INDIRECT AND DIRECT MICROWAVE REGENERATION OF Na-ETS-10

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Abstract

Indirect and direct microwave regeneration were assessed as potential techniques for desorbing a CO_2/CH_4 mixture from Na-ETS-10. Indirect microwave regeneration consists of desorption with water followed by microwave drying, while direct microwave regeneration consists of constant power microwave heating. Five adsorption-desorption cycles were completed to determine and compare the swing capacity, net energy consumption, and gas recovery obtained with these two regeneration techniques. During indirect microwave regeneration the swing capacity of Na-ETS-10 was stable at 0.3 mmol/g, with the exception of the first cycle where it reached 1.6 mmol/g. The first cycle swing capacity decreased however, by five fold during the subsequent desorption cycles. During direct microwave regeneration, the swing capacity remained stable at 0.7 mmol/g over all five consecutive cycles. On average, 2.46 KJ and 0.32 KJ were consumed during regeneration of one gram of Na-ETS-10 using indirect and direct microwave regeneration, respectively. With the exception of the first cycle, gas recovery for indirect microwave

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regeneration was only 20%. In contrast, an average of 50% gas recovery was achieved by direct microwave regeneration.

1. Introduction

Microwave heating is an emerging technology in the chemical process industries (Bykov et al., 2001) with the advantages of being cheaper and faster than conventional adsorbent regeneration techniques such as steam regeneration (Polaert et al., 2010). The fundamentals of microwave regeneration are unique and opposite to the mechanism of conventional thermal regeneration techniques (Das et al., 2009). In steam regeneration, the thermal energy is transferred from the surface to the bulk of the adsorbent bed. By contrast, the thermal energy in microwave regeneration is transferred from the inside to the outside of the adsorbent bed (Hashisho et al., 2005).

Finding an efficient method for regeneration of the adsorbents is important for chemical industries because it is a time-consuming process with high-energy requirements. In this context, microwave regeneration was shown to be successful in dehydrating and regenerating adsorbents saturated with volatile organic compounds (VOCs) (Roussy and Chenot, 1981; Cha and Carlisle, 2001; Hashisho et al., 2005). Dehydration of adsorbents using microwave heating was studied for zeolite 13X (Roussy and Chenot, 1981), zeolite 3A, 4A, 5A, (Ohgushi et al., 2001), Na-X, and Ca-X (Ohgushi and Nagae, 2003, 2005). Constant power microwave heating has also been used to regenerate dealuminated Y zeolite (DAY) (Reuβ et al., 2002; Turner et al., 2000), silicate (Meier, 2009), mordinate (Kim et al., 2005), faujasite (FAU) (Kim et al., 2007) and Engelhard titanosilicate (ETS-10) (Shi et al., 2010).

Natural gas is considered to be a cleaner alternative to other fuels and its supply currently meets one-fourth of the world's energy needs. Typically, natural gas contains traces of impurities such

as carbon monoxide, carbon dioxide, and nitrogen. In Australia and Germany, natural gas contains more than 10% carbon dioxide (CO_2) impurity (Caventi et al., 2004). This percentage needs to be reduced to meet the 'pipeline quality' (< 2% CO_2 impurity) set for methane (CH_4) in order to protect equipment and pipeline infrastructures (Cavenati et al., 2004). ETS-10, a titanosilicate molecular sieve with pores large enough to accommodate CO_2 and lighter hydrocarbons, is shown to purify CH_4 by preferentially adsorbing CO_2 over CH_4. (Kuznicki, 1991; Anderson et al., 1994). ETS-10 can preferentially adsorb CO_2 over C_2H_6 as well. (Anson et al., 2009). However like with adsorbents there is a need to efficiently regenerate and reuse ETS-10. Previous studies on regeneration of Na-ETS-10 with steam, conductive heating, and microwave heating techniques have shown that microwave regeneration is faster and more energy efficient than the other two techniques (Shi et al., 2010; Chowdhury et al., 2012). These studies reported that Na-ETS-10 provides maximum water desorption by mass action displacement but the energy requirement for drying wet Na-ETS-10 has yet to be determined (Shi et al, 2010, Chowdhury et al., 2012).

This study investigates an indirect method for regenerating Na-ETS-10 (water desorption followed by microwave drying) and compares it to direct microwave regeneration. In direct microwave regeneration CO_2/CH_4 mixture was adsorbed on a packed bed of Na-ETS-10 and then desorbed by water injection. The wet adsorbent was then dried and reactivated using microwave heating. In direct microwave regeneration, CO_2/CH_4 gas mixture was adsorbed on a packed bed of Na-ETS-10, and desorbed using constant power microwave heating. The swing capacity, gas recovery, and energy consumption achieved over five adsorption-desorption cycles were measured and compared between indirect and direct microwave regeneration.

2. Experimental

Na-ETS-10 was synthesized using the hydrothermal technique as described elsewhere (Kuznicki, 1991). Powder Na-ETS-10 was pelletized into 16-20 mesh pellets. A detailed method of pellet preparation can be found elsewhere (Shi et al., 2010).

Adsorption-desorption experiments were performed using an adsorbent bed 3.75cm long and 2.9cm in diameter containing 10g of Na-ETS-10 and a double ended cylindrical quartz column. Activation of the sample was completed at 200°C in a laboratory oven for 16h under 120ml/min helium gas flow. The feed gas mixture (Praxair) of 10%CO_2 and 90%CH_4 was introduced into the fixed bed column with a flow rate of 300 mL/min at 22°C and 101.325 kPa. The outlet gas was sampled and analysed by using a gas chromatograph (Agilent 5890) equipped with a thermal conductivity detector and Supelco matrix Heysep Q column. Adsorption proceeded under a continuous flow of feed gas until saturation when the outlet composition became the same as the feed composition. Na-ETS-10 became saturated with CO_2/CH_4 after 90 minutes.

In the indirect microwave regeneration, 5ml water was injected into the saturated adsorbent. The desorbed gas flowed to a downstream flask and was collected by water displacement. The experiment was carried out until no gas evolution was observed. The volume of the displaced water was equal to the volume of the gas that was collected at the outlet. After desorption with water, a microwave generation and propagation system was used to dry the adsorbent. The system consisted of a 2 kW switch-mode power supply (SM745G.1, Alter), a 2 kW variable output microwave generator (MH2.0W-S, National Electronics) equipped with a 2.45 GHz magnetron, an isolator (National Electronics), a three-stub tuner (National Electronics), and a

waveguide applicator connected to a sliding short (IBF Electronic GmbH & Co. KG). At the beginning of the experiment the tuner and the sliding short were manually adjusted for matching the impedance and improving the energy transfer to the adsorbent. To determine energy consumption, power was monitored using a dual channel microwave power meter (E4419B, Agilent), two power sensors (8481A, Agilent), and a dual directional coupler with 60 db attenuation (Mega Industries). To avoid interference with the electromagnetic field, a fiber optic temperature sensor with a signal conditioner (Reflex signal conditioner, Neoptix) was used to monitor the temperature of the adsorbent during microwave heating. A data acquisition and control (DAC) system (Compact DAC, National Instruments) equipped with a Labview program (National Instruments) was used to record the signals from the temperature sensor, and power meter and control the output of the power supply. The Labview program was used to monitor and control the heating. During the microwave drying, a 120 sccm nitrogen flow was used as purge gas to provide uniform heating. After microwave drying, the nitrogen flow was adjusted to 300 sccm for faster cooling of the bed to room temperature. Once the bed reached room temperature, further adsorption-water desorption-microwave drying cycles were initiated. A block diagram showing adsorption and regeneration by water desorption followed by microwave drying is presented in Figure 1.

The same microwave generation system was used for both indirect and direct microwave regenerations. After saturation of the adsorbent, the microwave generation system was turned on, and the heating was initiated using the Labview program. The adsorbent was exposed to a constant incident microwave power of 60W until the bed temperature reached its set-point, then the heating was stopped. During the regeneration, the desorbed gas flowed to a downstream flask and was collected by water displacement. Microwave regeneration was continued until no gas

evolution was observed. Then, the adsorbent was cooled to room temperature by purging with nitrogen at 120 sccm. After the bed reached ambient temperature, further adsorption-microwave desorption cycles were initiated. A block diagram showing adsorption and constant power microwave regeneration is presented in Figure 2.

For water desorption, the swing capacity of Na-ETS-10 is defined as the amount of gas desorbed during water injection. For microwave regeneration, the swing capacity is defined as the amount of gas desorbed during microwave heating. Gas recovery was calculated based on equation 1:

$$\text{Gas recovery (\%)} = (V_{W/M}/V_W)\, x100 \ldots\ldots\ldots\ldots (1)$$

where, $V_{W/M}$ = volume of gas desorbed by water desorption (W) or microwave (M) heating, and V_W = the volume of gas desorbed by water desorption from the fresh adsorbent.