



Development of an Innovative Circulating Fluidized-Bed with Microwave System for Controlling NO_x

Yuan-Yi Chang¹, Yeou-Lih Yan², Chao-Heng Tseng¹, Jin-Yuan Syu¹, Wen-Yinn Lin^{1*}, Yu-Chun Yuan¹

¹ Institute of Environmental Engineering and Management, National Taipei University of Technology, 1, Sec. 3, Chung-hsiao E. Rd., Taipei, 10608, Taiwan

² Department of Safety, Health and Environmental Engineering, National United University, 1, Lienda, Miaoli, 36003, Taiwan

ABSTRACT

Selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR) technique are widely used to control nitrogen oxides emissions. However, both techniques have a general shortcoming known as NH₃ slip.

This research introduced the design of activated carbon in a circulating fluidized-bed with microwave system and developed an innovative de-NO_x technique. The whole system demonstrated the capability and advantage of reducing reductant cost and continuous process. The experiments investigated microwave to regenerate activated carbon (AC) in order to increase adsorption and destruction efficiency while reducing energy consumption. In the NO_x abatement process, activated carbon adsorbed NO and NO₂ and then utilized microwave heating technology to regenerate itself because of microwave's high energy utilization and strong penetration ability. The specific surface areas of AC increased from 673.03 to 834.52 (m²/g) when microwave power was increased from 0 to 550 W, respectively, in this study. Through increasing the specific surface area, the microwave treatment further improved the NO_x adsorption capacity and rate.

In consequence, the results indicated that destruction efficiency of NO and NO₂ at 200 ppm could reach about 80% with microwave power of 350 W and above 85% of 550 W. The destruction efficiency at 550 W for NO_x was about 77%.

Keywords: Nitrogen oxides; Microwave; Activated carbon; Circulating fluidized-bed.

INTRODUCTION

The abatement of NO_x emissions has become a global issue, because NO_x are related to acid deposition and photochemical smog (Singoredjo *et al.*, 1993; Yang *et al.*, 2000; Yang *et al.*, 2000; Kumar *et al.*, 2008; Lee *et al.*, 2010; Han *et al.*, 2011; Li *et al.*, 2011; Colbeck *et al.*, 2011; Peng *et al.*, 2011). Selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR) technologies are extensively used to control NO_x emissions from chemical factories and stationary power generation sources (Wójtowicz *et al.*, 1993; Yang *et al.*, 2000; Chen 2006; Lou *et al.* 2003). Both techniques also have a universal imperfection known as NH₃ slip, which is simply the discharge of unreacted NH₃ into the stack. The utility of SCR and SNCR critically relies on good process control as poorly chosen NH₃/NO_x ratio, temperature, or location of NH₃ injection that may

bring on a raise in NO_x emissions. Furthermore, SNCR has been described to increase incomplete NO_x reduction and the formation of nitrous oxide (N₂O) (Hjalmarsson, 1992).

Activated carbon (AC) obtains special attention due to its unique characteristics for adsorption of atmospheric pollutants. Different NO_x abatement methods have been recommended. AC is considered valuable either as catalyst support or as solid reactant for NO_x reduction to nitrogen (N₂) and can evade reductant slip (Ahmed *et al.*, 1993; Teng *et al.*, 1992; Illán-Gómez *et al.*, 1996). AC is usually prepared from the pyrolysis and the following activation, either by physical or chemical processes, of coal, wood by-products, various types of cellulose materials and pitches (Ahmadpour and Do, 1996; Illán-Gómez *et al.*, 1996; Molina-Sabio *et al.*, 1996). These AC, with large porous structure and high surface area, is broadly studied and used in many practical applications (Mochida *et al.*, 2000; Shirahama *et al.*, 2002; Lee *et al.*, 2002). Nevertheless, the influence of the type of active carbon on NO_x adsorption capacity has shown equivocal consequences. The capacities of nitric oxide (NO) adsorption of AC are not directly correlated with its specific surface area or its pore volumes (Neathery *et al.*, 1997).

* Corresponding author. Tel.: +886-2-2771-2171 ext. 4131;
Fax: +886-2-2778-1598
E-mail address: wylin@ntut.edu.tw

Microwave techniques applied to a pyrolytic carbon such as activated carbon and char could promote the reaction of NO_x with carbon to produce N_2 , carbon oxide (CO) and CO_2 (Cha, 1994; Cha and Kong, 1995; Kong and Cha, 1995; Kong and Cha, 1996; Kong and Cha, 1996). Microwave is a kind of electromagnetic energy transpiring in the frequencies range from 300Hz to 300GHz (Thuéry, 1992). Within this range, there are four frequencies usually for industrial utilization: 915M, 2.45G, 5.8G, and 22.125GHz. Most of the industrial microwave facilities employ 2.45GHz as its working frequency. Microwave energy causes molecular motion by rotation of dipoles and migration of ions (Plazl *et al.*, 1997). Heating by microwave relies on the ion conductivity, volume of the sample, and dipole relaxation time (Barringer *et al.*, 1994). The heat resulting from microwave energy is mostly due to two different effects. In the case of polar molecules, the electric field component of the microwaves results in induced dipoles to rotate with the alternating field. This molecular movement produces friction among the rotating molecules, and the energy is subsequently dispersed as heat (dipolar polarization). This is the instance of water and other polar fluids. In the case of dielectric solid materials with charged particles which are free to move in a delimited zone of the material, such as π -electrons in carbon materials, a current migrating in phase with the electromagnetic field is formed. Since the electrons cannot couple with the changes of phase of the electric field, the energy is dispersed in the type of heat due to the so-called Maxwell-Wagner effect (Zlotorzynski, 1995; Meredith, 1998).

General conclusions deduced from the above literature review can be summarized as follows. AC will be a comely alternative material for NO_x reduction and it can be used either as catalyst support or as solid reactant. The application

of AC with microwave can enhance the reaction of NO_x reduction to N_2 . In addition, microwave technology can make AC to be recycled and reused numerous times. Hence, it is shown that microwave heating supports the porous structure of the regenerated AC more efficiency than treatment in a conventional device (Ania *et al.*, 2005).

In spite of many studies have been researched on the utilization of AC with microwave treatment in NO_x reduction, but none was found by the authors to carry out on AC in microwave equipment treating with cyclone collector continuously. There were two objectives of this study: (1) to investigate the design of AC in a circulating fluidized-bed with microwave system, and (2) to experimentally research the influence of destruction efficiency of NO_x by testing the different amount of AC with different microwave power.

METHODS AND MATERIALS

The experimental set-up, shown in Fig. 1, consisted of four major parts, i.e. the adsorption system, the microwave system, the outlet gas emission measurement system, and the gas feed system.

A commercial AC with diameter of 0.25–0.6 mm (30 × 60 mesh) was used as the adsorption material in this study. Different amount of AC (180, 210, and 240 g) was tested to understand its influence. These AC powders were pretreated at 105°C in oven for 2 hours and then cooled down to the room temperature in a dry box. The diameter of adsorption tube was 10 mm and the length of the tube was 3 m. The feed gas and the AC were separated by a cyclone collector which had a cutoff size (d_{50}) of 5.27 μm . After separation, the AC was collected into a quartz tube and the outlet gas was exhausted through the vent of the cyclone collector. The quartz tube ($\phi 5 \times 50 \text{ cm}$) was

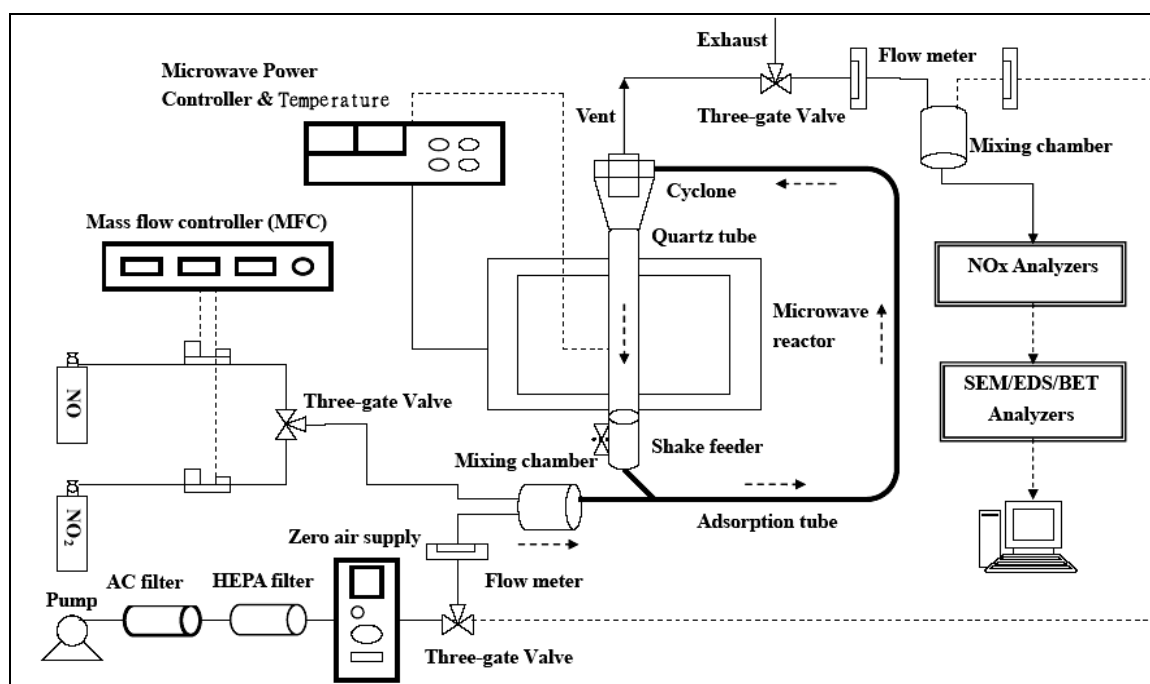


Fig. 1. Schematic diagram of the experimental system.

vertically centered in the microwave oven. All the experiments were performed under atmospheric pressure. In quartz tube, the retention time of AC increased as the amount of AC increased. While the quantity of AC was 180, 210, and 240 g, the average retention time was 148, 198, and 252 seconds, respectively.

A household microwave oven (MOB-201R, Sampo Corp., Taiwan), with a maximum power of 650W and frequency of 2.45 GHz, was modified to have variable power setting mode by using a power controller. The experiments were conducted at different microwave power of 350, 450, and 550 W.

The gas feed system consisted of cylindrical NO and NO₂, a zero air supply, and a mass flow controller. The concentrations of NO and NO₂ in the gas stream were controlled by the flow rates of zero air and the cylindrical gas. The total gas flow rate was maintained at 15 L/min. The concentrations of NO and NO₂ were 200 and 400 ppm, respectively. And the concentration of NO_x was mixed with 200 ppm NO and 100 ppm NO₂. The zero air supply (Thermo Electron Model 111) generated the pollutant free zero gas for different concentration of NO, NO₂, and NO_x requirements. A NO_x analyzer (Thermo Environmental Instruments Model 42) was used to continuously measure the concentrations of NO, NO₂, and NO_x in the vent.

The AC powders were characterized by measuring surface area and morphology. The surface area was determined by N₂ adsorption at 77 K, using the classical Brunauer-Emmett-Teller (BET) equation. The morphology was determined by scanning electron microscopy. Scanning electron microscopy and energy dispersive X-ray analyses were performed using a SEM/EDX, Hitachi S-4700 Scanning Electronic Microscope.

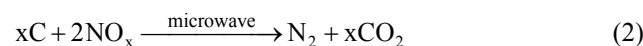
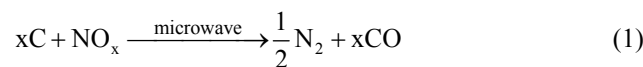
RESULTS AND DISCUSSION

The Effect of Microwave Power on Temperature of Activated Carbons

The AC was heated under microwave radiation at three power levels and each run was 30 minutes. Perhaps electronic

ions might induce AC flame in a longer operation time; however, no induced flame around AC was observed by microwave radiation during the experimental period. The trend illustrated that temperature increased speedily when microwave induced. Compared with conventional heating techniques, microwave heating proposes not only higher heating rate but also material selective heating. Chen *et al.* (1984) indicated that dark colored compounds could be heated rapidly with microwave radiation to high temperature. As shown in Fig. 2, the higher microwave power output was, the higher temperature of the AC was. This trend was similar to that reported by Kingman *et al.* (2004) and Lee *et al.* (2007). Ramesh *et al.* (1999) demonstrated that the total power absorbed by the material principally relied on the reflection coefficient and attenuation constant of the material. For a homogeneous material, reflection coefficient could be submitted as a function of the root-mean-square value of the complex dielectric constant. Nevertheless, attenuation constant was an outcome of the free space permittivity and the relative dielectric constant of the material. At a constant microwave frequency, both the attenuation constant and the loss tangent of the substance depend on the temperature.

The reaction temperature played an important role on NO_x–C reaction. Simplified reactions of carbon with NO_x were described below, which would depend on the carbon temperature:



Eq. (1) and (2) were exothermic and thus favorable at low temperature. Accordingly, NO_x might be reduced selectively by regulating the carbon-bed temperature. In the low temperature region (< 677°C), reaction (2) was the primary reaction and the products were N₂ and CO₂. Increasing the reaction temperature, the effect of reaction (1) gradually

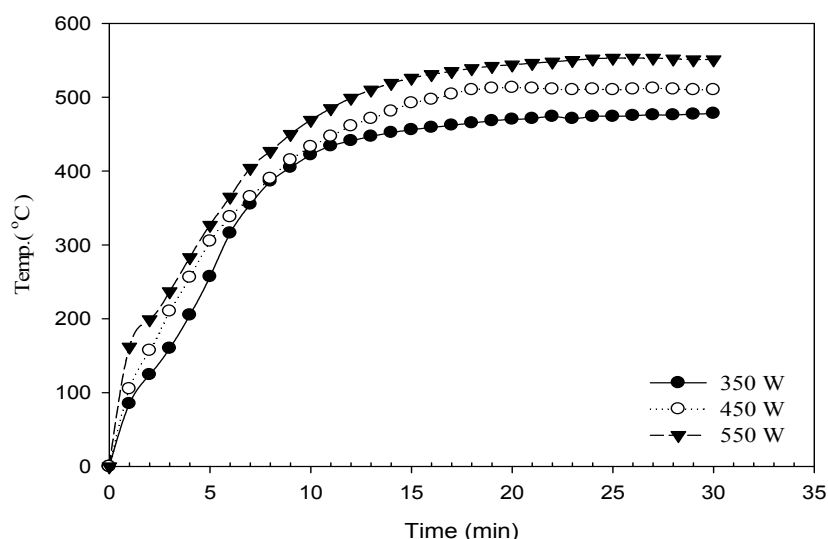
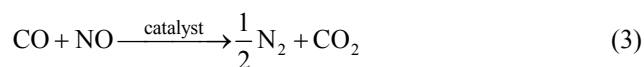


Fig. 2. Temperature variation of AC under various microwave power (180 g).

augmented (Jones *et al.*, 1999; Cha and Kim, 2001). Another possible reaction between NO and carbon is described as follow:



In the meantime, the increased CO enhanced the reaction (3), leading to the increase of the NO conversion (Furusawa *et al.*, 1985).

The Effect of Microwave Power on the Destruction Efficiency

Effects of microwave power on the NO, NO₂, and NO_x destruction efficiency were investigated for NO only (200

and 400 ppm), NO₂ only (200 and 400 ppm), and NO_x (200 ppm NO and 100 ppm NO₂). The quantities of AC, 180, 210, and 240 g were used, the average retention time was 148, 198, and 252 seconds, respectively. Considering the stability of microwave power, the lowest microwave power used for experiments was 350 W.

Figs. 3–5 demonstrated the decomposition efficiency as a function of retention time. As shown in Figs. 3–5, the efficiency increased with the retention time and microwave power increased. At 350 W, the destruction efficiency of 200 ppm NO and NO₂ increased from 74 to 79% and 77 to 81%, respectively, with increasing the retention time from 148 to 252 seconds. While the microwave power increased to 550 W, the destruction efficiency of NO and NO₂ increased from 80.5 to 87% and 81 to 85%, respectively, with increasing

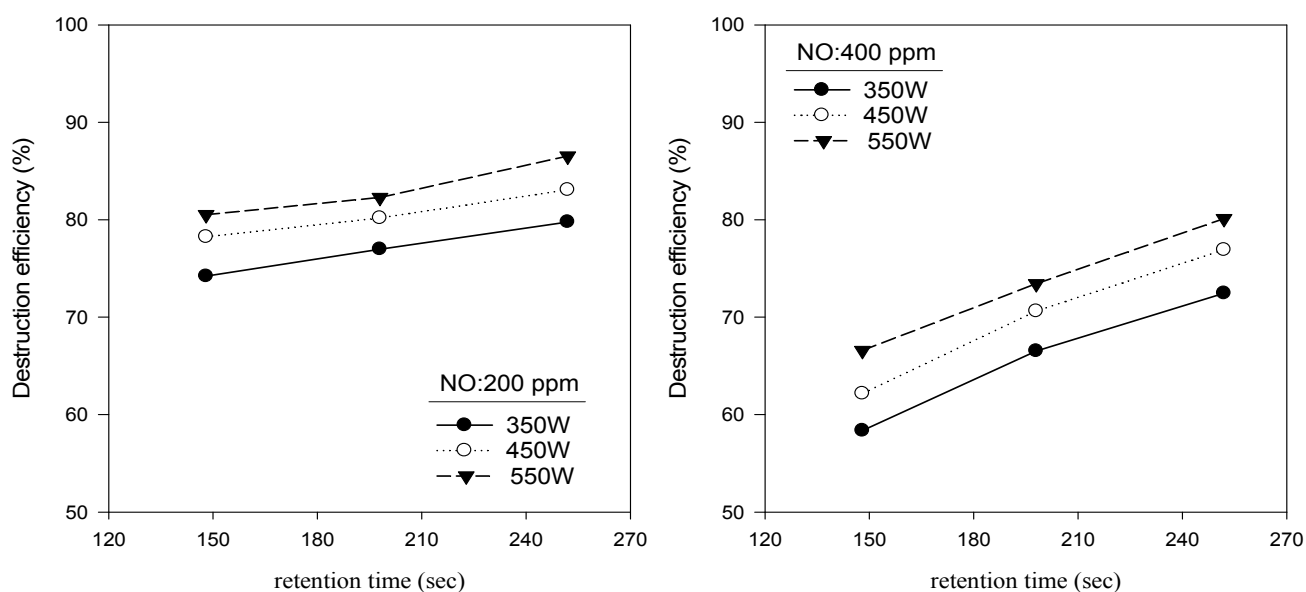


Fig. 3. Destruction efficiencies of 200 and 400 ppm NO with various microwave power and retention time.

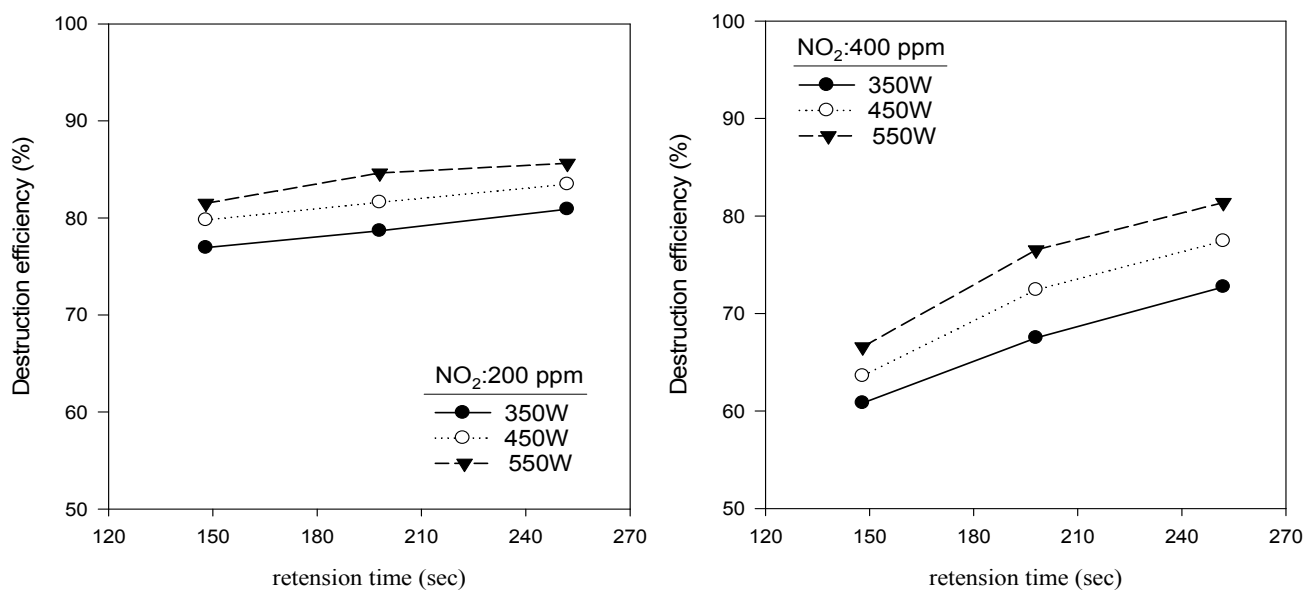


Fig. 4. Destruction efficiencies of 200 and 400 ppm NO₂ with various microwave power and retention time.

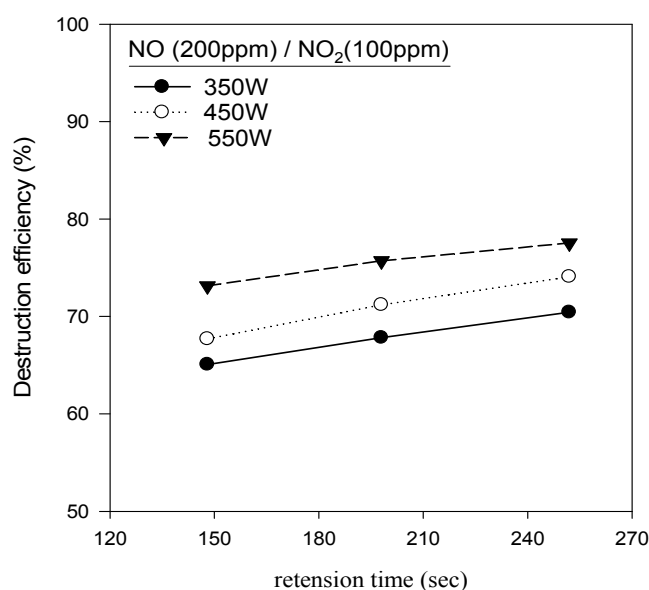


Fig. 5. Destruction efficiencies of NO_x (200 ppm of NO + 100 ppm of NO₂) with various microwave power and retention time.

the retention time from 148 to 252 seconds. At 350 W, the destruction efficiency of 400 ppm NO and NO₂ increased from 58 to 72% and 61 to 72%, respectively, with increasing the retention time from 148 to 252 seconds. When the microwave power increasing to 550 W, the destruction efficiencies of NO and NO₂ increased from 66 to 80% and 67 to 81%, respectively, with increasing the retention time from 148 to 252 seconds. The destruction efficiency of NO_x (200 ppm NO and 100 ppm NO₂) increased from 65 to 70% with increasing the retention time from 148 to 252 seconds at 350 W microwave power. At 550 W, the decomposition efficiency of NO_x raised from 73 to 77% as increasing the retention time. These results were similar to that reported by Cha and Kim (2001), Wójtowicz *et al.* (2000), Radoiu *et al.* (1998), and Radoiu *et al.* (2003). Rubel *et al.* (1995) indicated that under certain conditions the overall adsorption capacities might be independent of the amount of AC used. In the existence of oxygen, NO might be catalytically oxidized to NO₂ on the carbon surface (Kong and Cha, 1996); when in the poverty of oxygen ambient temperature, very little NO was adsorbed on porous adsorbents (Lee *et al.*, 2002; Kaneko and Imai, 1989). Yang *et al.* (2000) reported that NO could be reversibly adsorbed on the carbon surface forming a complex. Formation of a NO dimer was suggested as a possible mechanism to illustrate the adsorption of NO at 100°C (García *et al.*, 2002). At higher temperatures, desorption of these complexes could produce N₂ and carbon dioxide.

However, it was suggested by Radoiu *et al.* (2003) that the microwave treatment of NO_x was not a powerful method at power levels below 400 W because the reaction rate of reassociation was higher than that of destruction.

SEM Micrographs, EDX and BET Analysis

The adsorption on and desorption from AC with a porous system might differ from each of spherical shaped properties. The morphology of AC was shown in Fig. 6.

The surface was uneven and displayed microporosity without any cracks and breaks. In Figs. 6(b)–6(d), the substance showed higher porosity and broader pores after regeneration (350, 450, and 550 W). At 550 W, the surface of AC presented higher porosity, many cavities over the surface developing an advanced pore network system.

The composition and specific surface area of the AC were measured by EDX and BET analysis and the results were presented in Table 1. As the microwave power increased, the specific surface area of AC was increased. The plasma powder might cause the specific surface areas of AC increasing. Illán-Gómez *et al.* (1993) and Calo *et al.* (1999) indicated that the specific surface area of AC played a major role in the NO—C reaction. The higher available specific surface area of the AC, the more effective it was for NO_x reduction. Moreover, the ratio between pore diameter and molecular size of the adsorbate also played an important role in the adsorption potential for gas adsorption in micropores (Young and Crowell, 1962; Jovanović, 1969).

CONCLUSIONS

This research emphasized an innovative design of AC in a circulating fluidized-bed with microwave system for treatment of NO_x to understand the capability of an effective pollution control method. In this study, the exploratory results suggested that the reaction temperature and destruction efficiency were increased with increasing microwave power. Besides, the destruction efficiency was raised with increasing the AC dosage. According to SEM and EDX analysis, the AC showed higher porosity, broader pores after microwave treatment. In BET analysis, the results indicated that the specific surface area was increased as increasing the microwave power.

These results suggested that the NO_x decomposition efficiency of microwave associated would probably be more effective than the ones obtained using traditional techniques.

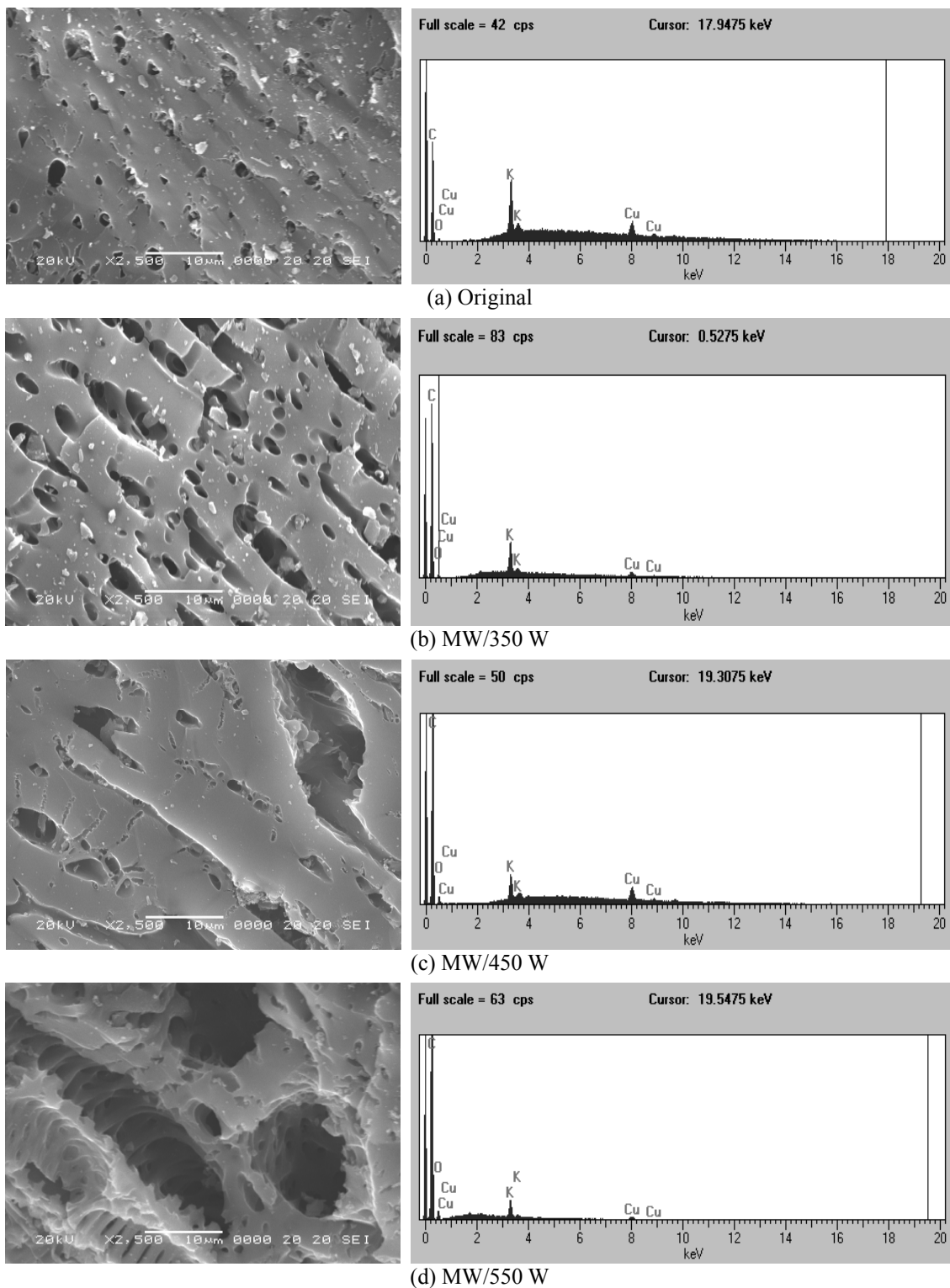


Fig. 6. SEM and EDX graphs of activated carbon (under 2,500 magnifications).

Table 1. BET and EDX analysis of activated carbon with various microwave power.

Sample	BET (m^2/g)	C (wt.%)	O (wt.%)	K (wt.%)	Cu (wt.%)
original	673.03	61.65	10.48	10.72	17.15
350W	702.97	76.04	8.85	8.06	7.05
450W	759.48	83.06	7.25	5.3	4.39
550W	834.52	88.25	4.07	3.9	3.78

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