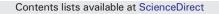
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Microscopic investigation on the adsorption of lubrication oil on microplastics

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ABSTRACT

The different size of microplastic in the environments may affect the fate and effects of traditional hydrophilic chemicals. The effect of reaction time, pH, salinity and concentration on the adsorption of lubrication oil on the 20–140 µm sized polystyrene (micro-PS) and 50 nm polyethylene (nano-PE) were presented. The adsorption kinetics and isotherms of nano-PE and micro-PS can be satisfactorily fitted by pseudo-second-order kinetic model and Langmuir, respectively. The adsorption of lubrication oil on nano-PE and micro-PS significantly increased with increasing concentration of salinity, indicating that the outer-sphere surface complexation dominated lubricating oil sorption on nano-PE and micro-PS. The maximum adsorption capacity of lubricating oil on nano-PE and micro-PS was 6.8 and 5.2 g/g at pH 5.0 and 293 K, respectively. These results are of crucial importance to the application of microplastics as the promising adsorbent in natural environments.

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1. Introductions

With the rapid development of industrial production and urbanization, the contamination of environmental impacts emerged in recent years [1–7]. Among the many classes of pollutants, oil is one of the most complicated organic contaminants with very low solubility to water due to the presence of hydrocarbons, fatty acids, soaps, lipids and waxes [8]. These pollutants have very low biodegradability, resulting in the influence of aquatic life via wastewaters. For example, thin layer of oil in aqueous solution decreased the penetration of light and the oxygen transfer between air and water. Therefore, the removal of oil from wastewaters is often challenging. Generally, there are three approaches, including physical (e.g., gravity separation, coagulation/ flocculation, flotation), chemical (e.g., electro-coagulation, bio-accumulation, chemical adsorption) and membranes methods [9-13]. Among these approaches, adsorption is a promising method to remove oils due to low cost, environmentally friendly, ease accessible and recovery [14–18]. Lubricating oils are often applied in the process to serve as interface between tools and work pieces. The rinsing, cooling and cleaning operation are carried away to the wastewaters of the facility [19].

Microplastics as a new pollutant pose a direct threat to aquatic organisms but are also used as a carrier for other organic pollutants [20– 26]. Lechner et al. reported that the load of plastic debris transported by the Danube River into the Black Sea was estimated to be 4.2 t per

* Corresponding author. *E-mail address:* hjq555918@sohu.com (J.-Q. Hu). day [27]. General Larger primary fragments of plastics were broken down into the smaller particles known as microplastics ranging in size from 5 mm to 1 µm. Nano- and micrometer-sized plastic particle are ingested by fish, worms and mussels, which could be caused the death of their living beings [28]. In order to transform waste into wealth, microplastic has been extensively regarded as a promising adsorbent in water purification. There have been a growing number of reports regarding the adsorption of organic pollutants on microplastics in recent years [22,24,26]. Huffer and Hofmann found that the high effective sorption of organic compounds (such as polyamide, polyethylene, polystyrene) on microplastics was attributed to the hydrophobic interactions [22]. However, there is little information available on the sorption of lubricating oils to microplastics.

In this study, $20-140 \ \mu m$ sized polystyrene (micro-PS) and 50 nm polyethylene (nano-PE) as two kinds of commonly found microplastics in the marine environment were used in the study. The objective of this study was to investigate the effect of water chemistry on lubrication oil adsorption to microplastics by batch techniques. This study provides the insight in the design of appropriate treatment for the removal of lubricating oils from aqueous solutions.

2. Materials and methods

2.1. Materials

The nano-PE and micro-PS as typical microplastics were purchased as powder from Goodfellow Cambridge Ltd. (Huntingdon, UK). The lubricating oil (>99% purity) was purchased from Sigma-Aldrich. The

Table 1

The selective properties of nano-PE and micro-PS.

Microplastics	Density ^a	T _G ^b	S_{BET}^{c}	D ^d
	(g/cm ³)	(C)	(m ² /g)	(µm)
Nano-PE	0.91	- 120	0.532	56.45
Micro-PS	1.06	100	0.294	169

^a Provided by the supplier.

^b Glass transition temperature, provided by the supplier.

^c Specific surface area, provided by the supplier.

^d Number-based diameter of sorbent particles in aqueous dispersion.

selective properties of nano-PE and micro-PS were summarized in Table 1. The density of nano-PE and micro-PS was 0.91 and 1.06 g/cm³, respectively. The specific surface area derived from BET methods of nano-PE and micro-PS was 0.532 and 0.294 m²/g, respectively. The average diameter of nano-PE (56.45 μ m) was significantly lower than that of micro-PS (169 μ m).

2.2. Experimental protocols

The triple adsorption experiments were carried out by using 10 mL glass beakers at a constant temperature (293 K) and an 48 h light-dark stirring conditions. Briefly, the lubrication oil was slowly added into 5.0 g/L micro-PS/nano-PE with 25 mg/L NaN₃ as biocide. Adsorption kinetics was set at 0.5, 1, 3, 6, 12, 24 and 48 h. The adsorption isotherms were spiked with working solutions in single-sorbate concentration that ranged over 1 or 2 orders of magnitude. The methanol content in aqueous solutions did not more lass 0.25% (v/v) to minimize co-solvent effects. All samples were reacted for 2 days under overhead shaker at 293 K. The adsorption process of lubricating oil on nano-PE and micro-PS was showed in Fig. 1. As illustrated in Fig. 1, the color was changed from yellow derived from lubricating into white when micro-PE and nano-PS were added into watch glass.

3. Results and discussions

3.1. Effect of reaction time

Fig. 2 shows the effect of reaction time on the adsorption of lubricating oil onto nano-PE and micro-PS. It is observed that the adsorption of lubricating oil onto nano-PE and micro-PS significantly enhanced with increasing reaction time starting from 10 min to 6 h, then remained the high-level adsorption at reaction time > 12 h, which were in a accordance with previous studies [29–32]. The data of adsorption kinetics were fitted by pseudo-first-order and pseudo-second-order kinetic models. The linear forms of pseudo-first-order and pseudo-secondorder kinetic models can be described by Eqs. (1) and (2):

$$\ln(q_e - q_t) = \ln q_e - k_f \times t \tag{1}$$

$$t/q_t = 1/(k_s \times q_e^2) + t/q_e \tag{2}$$

where k_f and k_s (g/(mg · h)) are the rate constants of pseudo-first and pseudo-second-order model, respectively. q_e and q_t (mg/g) are the adsorbed amount of lubricating oil at equilibrium and time t (h), respectively.

The fitted results and corresponding parameters were shown in Fig. 2 and Table 2, respectively. It is observed that the adsorption of lubricating oil on nano-PE and micro-PS can be satisfactorily fitted by pseudo-second-order kinetic model with high correlation coefficient ($R^2 > 0.9999$) compared to pseudo-first-order kinetic model ($R^2 < 0.97$), which was in accordance with the previous studies [33–38].

3.2. Effect of particle size

Fig. 2 also shows the effect of particle size on the adsorption of lubricating oil onto nano-PE and micro-PS by batch techniques. One can see that the adsorption of lubricating oil on nano-PE was significantly higher than that of lubricating oil adsorption on micro-PS. As shown in Table 1, the average diameter of nano-PE was significantly lower

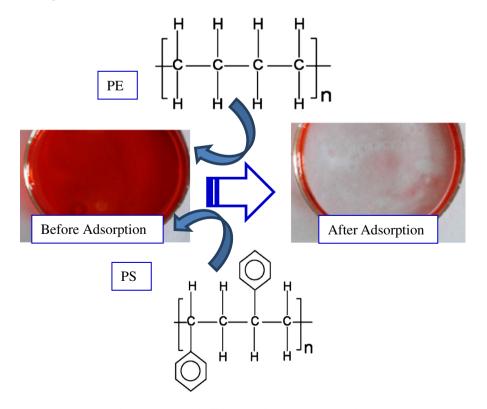


Fig. 1. Schematic graph of lubricating oil adsorption onto PE and PS.

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