# Synthesis of Mesoporous Silica Materials from Incineration Bottom Ash for the Removal of Toluene

Zhen-Shu Liu and Hai-Ming Lan

*Abstract*—In Taiwan, most of the municipal solid waste is incinerated, and more than 930 thousand tons of bottom ash is produced in 2014. Incineration bottom ash contains major element of silicon that could be as a silicon source for the synthesis of mesoporous materials. Our previous study investigated the conditions for alkaline fusion to extract silica from incinerator bottom ash and verified that the high specific surface area of mesoporous silica could been synthesized from bottom ash. To expand the application of mesoporous silica materials synthesized from bottom ash, the effects of operation conditions of adsorbent weight (0.2, 0.5, 0.8, 1.5 g), adsorption temperature (25, 30, 35 ℃) and toluene concentrations (900, 1100, 1300, 1500 ppm) on toluene removal were determined in this study. Moreover, the gas flow and the oxygen content were 500 sccm and 10%, respectively. The mesoporous materials synthesized from bottom ash possessed the best adsorption capacity of toluene (ca. 124 mg g-1) at the toluene concentration of 1100 ppm and at the reaction temperature of 25 ℃. The results showed that the mesoporous silica prepared with incinerator bottom ash have potential to be used as an adsorbent for the removal of toluene.

*Index Terms*—Adsorption, incineration bottom ash, mesoporous materials, toluene.

## I. INTRODUCTION

Mesoporous silica materials exhibited high specific surface area, and good thermal and hydrothermal stabilities have been used extensively as catalysts and adsorbents [1]-[4]. Silica is the main raw material to synthesize these mesoporous silica materials. However, silica derived from inorganic silicates (such as sodium silicate) is expensive. Therefore, an alternative source of silica must be found. In 1985, $SiO_2$ and $Al_2O_3$ were firstly extracted from coal fly ash using an alkaline hydrothermal reaction [5]. Then the $SiO_2$ and $Al_2O_3$ were used to synthesize microporous molecular sieves. Subsequently, many studies investigated the synthesis of mesoporous silica materials using silica source from coal fly ash [6]-[13]. Lee et al. indicated that the fly ash with high silica content has the potential for the synthesis of nanoporous materials, such as zeolites, mesoporous silica materials [8]. However, processing of mesoporous silica materials from incinerator bottom ash is obscure. The ash composition, pretreatment procedures, and hydrothermal synthesis conditions would affect the purity and yield rate of the synthesized nanoporous materials.

Incinerator bottom ash containing a large amount of silica can be used as a raw material for synthesizing mesoporous silica materials [14], [15]. This would not only increase the recycling rate of bottom ash but also reduce the cost of mesoporous silica materials. In Taiwan, the recovery rate of incineration bottom ash is approximately 77% in 2014, and 23% of bottom ash was disposed in landfill. Seeking new recycling techniques to increase the recovery rate and value added of bottom ash is important. Therefore, mesoporous silica materials synthesized from incinerator bottom ash was investigated in our previous study [14].

Our previous study verified that the high specific surface area of 992 $m^2/g$ of mesoporous silica could been synthesized from incineration bottom ash. The mesoporous silica also has been used for the removal of heavy metals from waste water [14]. It has potential for the synthesis of mesoporous silica from bottom ash and the application of mesoporous silica in the removal of pollutants. Therefore, the synthesis of mesoporous silica materials from incineration ash for the removal of toluene in a fluidized-bed reactor is investigated in this study.

## II. METHOD

The bottom ash used in this study was obtained from an incinerator in New Taipie City, Taiwan. The bottom ash was completely dried at 105 ℃ in an oven and then crushed, ground, and sieved (<0.35 mm). The ash less than 0.35 mm in size was mixed with $Na_2CO_3$ in a certain ratio, and then placed it in a high-temperature furnace at 900 ℃ for 15 min for alkali fusion. The fused mass was dissolved in deionized water and placed in an oven at 105 ℃ for 24 h for hydrolysis reaction. Subsequently, the mixed solution was filtered to obtain the supernatant. The templating agent for synthesizing mesoporous materials in this study was stirred and mixed with deionized water at 45 ℃ using cetyltrimethyl ammonium bromide (CTAB). The aqueous CTAB solution was slowly introduced into the silica-containing supernatant solution. Then, 1M sulfuric acid was added into the mixture until the pH value was 10. Subsequently, the solution was poured into an PP bottle, which was placed in an oven at 105 ℃ for hydrothermal reaction, and after being cooled to room temperature and filtrated. The filtered solid was washed with deionized water and placed in an oven for drying, and then placed in a high-temperature furnace for calcination. After cooling, the filtered solid yielded mesoporous silica.

The physical and chemical characteristics of mesoporous materials were analyzed using various instruments. To study the effect of the porosity of the mesoporous silica materials

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synthesized from bottom ash on toluene adsorption, an ASAP2020 vacuum volumetric sorption instrument was used to determine the porosities and BET surface area of the synthesized mesoporous silica materials. The specific surface area was determined by BET method. Since the micropore volumes of the silica materials was zero, the mesopore volumes (pore size: 2–50 nm) were the total pore volumes at a relative pressure of 0.98. A transmission electron microscope (TEM) (JEM-2100) and a field-emission

scanning electron microscopy (FE-SEM) (JEOL JSM-7600 F) were used to analyze the pore structure and morphologies of the mesoporous silica materials, respectively. An X-ray powder diffraction (XRD) system (SIEMENS D5000) was also used to observe whether the mesoporous silica has the hexagonal pore structure. The stability of the mesoporous silica can be determined by a thermogravimetric Analyzer (TGA) (Perkin Elmer STA6000).

TABLE I. EFFECTS OF VARIOUS ADSORPTION CONDITIONS ON THE ADSORPTION CAPACITY OF TOLUENE

| Test | Adsorbent weight (g) | Adsorption temperature ( ℃) | Volumetric flow rate (ml/min) | Toluene concentration (ppm) | Saturated adsorption capacity (mg/g) |
|---|---|---|---|---|---|
| Run 1 | 0.2 | 25 | 500 | 1100 | 124 |
| Run 2 | | 30 | | | 68 |
| Run 3 | | 35 | | | 87 |
| Run 4 | 0.5 | 25 | | | 112 |
| Run 5 | 0.8 | | | | 116 |
| Run 6 | 1.5 | | | | 115 |
| Run 7 | 0.2 | | | 900 | 79 |
| Run 8 | | | | 1300 | 88 |
| Run 9 | | | | 1500 | 94 |

Toluene adsorbed onto mesoporous silica was carried out in a fixed-bed reactor. Fig. 1 shows the experimental setup. The inner diameter of the adsorption reactor was 10 mm. The toluene gas was generated by bubbling $N_2$ into liquid toluene. $N_2$ and air were the balance gases to maintain the desired toluene concentration and oxygen content. The oxygen content and the volumetric flow rate of feed were 10% and 500 sccm, respectively. After the inlet concentrations of toluene and oxygen reached a steady state, the adsorption of toluene was proceeded in the adsorption reactor. The detailed adsorption conditions are shown in Table I. An online GC (Shimadzu GC-2014) was used to monitor he toluene concentration at the inlet and outlet of the reactor. After equilibrium adsorption of toluene was achieved, the saturated adsorption capacity of toluene was calculated using numerical integration of the breakthrough curves, which represent the outlet concentrations of toluene versus time [16].
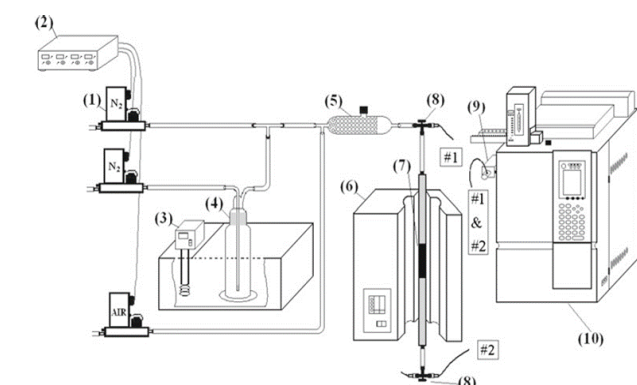


(1) Mass flow controller, MFC, (2) Power supply of MFC readout, (3) Thermostat water bath, (4) Gas bubbler filled with toluene, (5) Gas mixing chamber, (6) Heating unit, (7) Adsorption bed, (8) Three-way valve, (9) Gas sampling, (10) GC–FID
Fig. 1. Experimental setup for the adsorption of toluene.

## III. RESULTS AND DISCUSSIONS

### A. Physical and Chemical Properties of Mesoporous Silica Mmaterials

Fig. 2 shows the SEM image of mesoporous materials. SEM micrograph revealed that the particles of the synthesized mesoporous materials were crystalline and had a smooth surface. Fig. 3 shows the TEM image of mesoporous materials. TEM image showed that the hexagonal pore structure of mesoporous materials were unobvious. The result can be explained by the fact that other metal impurities are also present in the supernatant solution extracted from the bottom ash.
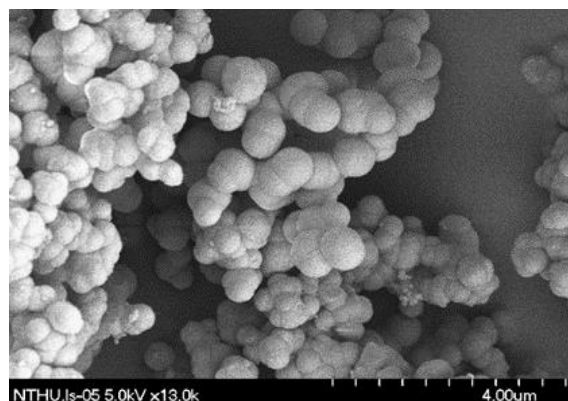


Fig. 2. SEM image of mesoporous materials.

The BET results indicated that the specific surface area and mesoporous volumes of mesoporous materials were 992 $m^2/g$ and 0.854 $cm^3/g$, respectively. Fig. 4 and Fig. 5 display the $N_2$ adsorption–desorption isotherms and the pore size distribution of the mesoporous materials. Fig. 4 indicated that the isotherm was of Type VI with an H1 hysteresis hoop due

to the capillary condensation in mesopores [17], [18]. Further, the silica materials exhibited a narrow pore size distribution (as shown in Fig. 5). The average pore size of the mesoporous silica was 3.4 nm.
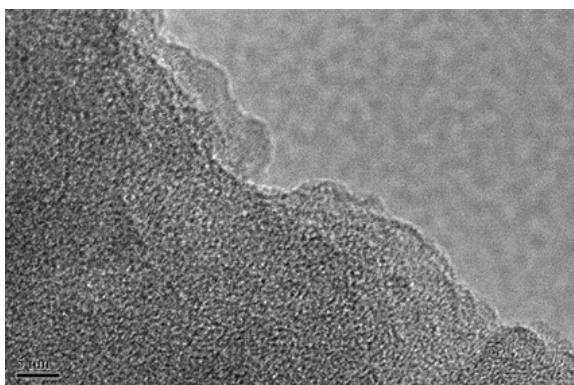


Fig. 3. TEM image of mesoporous materials.

Fig. 6 shows the XRD pattern of mesoporous materials. One peak corresponded to the (100) plane was observed in XRD patterns, indicating a hexagonal pore structure [19], [20]. Fig. 7 shows the TGA pattern of mesoporous materials. TGA patterns demonstrated that the synthesized mesoporous materials exhibited good thermal stability and hydrophobic property.
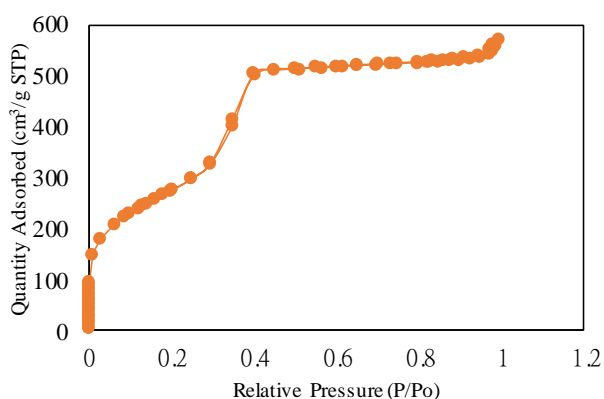


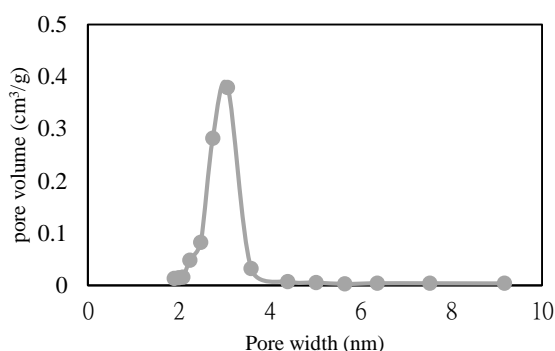Fig. 4. $N_2$ adsorption and desorption isotherms of mesoporous materials.



Fig. 5. Pore size distribution of mesoporous materials.

### B. Removal of Toluene

Table I shows the effects of various adsorption conditions on the adsorption capacity of toluene. The mesoporous materials synthesized from bottom ash possessed the best adsorption capacity of toluene (ca. 124 mg $g^{-1}$) at the toluene concentration of 1100 ppm and at the reaction temperature of 25 ℃. The data indicated that the adsorption capacity of toluene increased with increasing toluene concentration. The results also showed that the breakthrough times increased with increasing adsorbent weight, but the change in the adsorption capacity of toluene was insignificant. Ma and Ruan indicated that the adsorption capacity of toluene in mesoporous silica materials (S-MCM) synthesized from waste solar panel was 57, 104, 200 and 277 mg $g^{-1}$ for initial toluene concentrations of 250 to 1500 ppm respectively [21].
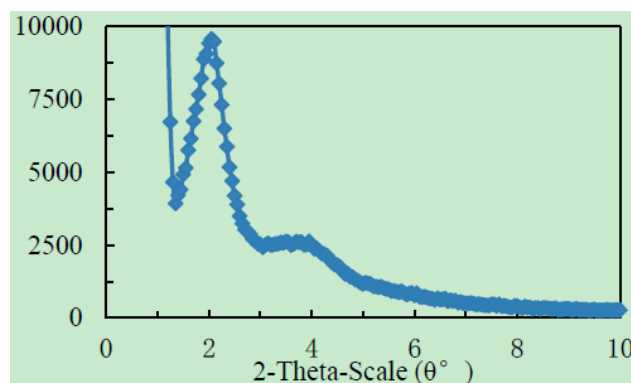


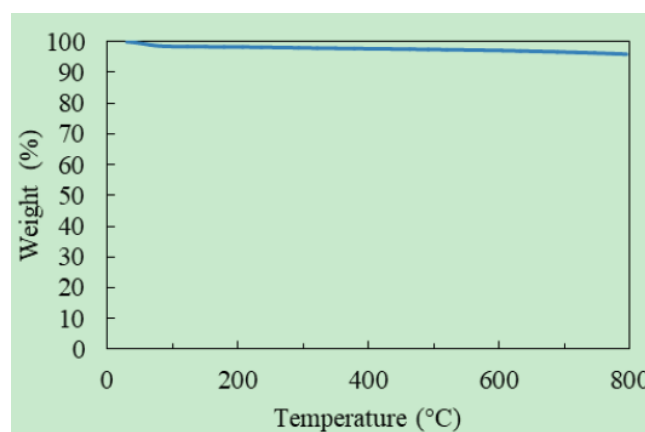Fig. 6. XRD pattern of mesoporous materials.



Fig. 7. TGA pattern of mesoporous materials.

## IV. CONCLUSIONS

This study investigated the synthesis of mesoporous silica materials from incineration ash for the removal of toluene in a fluidized-bed reactor. The results showed that the mesoporous silica has an optimum adsorption capacity of toluene (ca. 124 mg $g^{-1}$) at the toluene concentration of 1100 ppm and at the reaction temperature of 25 ℃. The mesoporous silica prepared with incinerator bottom ash has potential to be used as an adsorbent for removing toluene after being modified.

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