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Effect of initial concentration on sulfamethoxazole (SMX) removal using zinc oxide adsorbent coating

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Abstract – Sulfamethoxazole (SMX) is an antibiotic that have been detected in significant quantities in sewage treatment plants in Malaysia and other countries. However, the conventional pharmaceutical treatment plants are not effective to reduce the concentrations of the compounds. Therefore, this study introduces an adsorbent coating formulated using zinc oxide and sepiolite as the adsorbent and additive, respectively and chitosan was used as the binder, which was then coated on the cotton cloth as the support via brushing technique. The batch adsorption studies have been done to study the effect of initial concentration on SMX removal. Result shows the percentage removal of SMX decreased from 94.7 to 65.3 %by increasing the initial concentration from 10 - 50 mg/L.

Keywords – Adsorbent Coating, Sulfamethoxazole (SMX), Adsorption, Antibiotics Removal, Zinc Oxide Coating

I. INTRODUCTION

The production of chemicals and pharmaceuticals products as well as their usage and application has impacted the environment and caused serious health effects for a long time. In the 1990s, the synthesis of 1 kg of an active pharmaceutical compound generated around 50–100 kg of waste [1]. During the second half of the twentieth century, several tremendous progresses was made in order to prevent the pollution of the environment and to reduce the impact of the pollution on health. A pharmaceutical product, such as a pill or a liquid is basically consisting of one or several active pharmaceutical ingredients (APIs), and additives, as well as inorganic salts or other organic chemicals which might be sugars, scents, pigments, and dyes. In fact, some medicines may also contain endocrine disrupting chemical excipients. There are three main routes of pharmaceutical waste to enter the environment which are effluents discharged by manufacturers, improper disposal of unused or expired medications, and excretion from the patients. Apart from manufacturers and healthcare institution, pharmaceutical waste is also derived from households because not all pharmaceuticals dispensed or purchased over the counter are consumed by the patients depends on the different situation such as completion of treatment, expired medication, changes in the dosage, unpleasant side effects, and forgetfulness [2,3]. Thus, most of the unused medicines will eventually end up as the household pharmaceutical waste and found to highly contribute to the environmental pollution [4]. Generally, household pharmaceutical waste cover all medications including over the counter drugs, illegal drugs and cosmetic products that have already expired, unsealed syrups or eye drops, tubes of cream, all bulk or loose tablets and capsules. Some medications and personal care products might contain hazardous chemicals. Abahussain et al. [5] explained that household pharmaceutical waste is typically found in the form of syrup, topical ointment, or cream, drops, solid oral dosage forms, injections, or syringes. Apart from excretion process from the human body [6], household pharmaceutical waste enters the environment through household trash, which is taken directly to the landfills [7]. Among all the conventional and advanced technology for pharmaceutical waste removal, adsorption proved more advantages over other methods because of its simple design that can involve low capital investment in terms of both initial cost and space required. For water and wastewater treatment, it is applicable for low adsorbate concentrations and suitable for batch or continuous process. As for the adsorbents, the ability to be reused and regenerated become an interesting study. Furthermore, adsorption is a low consuming energy process which can be very efficient because it may lead to the removal up to 90%, even with less operation condition. One of interesting study on adsorption is the adsorbent coating which refers to a thin layer of material applied onto a substrate or surface to enhance its adsorption performance. The coating is expected to selectively bind or capture specific substances from a solution onto its surface while effectively removing or reducing their concentration. It is designed to be applied on to various materials such as solid particles, fibers, membranes, or even the inner surfaces of reactors or columns. Different adsorbents have been studied for pharmaceutical waste removal yet metal oxides is still a growing interest. Metal oxides were applied for water purification since early before the 1950s which have been considered as a promising class of adsorbent materials owing to their excellent adsorption performance, inexpensive and sustainable-produced properties, and also ease in modification and regeneration. In many cases, the adsorption process by using metal oxides as adsorbents has the potential to produce high-quality treated effluent, like removing contaminants of low concentrations to trace level. This study is aimed at synthesizing and preparing adsorbent coating with functional adsorption sites to remove antibiotics. Among numerous antibiotics, sulfamethoxazole (SMX) which belongs to the family of sulfonamide antibiotic group, is one of the frequently detected antibiotics in aquatic environments with reported concentrations reaching high levels expressed in μg/L in surface water and mg/L in wastewater [8]. According to Segura et al. [9] review, sulfonamide antibiotics concentrations in surface waters were higher than those in wastewater effluent, thus indicating that they are not easily degraded or eliminated and tend to accumulate in the environment. Therefore, the elimination of antibiotics from water is urgently needed to mitigate underlying health and environmental risks.

This study will provide fundamental information of adsorption concept as different adsorbent form. This is forecasted to be very economical and easily accepted by industries since it’s able to face the typical complexity of industrial effluents. Hence, the proposed methodology will be beneficial for pharmaceutical waste later. The objective of this study is to study the effect of initial concentration on the removal of SMX.

II. MATERIALS AND METHOD

An adsorbent coating was synthesized by a mass ratio of 1:5:5 of chitosan, zinc oxide and sepiolite were used. 0.2 g chitosan was first diluted into 2 % of acetic acid and stirred approximately 8 hours till the chitosan completely dissolved without any agglomeration left. Then, 1 g of zinc oxide was added into the chitosan solution. Mixtures was further mixing for roughly 3 hours for complete dissolution of zinc oxide. Next, 1 g of sepiolite was added to the mixture of zinc oxide and chitosan solution. The mixing was continued for another 3 hours for better blending of all materials. The final solution resulted in brown-greyish colour, to be coated as in flat sheet layer.

Then, brush-coating technique was used to coat an adsorbent solution onto a cotton cloth. Firstly, a clean cotton cloth was cut at 30 cm length and 5 cm width, and at a center of cotton cloth, measured at 20 cm length and marked as reference. The cotton cloth was then clipped using paper clipper tightly at left and right side of a coating stand. The readily prepared adsorbent coating solution was coated manually onto the cotton cloth using paint brush.

For batch equilibrium studies, the adsorption experiment was conducted in batch mode using 250 mL flat bottom beakers with 200 mL initial volume of SMX solution. The experiment was conducted using a hot plate stirrer and the stirring rate was controlled at 300 rpm. All the experiments were performed at 27 ± 2 °C except mentioned earlier. The adsorbent coating strip was properly surrounded in the interior part of the glass beaker. Cloth clips were used to prevent any movement of strips during the stirring. The SMX solution was stirred at a predetermined time interval of 15 minutes, and 2 mL of SMX concentration was drawn out to observe the changes in SMX concentration using UV-Vis spectrophotometer at λ_{max} = 286 nm The equilibrium was determined when the SMX concentration is not showing any difference in three consecutive measures. The adsorbent coating strip was then withdrawn from the water solution without post-treatment of filtration or centrifugation processes. Fig. 1 shows the SMX structure used in this study.

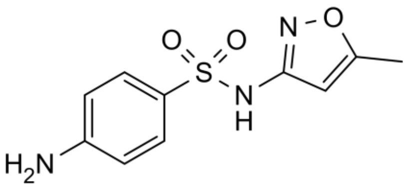

Fig 1 Sulfamethoxazole (SMX) structure

III. RESULTS AND DISCUSSIONS

The effectiveness of the prepared zinc oxide adsorbent coating in the adsorption of SMX were evaluated as a function of initial concentration and contact time. The effect of initial concentration on the removal efficiency (%) and adsorption capacities were studied by varying the initial concentrations at 10, 20, 30, 40 and 50 mg/L. Other reaction variables such as pH, temperature and dosage of adsorbent were fixed at its original and atmospheric condition. The experimental data were measured until a full equilibrium was attained. The profiles for zinc oxide sorption are presented in Fig. 2 shows by removal efficiency (%), and Fig. 3 is referring to adsorption capacity (mg/g) of SMX.

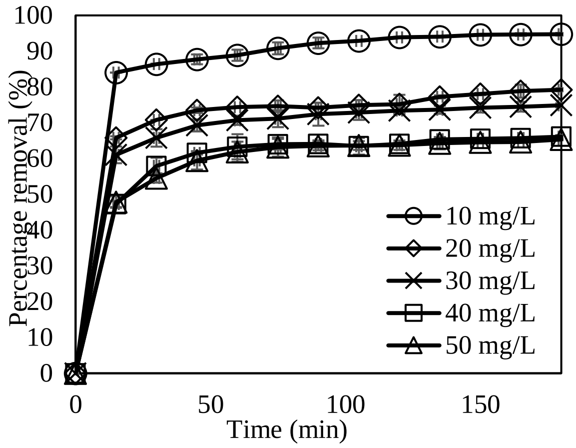

Fig. 2 Efficiency removal at different initial SMX concentration

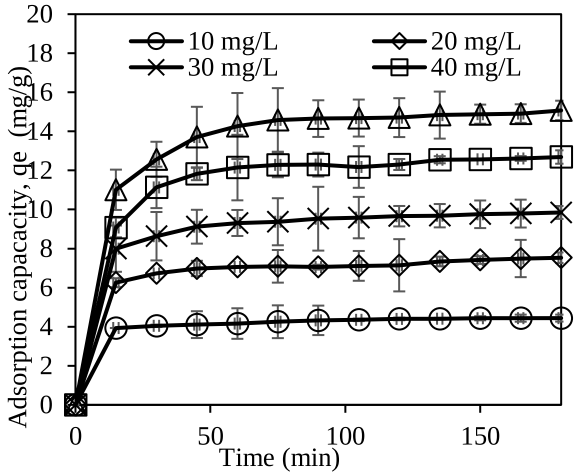

Fig. 3 Adsorption capacity at different initial SMX concentration

The experimental result depicted that increasing contact time and SMX concentration led to a rapid increment in the adsorption capacities of zinc oxide adsorbent coating. Variation of initial concentration from 10 to 50 mg/L, gave a corresponding increase in adsorption capacities which were from 4.44 to 15.07 mg/g. Similar trends reported by Dutta et al. [10], which adsorbate uptake increased with the increasing of initial concentration. The increased in adsorbate's initial concentration may lead to an increase in the driving force at the solid-liquid interface and also to an enhancement in mass transfer rate [11]. In term of removal efficiency, the percentage removal of SMX decreased from 94.7 to 65.3 % as initial concentration increase from 10 to 50 mg/L. Elimination of SMX for the first 15 minutes at every initial concentration (10 mg/L - 50 mg/L) of adsorption process was found faster throughout the experiment. This is due to competition between the particle adsorbed by adsorbent via pore filling and surface chemistry interaction between the adsorbent coating and SMX through diffusion forces. This in parallel with the finding observed by Zbair et. al., [12], the adsorption of SMX is divided by two parts, the first part is the diffusion of SMX molecule through external surface interaction (surface chemistry interaction) and the second part is an intraparticle diffusion where SMX will transport into pores to achieve equilibrium. This is because the amount of adsorbate adsorbed is directly proportional to the adsorption capacity. The effectiveness of zinc oxide on the adsorbent coating surface makes the SMX molecules probably compete each other to fulfil the vacant sites of adsorbent coating. Thereafter, the adsorption process become slower, and SMX removal continued at slower rate until the rate of adsorption reached a constant value, starting at first 1 hour up to 3 hours, at every concentration studied. This result could be caused by increasing accumulation of SMX, depletion, and saturation of available adsorption sites of the adsorbent wherein SMX molecules were no longer adsorbed on the adsorbent coating surfaces [14]. The finding was consistent with the study by [15, 16]. They mentioned that at the commencement of the adsorption process, faster decrease in the SMX molecules from the solution can be detected.

IV. CONCLUSION

Zinc oxide adsorbent coating is an interesting new adsorption method for pharmaceutical waste specifically SMX as one of the most occurrence antibiotics. The percentage removal of SMX decreased as the initial concentration increased due to the adsorbent saturation on the adsorption sites of the coating.

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