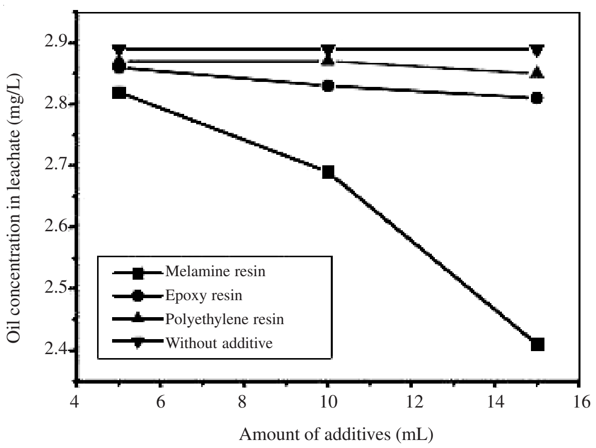

Fig. 4. Effect of the enforcing agents on petroleum leaching

leached reduced dramatically with the increment of amount of the melamine resin added and the variation amplitude of petroleum leaching for other reinforcing agents such as epoxy and polyethylene resins exhibits flatly. The diffusion quantity reached 2.69 mg/L at 120 h after adding melamine resin, which is lower than without reinforcing agent. The diffusion quantity could arrive 5 mg/L through 120 h, which meets the demands requested by national first order dispatching in China. The possible explanation could be that the high amount of reinforcing agents added could lead to the intensity of solidification of cement and reduce the performance of leaching of petroleum in solidified blocks, resulting in the lower leachability.

Conclusion

A leaching test was developed by modifying existing standard leaching tests and was applied to study the leaching behaviour of hydrocarbons from refinery oily sludge S/S with cement. On the basis of this test, the following conclusions were drawn. Macroscopic observation of the stabilized/solidified samples revealed macroencapsulation of the oily sludge into cavities, formed in the cement matrix. For higher cement addition, smaller and more abundant cavities were formed. The breakage of these cavities for the leaching test resulted in increased hydrocarbon leaching with cement addition. The quantity of petroleum diffusion varies from 1.18 at 24 h to 3.48 mg/L at 72 h. The amplitude of petroleum diffusion within 72 h is the most distinct and thereafter, the variation becomes slower. The diffusion quantity enhances from 2.01 to 2.77 mg/L with the increment of temperature under the range of 298-308 K. The trend displays flat when the temperature is beyond 308 K. The quantity of petroleum diffusion reduces from 3.82 to 2.80 mg/L with the increasing of the cement/sludge ratio under the range of 1.0-1.8. The diffusion quantity reaches 2.69 mg/L at 120 h after adding reinforcing agent of resins, which is lower than without reinforcing agent. The diffusion quantity could arrive at 5 mg/L through 120 h, which meets the demands requested by national first order dispatching in China.

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