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Regeneration of granular activated carbon loaded with toluene – Comparison of microwave and conductive heating at the same active powers

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ABSTRACT

This paper presents a comparison of the microwave and conductive heating regeneration of granular activated carbon loaded with toluene at the same active powers. The purpose of this research was to compare desorption efficiencies and local temperature profiles for the both regeneration methods. Mass balance in the adsorbent bed was carried out by automatic sampling of the outlet gas for the GC analysis instead of weighting the adsorbent bed itself. Five experimental series were performed at constant active power of 12 W and at changing active power in the two-stage program (24 W + 12 W). It was found that the mass profiles of desorbed toluene versus time were practically overlapped for the two regeneration methods when the same active power was applied and the non-insulated vessels were used. Moreover, another comparison demonstrated that proper thermal insulation can be a decisive factor when choosing a particular regeneration method in terms of its economic viability. A comparison of the textural properties of the fresh and regenerated (after 10 successive adsorption/desorption cycles) granular activated carbon (GAC) revealed only slight differences between them. Therefore, it was concluded that the adsorption capacity of GAC is only slightly affected by the both regeneration methods.

1. Introduction

Toluene is a common representative of volatile organic compounds (VOCs). It is used as an additive in gasoline mixtures, in benzene production, and as a solvent (i.e. paints, inks, and cleaners). In addition, it is utilized in the production of nylon, plastics, and polyurethanes [1]. The negative environmental effects of emissions of VOC are widely known. Therefore, VOC emissions have to be controlled and minimized. For instance, the problem is regulated in the European Union by the Paints Directive [2] which amended the original VOC Solvent Emissions Directive [3], and by the Industrial Emissions Directive [4].

There are several techniques for recovery of VOCs such as: condensation, absorption, adsorption and membrane separation. Basically, adsorption covers the range of VOC concentration from 20 ppm to one-fourth of the Lower Explosive Limit (LEL). However, it is economically feasible rather at the lower limit of this range. Incinerators, membranes, and condensers are better choice when the VOC concentration are at the other limit of this range [5].

The spent adsorbent needs to be regenerated to be used in the next adsorption cycle. This step is usually performed using pressure swing adsorption or temperature swing adsorption. However, the former process is more common. The known limitations of temperature swing adsorption are long regeneration time and dilution of desorbed compound in a stream of the purge gas which means low condensation temperature in the case of solvent's recovery. These drawbacks can be diminished by using microwave regeneration. Contrary to conventional regeneration in which heat needs to be delivered to the adsorbent bed from outside, heat is produced directly inside the adsorbent in microwave regeneration. In principle, the direct interaction of microwaves with the adsorbent and selective interaction of microwaves with the adsorbate can enable a faster process with a lower purge gas flow rate and lower process temperature that can be translated into energy savings.

The use of microwave heating for regeneration of adsorbents has been investigated since 1980s [6–8]. There are several review papers devoted to this topic [9–11]. Over the past decades various types of adsorbents have been examined, including silica gel [12–14], zeolites [12,15–22] and activated carbon [14,19,20,23–38].

While most articles concern microwave regeneration itself, only some of them compare microwave and conventional regeneration. In these cases, the problem of energy consumption is usually addressed [24,34,35,38].

Mao et al. [34] investigated the regeneration of spent activated carbons prepared from pine and wheat straw. These adsorbents were saturated with toluene and acetone. The effect of flow rate, microwave power, and humidity of purge gas on microwave regeneration of these adsorbents was investigated. Three regeneration methods were compared in terms of the electric energy consumption: constant power

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(CPMH) and constant temperature (CTMH) microwave heating, and conductive heating (CH). For toluene, they found that CPMH and CTMH required 13.5 and 27 kJ/g of electric energy to achieve 99% regeneration ratio, respectively. The CH required 60.8 kJ/g. They concluded that the two microwave methods are more efficient than the CH. The regenerations were carried out at different powers. While CPMH were conducted at constant microwave power of 600 W, variable powers were applied for CTMH and CH. For CTMH, manual power control was employed. For the CH, power consumption increased from 0 to 90 W and then stabilized at around 45 W.

Fayaz et al. [35] compared microwave heating and conductive heating for the regeneration of beaded activated carbon loaded with *n*-dodecane. The measurements conducted for different times at constant applied power of 180 W showed that the energy required to attain equal desorption efficiencies was lower for microwave heating in every case. For instance, the total energy needed for complete regeneration with microwave and conductive heating was 56 and 972 kJ, and the corresponding regeneration time was 5 and 90 min, respectively. For the lowest of tested powers (105 W for microwave heating and 100 W for conductive heating), assuming total energies as before (56 and 972 kJ), the desorption efficiency was 100% and ~65% for microwave and conductive heating, respectively.

Ania et al. [24,38] investigated regeneration of activated carbon loaded with phenol. Two methods were compared – microwave and conventional. Both regenerations were conducted at the same temperature of 1123 K. The temperature was reached after 4–6 and 10–13 min for microwave heating and conventional heating, respectively. It was concluded that the main benefits of microwave heating compared to conventional heating were the relatively short regeneration time and lower consumption of gas and energy. It was also reported that microwave heating was initially very fast and it was slower for the saturated samples.

Successive adsorption/desorption cycles may change the internal structure of activated carbon due to a partial collapse of the porous structure (structural annealing) or formation of coke inside the pores decreasing the active surface and volume of the porous structure [24,38]. This in turn manifests itself by changing the adsorption capacity. Surprisingly, previous comparative studies showed that microwave regeneration maintained the original adsorption capacity better than conventional regeneration [24,38]. This seems likely that the opposite temperature gradient due to microwave heating enhance diffusion of molecules from the inside to the outside of an adsorbent. Besides, there are several papers showing that the adsorption capacity were unaltered [19,30,33,35,36,39] or higher [23,25,26] than the original adsorption capacity of activated carbon after several successive adsorption/desorption cycles of microwave regeneration. In contradiction with these previous findings, Çalişkan et al. [29] found that microwave regeneration did not lead to better desorption efficiencies compared to conventional heating. They reported on the low temperature regeneration of activated carbon saturated with promethazine by the microwave and conventional methods. The regeneration was examined in the range of temperatures 300–500 °C that was too low to desorb the chemisorbed fraction of the compound. The observed phenomenon was explained by an enhanced cracking of promethazine in the internal structure of the adsorbent due to direct interaction of microwaves with promethazine (polar compound) at the low temperatures (microwave-lysis).

Separately from the topic of microwave regeneration, there are a large number of papers devoted to microwave activation and modification of activated carbons [11,31,40–42]. Different carbonized precursors as tobacco stems, coconut shell chars, waste tea or oil palm stone chars are used for physical or chemical activation using microwaves. To achieve the required high temperature of activation process (about 1000 °C for physical activation and 500–900 °C for chemical activation) in short time high specific microwave powers are usually applied. A microwave-assisted activation is supposed to be more efficient compared to the conventional thermal process due to direct heating and high dielectric loss of different carbonaceous materials.

In the present work, regeneration of activated carbon loaded with toluene were investigated by microwave and conductive heating. The novelty of this work is that the two methods are compared at the same active powers (or real powers). Moreover, the temperature of the adsorbent bed is measured locally at four points to capture non-uniform temperature distribution. In addition, mass balance over the adsorbent bed is carried out by automatic sampling of the outlet gas for the GC analysis instead of weighting the adsorbent bed itself. To the best of author's knowledge such the approach is applied for the first time.

Our previous paper presents a comparison of microwave swing regeneration (MSR) and temperature swing regeneration (TSR) of acetone and toluene from 13X molecular sieves in terms of desorption kinetics and desorption efficiencies [22]. In this previous work, the comparison was made using different criterion than was used in the present work. These prior regenerations were carried out at similar temperatures in the both methods using a constant microwave power of 140 W and adjustable power in TSR. It was found that MSR runs faster even when the adsorbent temperature is lower than the gas temperature in TSR. The observed enhancement of microwave-driven desorption was more pronounced for the polar adsorbate (acetone) or high heat transfer resistances. Moreover, it was verified that microwaves do not affect the adsorption capacity of the molecular sieves after several consecutive adsorption-desorption cycles.

2. Experimental

2.1. Materials

Granular activated carbon (GAC), purchased from Chempur (Poland), was used in the experiments. The physico-chemical properties of GAC are listed in Table 1. Toluene (CAS: 108-88-3; purity ≥99.5%), purchased from Chempur (Poland), was used as the adsorbate. Nitrogen (purity ≥99.999%), delivered by Multax (Poland), was used as the purge gas.

2.2. Setups

Two experimental setups were utilized to perform (1) adsorption from the gas phase (Fig. 1a), and (2) microwave regeneration (MR) and conducting regeneration (CR) (Fig. 1b).

These experimental setups consist of a gas chromatograph GC-2014 (Shimadzu, Japan) with a two-position valve (6 port valve, Valco Instruments Co. Inc.), a laboratory microwave oven RM-800pc (Plazmatronika, Poland) with a monomode applicator, a mass flow controller ERG-1-MPSb (BetaErg, Poland), an ultrathermostat type 9506 (Polyscience, USA), a peristaltic pump PP1-05A (Zalimp, Poland) and a personal computer. Additional equipment (not shown in Fig. 1) comprise four fibre optic (FO) sensors type FOT-L-SD with a multichannel signal conditioner TMI-4 (FISO Technologies, Canada), a glass

Table 1
Physico-chemical properties of GAC.

Parameter	Value
Shape	cylindrical
Diameter	approx. 1 mm
Length	approx. 4 mm
Bulk density	454 g/dm^3
Mechanical strength	95.9%
Abrasion resistance	0.18%
Moisture content	2.1%
Methylene blue number	25 cm^3
Iodine number	997 mg/g
Water absorbability	0.88 cm^3/g
pH of water extract	10.4