

Petroleum Science and Technology



ISSN: 1091-6466 (Print) 1532-2459 (Online) Journal homepage: http://www.tandfonline.com/loi/lpet20

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To cite this article: Anh Tuan Hoang, Van Vang Le, Abdel Rahman M.Said Al-Tawaha, Duong Nam Nguyen, Abdel Razzaq M.Said Al-Tawaha, Muhamad Mat Noor & Van Viet Pham (2018): An absorption capacity investigation of new absorbent based on polyurethane foams and rice straw for oil spill cleanup, Petroleum Science and Technology, DOI: 10.1080/10916466.2018.1425722

To link to this article: https://doi.org/10.1080/10916466.2018.1425722

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An absorption capacity investigation of new absorbent based on polyurethane foams and rice straw for oil spill cleanup

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ABSTRACT

Spilled oil has been considering as extremely serious disaster in the maritime field and oil exploration, and its effects lasted for decades, even hundreds of years. Oil spill treatment and recovery were the difficult and complicated issues due to the conditions of nature environment. In this work, rice strawagricultural residue in Vietnam – with 205 k g/m³ of low density was filled in porous polyurethan matrix in fabrication process of absorbent. The experimental results about oil absorption capacity for this new absorbent material showed that, the absorbed oil mass in case of filling 25% of rice straw mass with 0.5 mm of size was highest, equal to 12.012 g oil/ g absorbent material after 120 minutes of treatment. The result of oil absorption capacity was around 3–4 times higher than that of material fabricated by pristine polyurethan or xenlulozo/lignocellulosic. Besides, fabrication process and SEM analysis were also experimentally carried out in this work.

KEYWORDS

rice straw; polyurethane; absorption capacity; oil spill; absorbent

1. Introducton

Oil spills from any reasons are considered as "serious environment disasters" that cannot predict its negative impacts. As reported, there were many oil spill incidents because of oil rig such as Gulf of Mexico blew up on 20th April in 2010 (Welch et al. 2010); the Torrey Canyon in 1967 (Abdul, Abdulrauf, and Hossain 2012); the Amoco Cadiz in 1978; the shipwreck of Erika oil tanker in France, 1999; the Prestige in Spain, 2002 (Robertson et al. 2010); and maritime accidents related to soil shipwreck such as the ABT Summer ship in Angola, 1991; Castillo de Bellver ship at Saldanha Bay, South Africa in 1983; Amoco Cadiz ship in 1978; Odyssey ship in 1988 (Kapoor et al. 1994). Products originated from marine oil were complex hydrocarbon mixtures with very different molecular structure and physic-chemical characteristics that result in changing quickly while spilled and released into the seawater, and marine environment (Amin et al. 2015). In combination with wind, wave, condition weather, and solar radiation, the oil spills can be disintegrated or in-water emulsified, spread, evaporated, photo-oxidized, sunk, resurfaced, a weathered-oil in new formation of tar-ball or small lumps, thin sheens, and biodegradation to form or convert into different component enhanced more and more by wave (Schoenbaum 2012; Hussein et al. 2009). Therefore, it cannot help overemphasizing to the oil spill impact on the marine ecosystem such

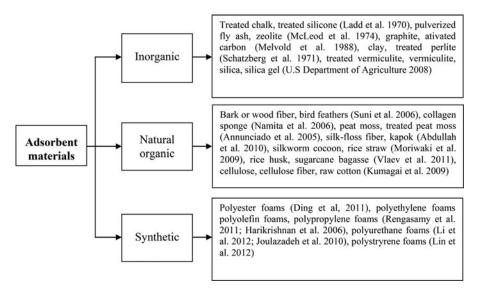


Figure 1. Classification of absorbent materials.

as offshore and coastal waters, plankton, fish, seabirds (Lucas 2006; Webler et al. 2010), marine mammals and reptiles (Annuciado 2005; Roger et al. 2015), shallow inshore waters, seagrass, corals, shorelines (Aisien, Hymore, and Ebewele 2006) rocky and sandy shores (Aguilera et al. 2010), soft sediment shores, salt marshes, mangroves (Burton et al. 2010) ... and so on. In among of physical methods for oil spill recovery, adsorption method was usually used for recovering oil spill by adsorbent materials with high capacity after using skimmers (Fingas 2012a; OSS 2010). The oil adsorption capacity based on adsorbent materials, which were divided into three types such as organic, inorganic, and synthetic materials, was shown in Figure 1.

Some physical methods from Figure 1 showed that, non-toxic, decontamination capability, complete oil recovery possibility from surface, well-managed, less harmful, low cost, and high capacity characteristics were the highlights of adsorbent material (Amin et al. 2015; Venkates et al. 2006). Therefore, detection of new adsorption materials with not only biodegradable, economic, but also hydrophobic and oliophilic characteristics to enhance and improve the oil adsorption and recovery capability plays an extremely important part in oil spill treatment.

Polyurethane (PU) was considered as a natural material with special features and properties such as high porosity, open-cell, low density, and industrial production (Javier et al. 2016; Wu et al. 2012). PU tended to show its higher oil sorption with low viscosity of oils and derivatives (Shi et al. 2014; Keshavarz et al. 2015). With more viscous oils and derivatives, the pores in PU were jammed and obstructed, they resulted in deceasing the oil sorption capacity. Besides, rice straw was an endless biomass resource in Vietnam, it was used to adsorb and recover oil spill and oil slick (Hassanein et al. 2014; Phan et al. 2017). In Vietnam, rice straw is usually used as combustive material for everyday cooking in the rural, material for planting mushroom, compost, food for buffalo and cow. The firing or elimination of the huge rest to the surrounding environment caused the serious pollution in the water and air environment (Phan et al. 2017). Above analysis and review resulted in the consideration and suggestion the idea in mind about fabrication a new adsorbent with high efficiency in oil absorption based on PU foams and natural residue-rice straw. In this reported work, the combination between PU foam with high oil absorption capacity (OAC) and nature rice straw that appeared as the residue in Vietnamese agriculture production to fabricate a new adsorbent with high efficiency was considered as a meaningful one. The PU-rice straw sorbent material might achieve and get several advantages such as capable of recycling, reduction in environment pollution, safety, improving the agricultural development, and low cost.

2. Materials and methods

2.1. Sorbent

Straw from rice production originated from Oryza sativa TBR225 grown in Thai Binh province, Vietnam was used in this experiment study to fabricate the PU-based sorbent material. The rice straw was cut by the cutter, and was screened by the sieves to get as-desired length. A part of rice straw was used to replace the PU to fabricate the absorbent. PU foams made in this work were based on component A type, and component B type bought from China. The component A, B were mixed with the proper weight ratio, and rice straw were added with the different weight ratio to aim to determine and evaluate the effects of these compositions on the capacity of fabricated absorbent.

2.2. Oils

The oil in this work included diesel oil (DO) has been using as main fuel in marine engines. The viscosity and density of DO at room temperature was 4.5 mm²/s, 860 k g/m³, respectively.

2.3. Material characterization

TAPPI methods were the standard for characterizing the chemical compositions of rice straw. The JSM-7000F, JEOL, Japan machine was use to perform scanning electron microscopy (SEM).

2.4. Sorption capacity test

ASTMF726 99 (Standard Test Method for Sorbent Performance of Adsorbents) was a standard aiming at developing the method for measurement and determination of OAC of each sorbent (Li et al. 2012; Annunciado et al. 2005). In this experiment setup shown in Figure 2, a 50 ml of DO was poured into a beaker containing 100 ml of water. This amount of oil was to ensure the thickness of oil film bigger than 5 mm and plenty oil remaining after carrying out the sorption experiments. The adsorbent material was compulsory for weighting and the value was recorded. Thereafter, a piece of fabricated sorbent material was immersed gently into the oil at room condition. Generally, after 15, 30, 60, 90, and 120 min \pm 15 s of immersion, the absorbent material was removed and treated to drain for 5 min \pm 5 s under vacuum condition before weighed. However, because of water incorporation into the sorbent material, the determination of water uptake into the sorbent material needed to conduct with further experiments. The water uptake (G_s) on a weight was calculated and determined as following:

Water uptake,
$$G_{wu}(g/g) = \frac{G_{ws} - G_o}{G_o}$$

where: G_{ws} , G_o was respectively the weight of sorbent samples after water sorption and the weight of initial dry sorbent samples. All experimental tests were performed at room condition with the mean of the three times of determination and the mean value errors must be less than 15%. Besides, the mass of adsorbed oil was consider as a primary and important factor to evaluate the capacity of sorption material and the sorption coefficient that was calculated versus the unit of g adsorbed oil per g dry sorbent material, and following equation:

Oil sorption coefficient,
$$C_s\left(g/g\right) = \frac{G_{os} - G_o}{G_o - G_{wu}}$$

Where: G_{os} was the weight of sorbent samples after oil sorption.

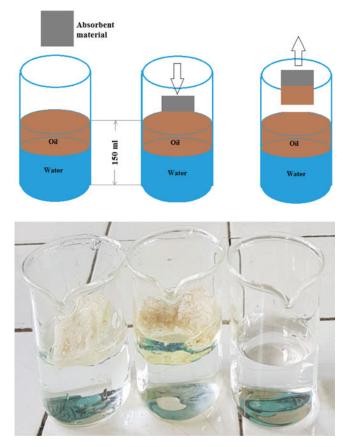


Figure 2. Experimental setup of sorption test at room condition.

3. Results and discussion

3.1. Sorbent material synthetic

Rice straw used for implementing into the PU cell structure based on TAPPI methods with 56–60% of cellulose, 18–24% of hemicellulose, 14–17% of lignin, 4–8% of others was grown in Thai Binh province, North of Vietnam. The open-cell structure of PU foam increased the OAC. However, as saturated or supersaturated state occurred, PU might be easy to sink into the seawater. Thus, the ability of retaking PU under seawater was extremely difficult. Floating improvement in the saturated or supersaturated state of each sorbent material by adding and filling the porous materials with low density such as rice straw, which has been considered as the waste in agricultural production process, was an acceptable, applicable, practical, eco-environmental, useful ponder and way of calculation. The pre-treatment and fabrication process of sorbent material from PU and rice straw was introduced in Figure 3.

The fabrication process of PU-rice straw sorbent material was denoted and explained as two-phase one that fixed pre-treated rice straw with the 5%, 15% and 25% respectively of mass and PU-component A was mixed by a rotor at around 3000 ± 30 rpm within 30s in the first period. In the second period, PU-component A and rice straw were strongly stirred in some seconds at room condition after adding PU-component B. New material was cut into same dimension for each experiment after completely soliditying reaction. In this study, rice straw length after treating to carry out mixing with PU-component A was 0.5 mm and 3 mm. As-fabricated sorbent material was shown in Figure 4.

The SEM images were used to characterize the surface properties of sorbent. As seen from the Figure 5b and Figure 5c, the roughness and porosity of sorbent material several pores and fissures with was obviously observed. The open-cell structure was showed on the sorbent material from rice straw

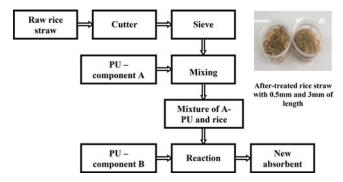


Figure 3. Fabrication process of PU-rice straw sorbent material.

based on PU in comparison with SEM of rice straw in Figure 5a. The surface characteristics effect dramatically on OAC (Pinto et al. 2016). Special characterization due to a good combination between porous PU foam and fibrous rice straw results in a hope about using new sorbent material with high potential for recovering oil spill to protect environment.

3.2. Effect of PU component on OAC

In order to select the appropriate ratio between the two components A and B of PU, study on the impact of A:B ration has been conducted with the following parameters: 25% of rice straw ratio; 0.5 mm of rice straw length; 100:100, 100:60, 150:60, respectively, of the ratio between two components A and B.

Through the experiments to determine the OAC at 120 minutes, the ratio between A and B was extremely important, and it affected the properties of the adsorbent. PU was made by reacting of an

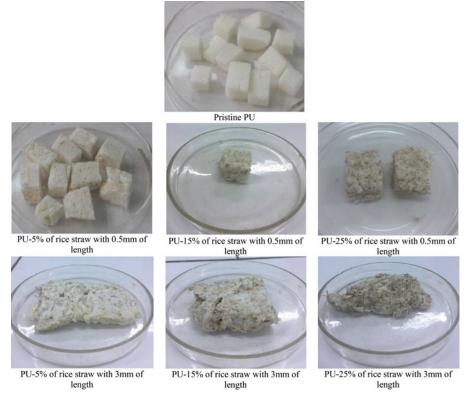


Figure 4. Sorbent material based on different mass and length of rice straw.

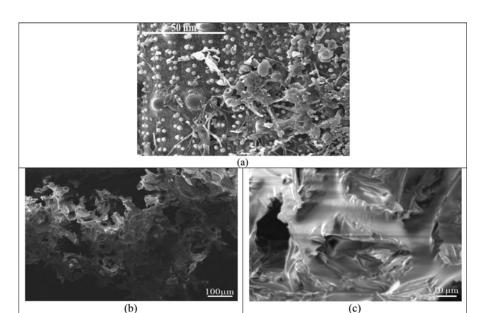


Figure 5. SEM of rice straw with $50\mu m$ of scale (a) and sorbent material with $100\mu m$ of scale (b) and $10\mu m$ of scale (c).

isocyanate containing two or several isocyanates A groups (R (N=C=O)_n) with a polyol B containing an average of two or several hydroxyl groups per molecule (R '-(OH)_n in the presence of a catalyst or activated by ultraviolet light. Thus, PU properties were greatly affected by the ratio of isocyanates A, and polyols B. The OAC depended on components A and B was shown in Figure 6.

Figure 6 showed that, as using 100:100 ratio, OAC of the adsorbents was very low, around 1.82 g/g. However, with the same amount of fillers and at the same experimental time, OAC at 150:60 ratio was around 3 times higher compared to 100:100 ratio. OAC at 100:60 ratio was the highest and many times higher than those of 150:60 ratio, and 100:100 ratio. This might be explained that, at a ratio of 100:100, the amount of component A was retained but the amount of component B was increased, the curing agent increased, and the adsorbent material was tougher than that of ratio of 100:60. The capillary pores were also small and less, it means that the lower porosity makes a difficulty of oil absorption into the material. At the ratio of 150:60, an increase in component A and remaining of component B result in brittle, fragile and humus material. At the ratio of 100:60, the porosity of the adsorbent material was not

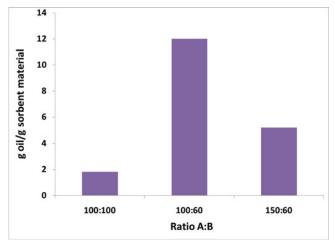


Figure 6. Effect of polymer composition on oil adsorption.



Table 1. Oil absorption capacity with 0.5 mm of rice straw size.

Time (min)	Oil absorption capacity (g oil/g absorbent)				
	No rice straw	5% of rice straw	15% of rice straw	25% of rice straw	
15	2.472	3.107	5.516	6.18	
30	2.557	3.473	6.562	8.918	
60	2.956	3.664	7.262	11.156	
90	3.289	4.152	8.312	11.701	
120	3.693	4.227	8.91	12.012	

compromised, adsorbent material was not humus, brittle with high adsorption capacity. Thus, the ratio of component A and component B for fabricating PU was 100:60.

3.3. Effect of rice straw ratio and size on OAC

The combination by utilizing the two types of material such as adsorbent and natural materials created a new material with both economic and technical advantages. The effect of the lignocellulosic component ratio from rice straw to the oil adsorption efficiency with the purpose of minimizing the cost of the adsorbent material (maximizing the amount of rice straw) without affecting, even the increase in its properties and the oil cleavage out of the adsorbent material was investigated. The experiment has been conducted with 5%, 15%, 25% of rice straw ratio; 0.5 mm and 3 mm of rice straw length, respectively. The results of the experimental process are shown in Table 1 and Table 2.

The results in Table 1 and Table 2 showed that, the amount of filled rice straw greatly affected the OAC of absorbent material. With different sizes of filled rice straw, the OAC of the sorbent increased with the increase in filled rice straw ratio. Specifically, with all two rice straw sizes at 0.5 mm, and 3 mm, the OAC was 12.012, and 6.117 g/g, respectively, in comparison with 3.693 g/g of pristine PU. This result might be due to the greater the amount of fillers were, the greater the amount of capillary or the space between adsorbents were, which increases the surface area of the sorbent material which gave the oil more osmotic potential inside the sorbent material. In addition, lignocellulosic fiber itself was also a porous material (after treatment) with several capillaries resulted in high OAC. As combined with PU, a synthetic material was fabricated with high applicability. However, in this study, only lower than 30% of rice straw ratio was considered, as the higher filled rice straw ratio, the fabricated material was less hard, less adhesive, and crumbly. Hence, the optimum fill ratio of rice straw was 25% for the fabrication process of adsorbent. The relationship between OAC and oil absorption time with different rice straw size in case of the same fill ratio was plotted in Figure 7.

From the results shown in Figure 6, the size of the rice straw used for the sorbent fabrication also toke an important influence to the OAC of the sorbent material. In particular, as the rice straw size increased, the OAC decreased. Under experimental conditions, as using 0.5 mm of rice straw, the OAC after 120 minutes was 12.012 g/g, with 3 mm of rice straw was 6,117 g/g. The maximum of OAC achieved with to 0.5 mm of rice straw size. This result might be due to the decreasing of filled rice straw size, the surface area of rice straw increased, it resulted in an increase in the surface area of the adsorbent. Moreover, with the same filled ratio 25%, small rice straw size made an increase in the number of pieces that

Table 2. Oil absorption capacity with 3 mm of rice straw size.

Time (min)	Oil absorption capacity (g oil/g absorbent)			
	No rice straw	5% of rice straw	15% of rice straw	25% of rice straw
15	2.472	2.711	2.904	4.052
30	2.557	2.822	3.110	4.498
60	2.956	3.145	3.682	5.277
90	3.289	3.476	4.056	5.683
120	3.693	3.823	4.978	6.117

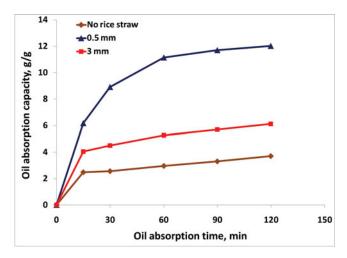


Figure 7. The relationship between OAC and oil absorption time.

led to increase in the amount of capillary in the adsorption material. As known, the OAC depended much on the surface area and capillary volume of the sorbent material. Therefore, using small rice straw size gave a higher oil adsorbent capacity. Besides, the OAC also depended on the time sorption where this relationship was considered as proportional but it seemed not linear. In comparison with the whole experiment time, the OAC increased dramatically in the first 60 minutes, the mass of absorbed oil in this period was 92.87% for sample with 0.5 mm of filled rice straw and 82.26% for sample with 3 mm of filled rice straw. However, the mass of absorbed oil in next period (from 60-120 minutes) increased unsignificantly. The ratio of absorbed oil mass and time was only 0.014 g oil/min compared with 0.088 g oil/min of the first 60 minutes with 3 mm of filled rice straw, and 0.0143 g oil/min compared with 0.186 g oil/min of the first 60 minutes with 0.5 mm of filled rice straw. The saturation of absorbed oil into the porous space of sorbent material occurred. This result was the same as other past study (Annunciado et al. 2005). However, the new as-fabricated sorbent material in this study offered the OAC higher than that of other absorbent material from Azolla plant (Amin et al. 2015), carbonized pith bagasse (Hussein et al. 2009), recycled rubber (Aisien, Hymore, and Ebewele 2006), other polymer foams (Javier et al. 2016) or rice husks ash (Vlaev et al. 2011).

4. Conclusions

A utilization of residue from agricultural production in Vietnam for fabricating a new sorbent material as combined with porous polyurethane foam was considered as useful, efficient work in oil spill treatment and recovery. In this study, new sorbent material showed its highlights such as high oil absorption capacity as filled with 25% of rice straw mass and 0.5 mm of rice straw size. The mass of absorbed oil increased dramatically in the first 60 minutes, especially the first 15 minutes. After 120 minutes of experiment, the maximum of absorbed oil mass of new sorbent material was 12.012 g/g in comparison with 3.693 g/g of absorbed oil mass for pristine polyurethane. Thus, as-fabricated absorbent material in this study not only contributed to verifying the materials for environmental protection from oil spill or oil slick, but also improving the eco-technical, utilizing the rice straw waste from agriculture, reducing the production cost.

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