

# Recycling C-14 generated from PHWR as $\text{CaCO}_3$ through an aerosol process

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## 1. Introduction

Considering the amount of C-14 in radioactive waste generated from domestic nuclear power plants, it is necessary to be treated as new materials including C-14. We investigated the preparation of sphere-shaped  $\text{CaCO}_3$  for high energy density and uniform properties. We tried to convert  $\text{CaCO}_3$  into CaO form and then re-adsorb  $\text{CO}_2$  generated from the PHWR waste resin to CaO form.  $\text{CaCO}_3$  containing C-14 could be used as a variety of materials. An ultrasonic aerosol process was introduced to produce sphere-shaped  $\text{CaCO}_3$ .

## 2. Methods and Results

### 2.1 Methods

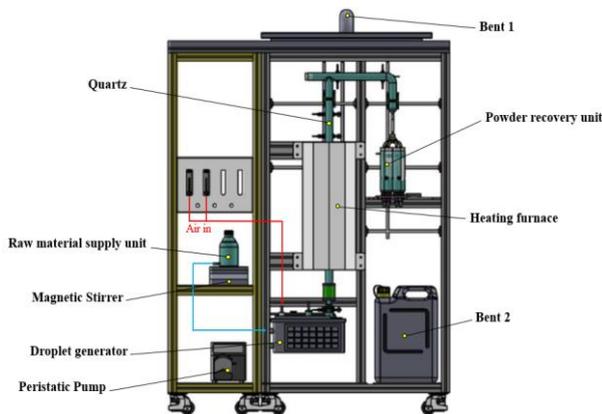


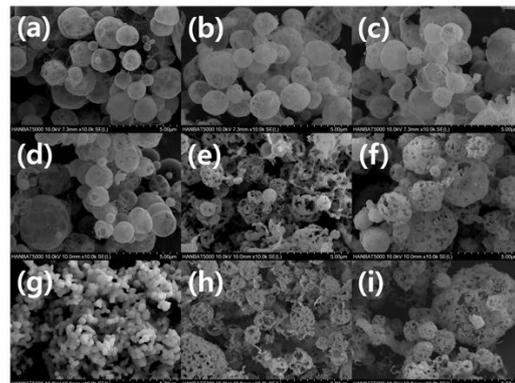
Fig. 1. Aerosol processing apparatus

In this study, an aerosol processing apparatus equipped with an ultrasonic droplet generator was used to make appropriate calcium carbonate particles. Aerosol processing apparatus shown in Fig. 1. The ultrasonic aerosol processing apparatus is composed of a raw material supply unit, a droplet generator, a heating furnace, a powder recovery unit and an exhaust line. The raw materials are composed of gas and liquid phase, and each raw material is introduced into the droplet generator through the raw material supply unit. At this time, the introduced gas acts as a carrier gas and transports the droplets generated by ultrasonic waves to the inside of the heating furnace. Liquid and gas react to form the final product. De-ionized water, citric acid, ethylene glycol, calcium nitrate and calcium acetate were used as the precursors of  $\text{CaCO}_3$ , and compressed

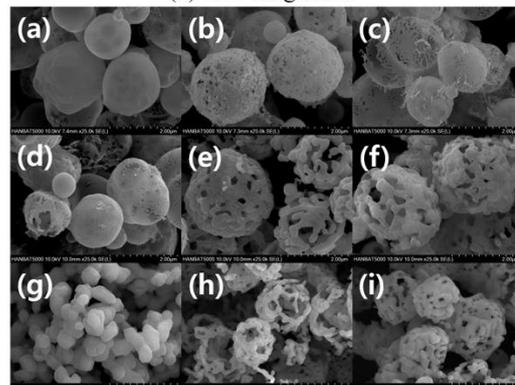
air was used as a carrier gas. The furnace temperature was 980 °C, and the flow rate was 20 l/min.

2.2 Ca compounds from  $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$  (Calcium nitrate) 0.6 M,  $(\text{CH}_2\text{CO}_2\text{H})_2$  (Citric acid) 0.4 M, and  $(\text{CH}_2\text{OH})_2$  (Ethylene glycol) 0.4 M.

Through the aerosol process, small primary  $\text{CaCO}_3$  particles were collected to form sphere-shaped secondary  $\text{CaCO}_3$  particles. Fig. 2 shows SEM images (low magnification, high magnification) of heat-treated samples of Ca compounds at 1 L/min air conditions for 3 hours. From 500 °C (Fig. 2(e)), voids are formed. It can be seen that as the carbon component is removed, secondary particles are collapsed to form primary particles.



(1) Low magnitude



(2) High magnitude

Fig. 2. SEM photographs of Ca compounds using calcium nitrate : (a) as-prepared (b) 200 °C, (c) 300 °C, (d) 400 °C, (e) 500 °C, (f) 600 °C, (g) 700 °C, (h) 800 °C, (i) 900 °C

The conversion of  $\text{CaCO}_3$  to  $\text{CaO}$  was confirmed by SEM-EDS results (Fig. 3). As the heat treatment temperature increases, it can be seen that the carbon component (red) decreases, and the calcium component (green) relatively increases accordingly.

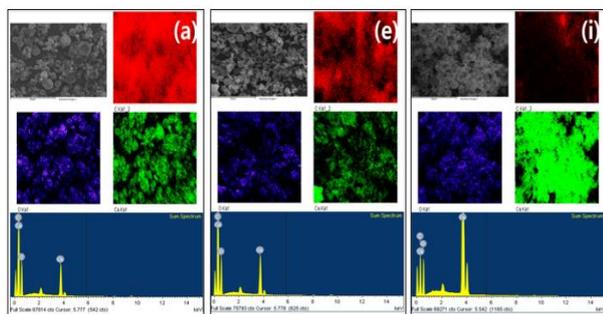


Fig. 3. SEM-EDS images of Ca compounds using calcium nitrate: (a) as-prepared, (e) 500 °C, (i) 900 °C

The XRD results of the heat-treated powder are shown in Fig. 4. The calcite phase disappears from 600°C. It seems that carbons bonded to Ca ions are removed.

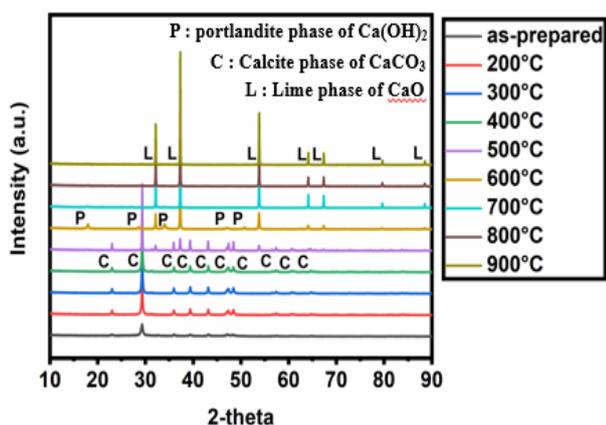


Fig. 4. XRD pattern of heat-treated Ca compounds using Calcium nitrate

### 2.3 Ca compounds from Calcium acetate ( $\text{Ca}(\text{CH}_3\text{COO})_2 \cdot \text{H}_2\text{O}$ ) 0.5 M.

The SEM-EDS image of the powder using Calcium acetate as a precursor is shown in Fig. 5. Removal of carbon and phase change according to the time of heat treatment at 1L/min air condition were observed. As-prepared samples are spherical, but as heat treatment proceeds, these spheres partially collapse.

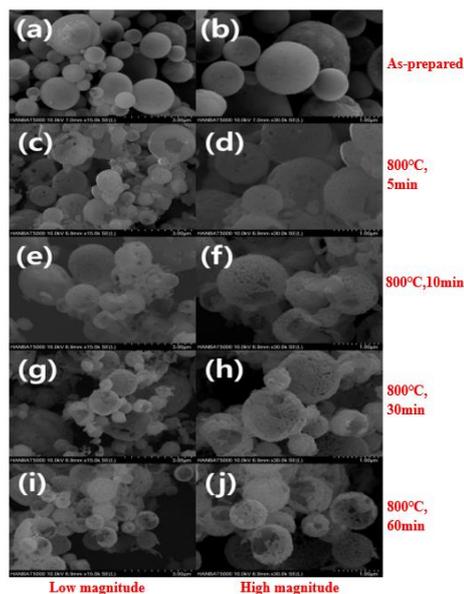


Fig. 5. SEM-EDS images of Ca compounds using calcium acetate

## 3. Conclusions

In this study, an aerosol process was introduced to immobilize carbon into a stable structure. Effects of precursor composition and heat-treatment conditions on the powder were evaluated. As a result of the study, round-shaped  $\text{CaCO}_3$  secondary particles were converted into  $\text{CaO}$  through heat treatment. It is expected that it will be possible to make  $\text{CaCO}_3$  with C-14 by absorbing  $\text{CO}_2$  generated from PHWR to  $\text{CaO}$  with pores. The results of this study would be applied to the process treating C-14.

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