Asian Journal of Chemistry Vol. 21, No. 4 (2009), 3117-3132

Drying of Sodium Bicarbonate with Microwave Energy

ABDULLAH SEYRANKAYA
Department of Mining Engineering, Faculty of Engineering
Dokuz Eylul University, Division of Mineral Processing, Buca, Izmir, Turkey
Fax: (90)((232)4530868; Tel: (90)(232)4127519; E-mail: a.seyrankaya@deu.edu.tr

Drying of sodium bicarbonate ($NaHCO_3$) with microwave energy was investigated in this study. In industrial applications, sodium bicarbonate is dried in steam tube rotary or fluidized bed dryers by heating with air below 60 ºC. However, drying in rotary dryers is slow due to limitations in mixing and low gas temperatures. Higher temperatures are not permitted due to possible degradation of the product. Fluidized bed dryers cause dusting and powder formation as a result of attrition caused by collision of particles in the bed. Using microwave energy, sodium bicarbonate powder containing 7 and 15 % moisture was dried using microwave powers ranging from 136 to 800 W and 2450 MHz frequency at different layers of sample thicknesses. Sodium bicarbonate samples with 7 % initial moisture and about 2 mm sample thickness were dried successfully at all powers with no decomposition or disintegration of the crystalline molecules. Decomposition and caking of the product started to occur at 15 % initial moisture content and increased sample bed thicknesses (> 2 mm) at all power levels used.

Key Words: Sodium bicarbonate, Drying, Microwaves, Mineral processing.

INTRODUCTION

Sodium bicarbonate, $NaHCO_3$ or baking soda, is widely used in chemical and pharmaceutical industries, in animal feeds, in human food and in other industrial and manufactured products[1,2]. Sodium bicarbonate is refined in 3 different ways: (a) from the mineral nahcolite (referred to as natural sodium bicarbonate), (b) from the mineral trona or naturally occurring sodium-carbonate-bearing brines (both referred to as natural soda ash) and (c) manufactured with using Solvay process which requires common salt and limestone (referred to as synthetic sodium bicarbonate)[3].

A simplified flowsheet of the sodium bicarbonate production process from soda ash solution is presented in Fig. 1. The soda ash solution is purified by filtration to eliminate impurities. Next step is carbonation, with pure concentrated carbon dioxide and simultaneous cooling to remove the heat of reaction as shown below[4],

$$Na_2CO_3 + CO_2 + H_2O \longrightarrow 2NaHCO_3 \qquad (1)$$

As carbonation proceeds, sodium bicarbonate precipitates and is recovered by centrifuging and drying with hot air.

With increasing heat, sodium bicarbonate decomposes into sodium carbonate, carbon dioxide and water, *i.e.*, reversal of eqn. 1. Fig. 2 presents the dissociation pressures of carbon dioxide at different temperatures. As seen from the figure, appreciable decomposition to sodium carbonate occurs[4] above 60 ºC.

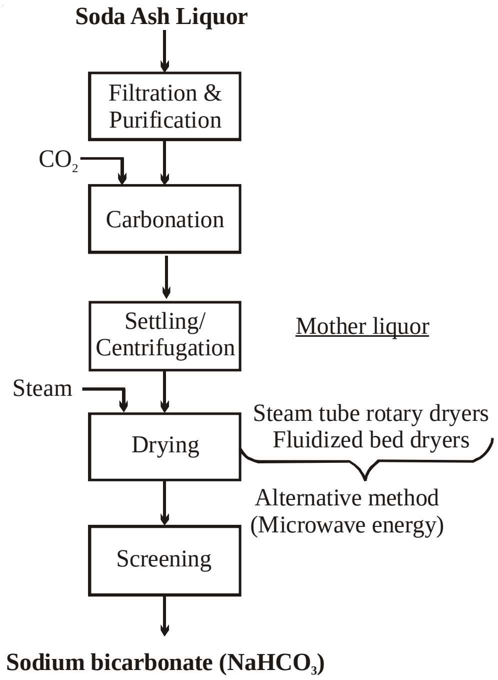

Fig. 1. Simplified flowsheet of the $NaHCO_3$ production process [modified from Ref. 1]

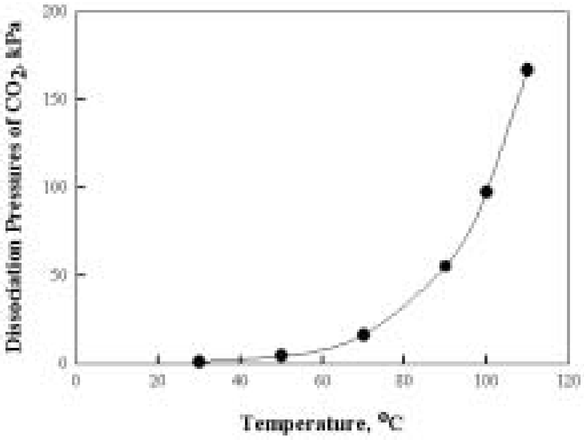

Fig. 2. Dissociation pressures of carbon dioxide at different temperatures [modified from Ref. 4].

In industrial applications, sodium bicarbonate is dried in steam-tube rotary, or fluidized bed, dryers by heating with air below 60 ºC. However, such drying methods have common limitations, that is, the heat transfer from the surface to the inside of the particle is principally by conduction and the process is slow. The maximum temperature of the heated surface, to avoid product degradation, is a limiting factor in the use of conductive heat transfer for drying. In addition, in rotary dryers, mixing is not good and gas temperatures are low. In fluidized bed dryers, the possibility of powder formation and dusting of the product is quite high, as a result of attrition caused by collision of particles[5,6]. To overcome these problems, microwave energy has been considered as an alternative process for drying sodium bicarbonate.

The mechanism of drying with microwave energy is quite different from that of conventional drying. In contrast to conventional dryers, microwave drying eliminates the need to use the surrounding air as a mode of heat transfer to the contained water in the material. With internal heat generation, in microwave systems, mass transfer is primarily due to total pressure gradients established because of the rapid vapour generation within the material. The quick energy absorption by water molecules causes rapid evaporation of water (results in higher drying rates of powder), creating an outward flux of rapidly escaping vapour. Most of the liquid is vapourized before leaving the sample. Ideally, the heat would be transferred only to the water molecules[7,8].

In recent years, microwave energy has found increasing application to the drying of various kinds of materials and products, such as agricultural, chemical, mineral, food, textile, paper and lumber[9-14]. The unique heating mechanisms of microwaves permit dramatic energy savings in many instances, as well as providing competitive benefits[7,15-18].

Microwave drying offers many advantages over conventional heating such as non-contact heating, energy transfer (not heat transfer), rapid heating, material selective heating, volumetric heating, quick start-up and stopping, higher level of safety and automation. In addition, heating is quite fast, so dryer sizes and necessary plant areas are rather small[6,8,17].

In this paper, the results of the experimental work to dry off the free moisture from sodium bicarbonate powder using microwave energy is presented.

EXPERIMENTAL

Drying equipment and procedure: In this study a Premier, PMO-20 microwave oven was used. It has a frequency of 2450 MHz with a maximum output power of 800 W. Drying experiments were conducted at 6 different microwave output powers of 136, 168, 264, 440, 616 and 800 W.

In industry, the suspension of bicarbonate in the mother liquor (Fig. 1) leaves the carbonation stage at 30 ºC and the centrifuged products may contain about 7 % water[4]. In the case of using vacuum filters, the product may contain up to 18 % water[19]. In this study, sodium bicarbonate was wetted with aqueous saturated sodium bicarbonate solution to obtain samples containing 7 and 15 % water. The wet samples were placed in closed containers and preliminary heating to 30 ºC was carried out in a laboratory thermostatic oven to simulate heating under a saturated atmosphere.

Drying experiments were performed with 50.04 ± 0.16 g samples (where ± shows standard error of the mean, SEM). For mass determinations, a digital balance of ± 0.01 g accuracy (Sartorius GP4102-OCE) was used. A teflon line sheet (15 cm diameter), containing a layer of sample of 1-2 mm thickness, was placed on the centre of the turntable inside the microwave cavity during treatment, for even absorption of microwave energy. A new sample was used independently at each microwave output power selected for drying. Each experiment was replicated at least three times.

Simultaneous temperature measurement in a continuous electromagnetic field using a thermocouple is a major problem as the thermocouple-field interaction leads to gross errors[17,20]. To avoid such a problem, the temperature of the irradiated sample was measured by inserting a K-thermocouple (Hanna instruments, model HI 93551) into the sample immediately after turning off the microwave power. The temperature was measured at three different locations in the bed for a duration of fixed time. An average bulk surface temperature of each sample was reported in this study. Although microwaves directly heat the moisture in the samples instead of sodium bicarbonate powder which is transparent to microwaves, it is not possible to conveniently measure the temperature of the water only. It is likely that the moisture in the sample acquired significantly higher 'peak' temperatures especially at the higher output power rates.

The thermocouple was calibrated using a thermometer at different water temperatures. Completion of drying was determined by chemical analysis of the product, *via* determination of its $NaHCO_3$ and Na_2CO_3 content.

The heating profile of the oven conducted with water showed an almost uniform magnetic field. The test was conducted by heating 100 mL water in a beaker and reading its temperature (about 5 s delay), *i.e.,* interrupting the power and reading the temperature.

Particle size analysis of the samples was determined by the Retsch standard sieves using a mechanical sieve shaker (Retsch AS 200 basic) for 0.5 h. Optical analyses were also performed on products placed on a sampling tray by a digital camera connected to an Olympus SZ61 microscope to evaluate the physical changes caused by microwave application at different sample thicknesses.

Batch conventional drying experiments were performed using a laboratory type of indirect rotary tube dryer for a comparison of drying time. The dryer consists of a muffle furnace, a steel cylindrical drying chamber passing through it (inner diameter of 10 cm and a length of 30 cm) and a 1600 W electrical heater with a temperature controller. Three internal flights of 30 cm length and 2 cm width were fixed to carry the samples. During the dryer's operation, the particles were exposed to the drum surface to be heated. The vapourized constituents were removed by a carrier gas carbon dioxide. Before each experiment, the dryer was operated for 0.5 h to obtain a steady temperature reading of 55 ºC. All the experiments were conducted at a rotary dryer speed of 10 min^{-1}, so that the sample could be mixed uniformly. Experiments were conducted with samples containing 7 and 15 % moisture.

Purity analysis of the sodium bicarbonate product: After irradiation of the sodium bicarbonate samples in the microwave oven, about 2 g of sodium bicarbonate powder was taken and dissolved in CO_2 free, double distilled, deionized water. The sample was then transferred into a 100 mL volumetric flask and brought to volume. 10 mL of the solution was placed into a 100 mL beaker and 10 mL of water added. Three drops of phenolphthalein indicator were added to this solution with stirring. If the colour of this solution turned pink, decomposition was considered significant. If the test appeared to be negative, the sodium bicarbonate solution was titrated with a standardized hydrochloric acid solution (0.1 N) using bromocresol green indicator. Titration continued until its colour turned yellow. Finally, the amount of sodium bicarbonate in each sample was calculated.

Determination of moisture and weight of sodium bicarbonate and sodium carbonate: The amount of moisture and decomposition products was determined by acid titration method. If the phenolphthalein test indicated that the amount of carbonate was significant, solution was titrated with the standardized hydrochloric acid until the colour turned colourless. If the phenolphthalein test indicated that the amount of carbonate was negligible, the titration procedure was performed, to accurately determine the sodium bicarbonate content, with standardized hydrochloric acid using bromocresol green indicator. Hydrochloric acid (0.1 N) was always pre-standardized using with pre-dried sodium carbonate. Titration continued until its colour turned yellow. The amount of sodium bicarbonate and sodium carbonate were calculated by acid consumption of the solution. The amount of moisture content was derived from mass balances before and after titration.

RESULTS AND DISCUSSION

Sodium bicarbonate powder containing 7 and 15 % initial moisture was dried in the microwave oven at different energy levels. Drying of the sodium bicarbonate powder is dictated by the rate of moisture removal and the rate of sodium bicarbonate decomposition to sodium carbonate.

Firstly, the drying behaviour of the dry sodium bicarbonate powder was investigated in the microwave oven at 3 power levels of 136, 440 and 800 W. The results of irradiation tests, presented in Fig. 3, showed that the sodium bicarbonate was a poor absorber of the microwave energy for up to 5 min of irradiation. The sample's temperature increased to 38 ± 0.83 ºC during 5 min at the highest power level of 800 W.

Particle size distributions of the crystalline sodium bicarbonate samples before and after microwave heating at 800 W were determined by dry screening and presented in Table-1. The results indicate that the size distributions after microwave

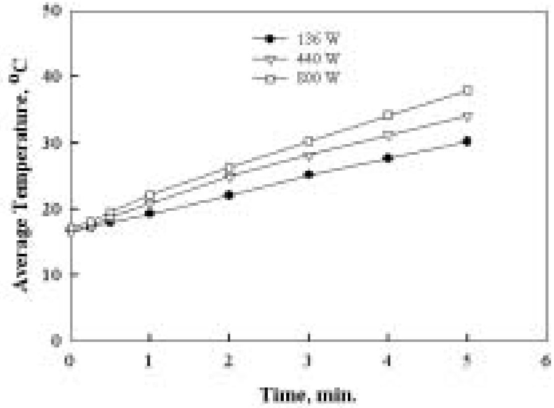

Fig. 3. Heating behaviour of dry sodium bicarbonate in the microwave oven at different power levels

TABLE-1
SIZE DISTRIBUTIONS OF THE SODIUM BICARBONATE POWDER BEFORE AND AFTER MICROWAVE IRRADIATION AT 800 W (INITIAL MOISTURE: 7 %, BED THICKNESS: 2 mm)

Size range (µm)	Weight % [before]	Weight % [after]
+150	1.44	1.95
-150+75	70.32	71.31
-75+45	21.61	21.00
-45	6.63	5.74

irradiation remained the same. Size distributions of the samples with initial 7 % moisture content and 2 mm sample thickness did not change at different power levels selected. Caking started to form, however, with both increasing the initial moisture content and sample thickness.

The results of drying experiments with the sodium bicarbonate samples, containing 7 and 15 % moisture, at 2 mm bed thickness and at different microwave energy levels are presented in Figs. 4-7 and Table-2. Figs. 4 and 6 show the results for the effect of initial moisture content and power level on the microwave heating behaviour of sodium bicarbonate samples. As seen from the Figs. 4 and 6, the rate of microwave energy absorption by the samples was determined by water molecules per volume and power levels of the oven. The heating behaviour is strongly dependent on the moisture content at the same power levels. However, it is strongly dependent on the power level for the same initial moisture content. The sample temperatures increased to a maximum and then their temperatures dropped indicating the removal of the moisture. This phenomenon can be used for easy control of the drying operation. In the last experiment at a microwave power level of 136 W, the temperature did not rise excessively, because microwave energy was not sufficient. As seen from the Figs. 4-7, at high power levels, drying times were reduced substantially.

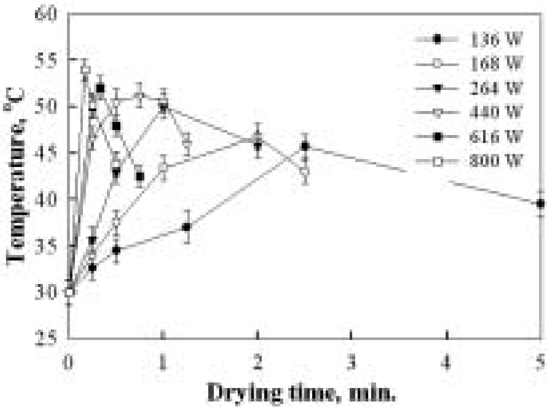

Fig. 4. Drying results for sodium bicarbonate, containing 7 % moisture at different microwave energy levels

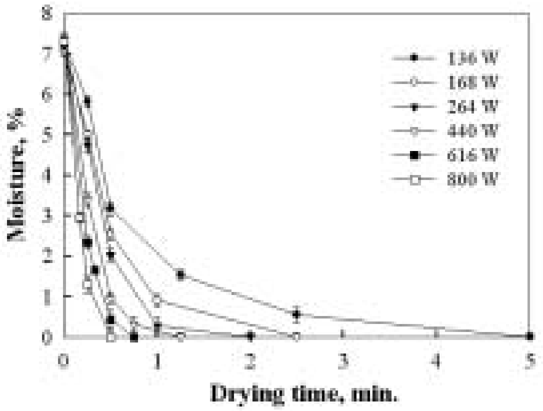

Fig. 5. Reduction in moisture in sodium bicarbonate samples, containing 7 % moisture at different microwave energy levels

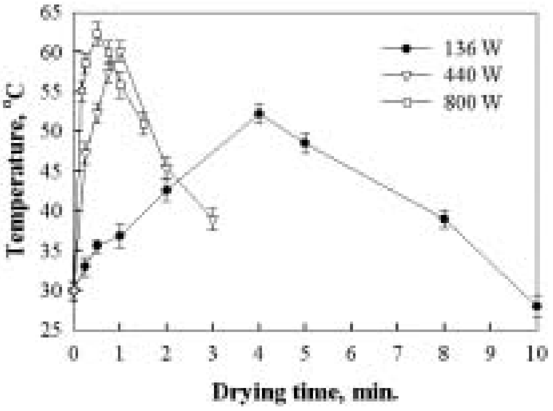

Fig. 6. Drying results for sodium bicarbonate samples, containing 15 % moisture at different microwave energy levels

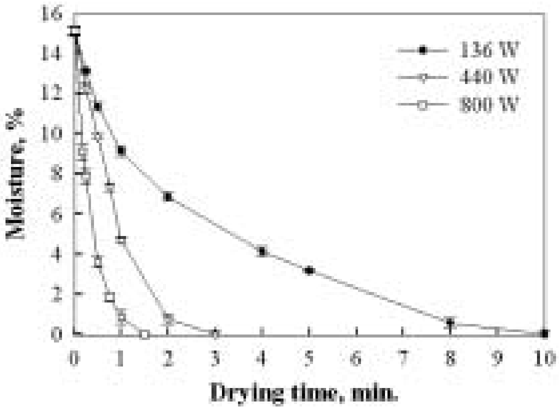

Fig. 7. Reduction in moisture in sodium bicarbonate samples, containing 15 % moisture at different microwave energy levels

TABLE-2
DECOMPOSITION OF $NaHCO_3$, CONTAINING DIFFERENT MOISTURE CONTENTS, DURING MICROWAVE IRRADIATION

Microwave power (W)	% Na_2CO_3	
	@ 7 % moisture	@ 15 % moisture
136	0.0	3.81
440	0.0	2.54
800	0.0	2.23

The tests were repeated at an initial moisture level of 15 % to see the effect of moisture content on the decomposition of the sodium bicarbonate powder. As seen from the Table-2, the sodium bicarbonate decomposed at all three microwave power levels indicating that the water temperatures rose above the decomposition temperatures of the CO_2.

Complete drying of the sodium bicarbonate, containing 7 % moisture, was achieved, without any loss of structural water, *i.e.*, no decomposition, as seen from the Figs. 4 and 5 and Table-2. Mass balance determinations after the titration showed a good correlation between the amount of evaporated water and the chemical composition of the dried mass.

Almost complete drying of sodium bicarbonate was also achieved in the case of 15 % moisture as seen from the Figs. 6 and 7, but structural water was lost as shown in Table-2. The amount of dissociation decreased with increasing microwave power output indicating that the matter dried more rapidly at high energy levels than at the low energy levels, as shown in Table-2. With the lower radiation intensities the solid remains longer at a temperature level of 50 °C, (because the power is sufficient to heat up the water, but not to evaporate it fast). The increased degree of decomposition is the result of longer drying times (*i.e.* more time at the peak-temperatures) with logically more time for the decomposition reaction and CO_2 desorption (Figs. 5 and 7). As the bulk temperature at 15 % is higher than at 7 % moisture, the peak temperature at higher moisture levels might not only have lasted longer, but also might have been higher than at the lower moisture content-accelerating the decomposition reaction.

Kuu *et al.*[21] reported that the solubility of CO_2 in the aqueous moisture layer is lower at higher temperature. Thus, the mass transfer rate of CO_2 increases, resulting in higher hydrolysis and decomposition rates of bicarbonate powder. They proposed the following decomposition schemes:

$$NaHCO_{3(s)} \rightleftharpoons NaHCO_{3(aq)} \quad (2)$$

$$2NaHCO_{3(aq)} \rightleftharpoons 2Na^+ + 2HCO_3^- \quad (3)$$

$$HCO_3^- \rightleftharpoons H^+ + CO_3^{2-} \quad (4)$$

$$HCO_3^- + H^+ \rightleftharpoons H_2CO_3 \quad (5)$$

$$H_2CO_3 \rightleftharpoons H_2O_{(aq)} + CO_{2(aq)} \quad (6)$$

$$CO_2\,(aq) \rightleftharpoons CO_{2(g)} \quad (7)$$

The chemical reaction mechanism (eqns. 4 to 6) is rather unlikely. Since bicarbonate creates a basic solution (especially in adsorbed water), it is more likely, that HCO_3^- decomposes directly to CO_2 and OH^-, as found by Himmelblau and Babb[22]. They determined the kinetic reaction rate constants in the CO_2 - $NaHCO_3$ - Na_2CO_3 - H_2O system at various temperatures and pH values by a radioactive tracer technique. Thus, eqns. 8 and 9[22,23] are more likely to characterize the reaction kinetics and mechanisms rather than eqns. 4 to 6. As the adhering moisture is saturated with $NaHCO_3$, it is alkaline and a few free H^+-ions are available for acid-base neutralization.

$$HCO_3^-{}_{(aq)} \rightleftharpoons OH^-{}_{(aq)} + CO_{2(aq)} \quad (8)$$

$$OH^-{}_{(aq)} + HCO_3^-{}_{(aq)} \rightleftharpoons H_2O_{(aq)} + CO_3^{2-}{}_{(aq)} \quad (9)$$

Eqns. 2 and 7 are physical processes, whereas eqns. 3-6 or eqns. 3, 8 and 9 are chemical equilibrium equations established in the aqueous phase. The overall chemical reaction for both mechanisms, from eqns. 3-6 or eqns. 3, 8 and 9, becomes:

$$2NaHCO_{3(aq)} \rightleftharpoons 2Na^+ + CO_3^{2-} + CO_2 + H_2O \quad (10)$$

which is similar to eqn. 1. It can be concluded that the rate of weight change of the bicarbonate powder depends on the rate of moisture sorption rate and the rate of losing carbon dioxide and water molecules.

The effect of bed thickness on the drying process was also investigated and the results are presented in Figs. 8-11. In general, drying takes longer with increasing bed thickness, because water is removed more slowly. It is also obvious that a material with a large initial moisture content has a much longer drying time (Fig. 10). Figs. 9 and 11 show the temperature profiles measured at various times for different sample thicknesses and initial moisture content. Higher bulk temperatures were observed with increasing bed thicknesses. In contrast to that in conventional drying, microwave drying produces higher temperatures inside the drying sample while

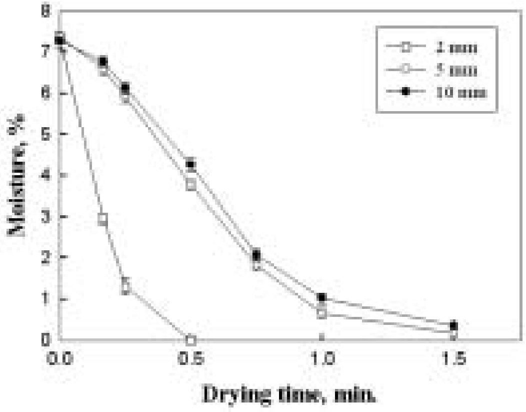

Fig. 8. Reduction in moisture in sodium bicarbonate at different bed thicknesses, containing 7 % moisture at 800 W power level

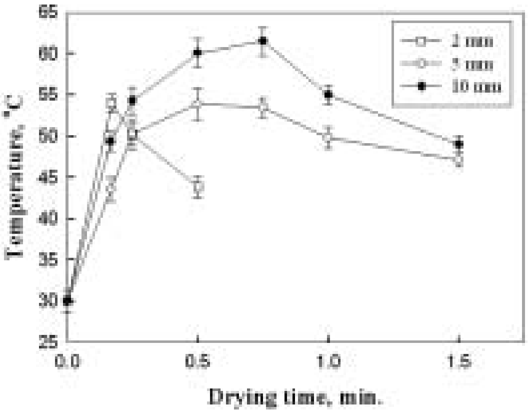

Fig. 9. Drying results for sodium bicarbonate at different bed thicknesses, containing 7 % moisture at 800 W power level

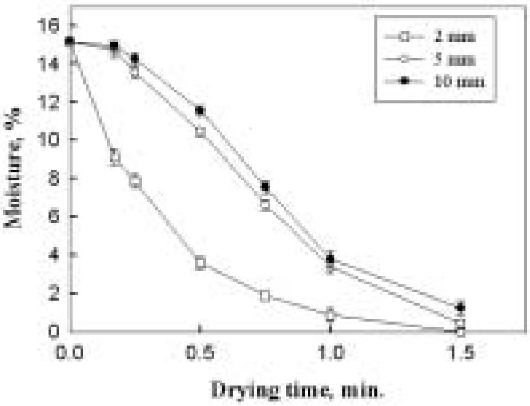

Fig. 10. Reduction in moisture in sodium bicarbonate at different bed thicknesses, containing 15 % moisture at 800 W power level

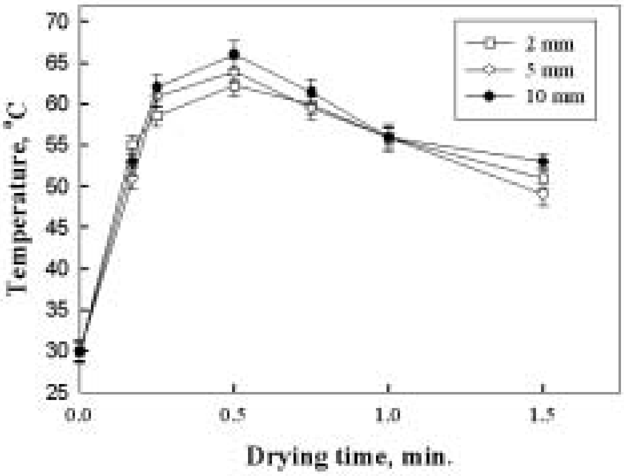

Fig. 11. Drying results for sodium bicarbonate at different bed thicknesses, containing 15 % moisture at 800 W power level

the surface temperature stays colder due to the cooling effect of surrounding air. At the same time the evaporation occurs at the surface of the sample at a lower temperature due to evaporative cooling. It can be seen in Figs. 9 and 11 that the temperature profiles of the sample rise up sharply. Later, the temperature starts to drop continuously since the rate of microwave absorption is lower after a majority of moisture content is removed from sodium bicarbonate samples. Similar trends were observed at other microwave power levels.

Drying times of samples in the microwave and conventional rotary drying ovens were compared in Table-3. As seen from the table, microwave oven power levels affect the drying times substantially. At 7 % initial sample moisture, drying the sample, at 136 W in the microwave oven took 5 min and this reduced to 0.5 min at

TABLE-3
COMPARISON OF DRYING TIMES WITH MICROWAVE OVEN AND INDIRECT ROTARY DRYER (BED THICKNESS OF 2 mm FOR MICROWAVE DRYING)

Drying method		Final drying time, (min.)	
MW	Rotary drying	@ 7 %	@ 15 %
Power level (W)	Temp. (°C)	Initial moisture content	Initial moisture content
136*§		5	10
440*§		1.25	3
800*§		0.5	1.5
	50 ± 2	30	-
	55 ± 2	27	45

*For microwave temperature readings, see Figs. 4 and 6.

§Bed thickness of 2 mm for microwave drying.

800 W. Similar trends but longer drying time were observed at 15 % initial sample moisture dried in the microwave oven. In general, drying times for samples are substantially higher in the rotary dryer than in microwave oven. Drying time were shortened by about 90 % for samples irradiated at 800 W instead of 136 W at conditions of 2 mm sample thickness, 7 and 15 % initial moistures. Microwave drying technique can greatly reduce the drying time of sodium bicarbonate compared to conventional drying (Fig. 12 and Table-3). Conventional drying at 50 and 55 ºC shortened the drying time by about 60 fold at 800 W, 7 % initial moisture and 2 mm bed thickness.

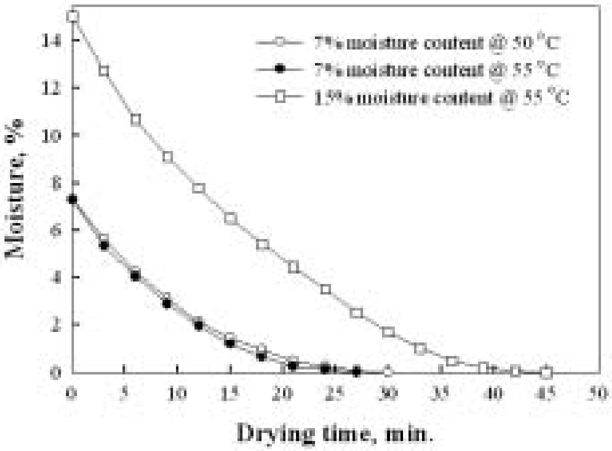

Fig. 12. Reduction in moisture in sodium bicarbonate in the rotary dryer at different temperatures

Optical microscopic studies were carried out to investigate the visual changes at the surface of the sodium bicarbonate powder. The pictures taken were presented in Fig. 13a-b. There were no visual changes in the samples of 7 % moisture. When the sample thickness increased from 2 mm to 5 and 10 mm, caking of the particles occurred, *i.e.* adhering between the crystals (Fig. 13c-e).

The caking of the sodium bicarbonate with increasing moisture content and bed thickness is expected, because the adhering moisture is saturated with sodium bicarbonate. With a solubility of 9.8 %-wt at room temperature (and 13 %-wt at 50 °C), there is 1.5 to 2 %-wt dissolved $NaHCO_3$ present-which will precipitate on the $NaHCO_3$-crystals, when the water is removed during drying. The higher the packing thickness, the more slowly the water is removed, giving the precipitating $NaHCO_3$ more time to form larger crystallites and bridges between the existing crystals. Caking was also seen with the sodium bicarbonate samples containing 15 % moisture, in samples of thickness as low as 2 mm.

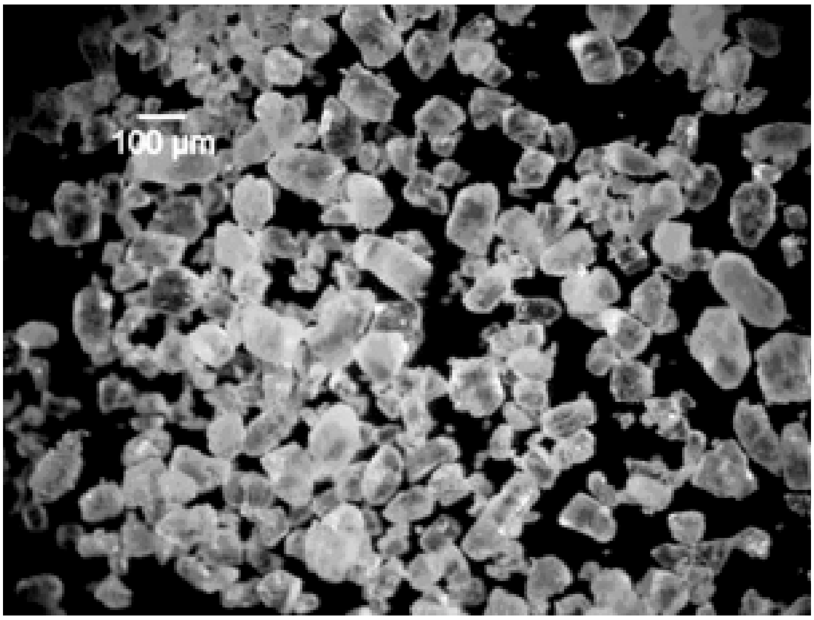

Fig. 13a. Optical micrographs of the original sodium bicarbonate

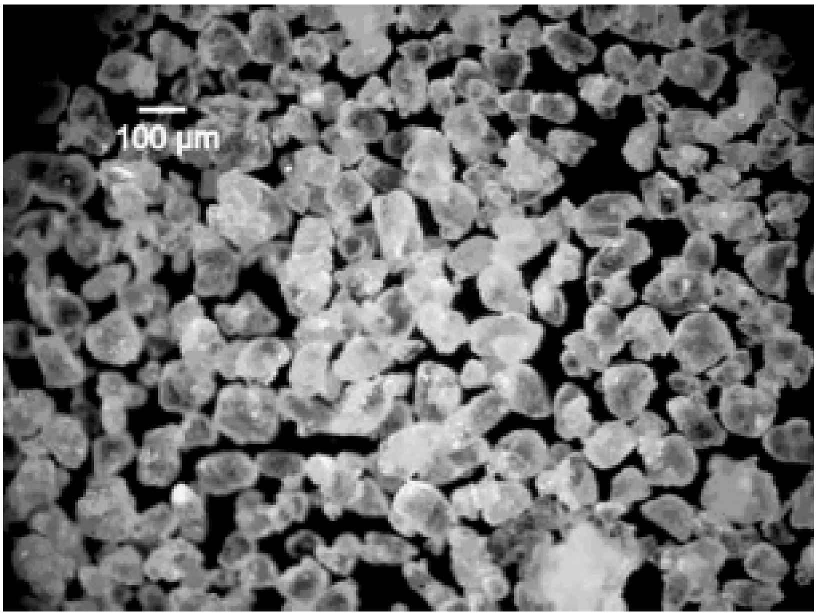

Fig. 13b. Optical micrographs of the sodium bicarbonate, after 70 s microwave irradiation (Initial moisture: 7 %, bed thickness: 2 mm, microwave power: 440 W)

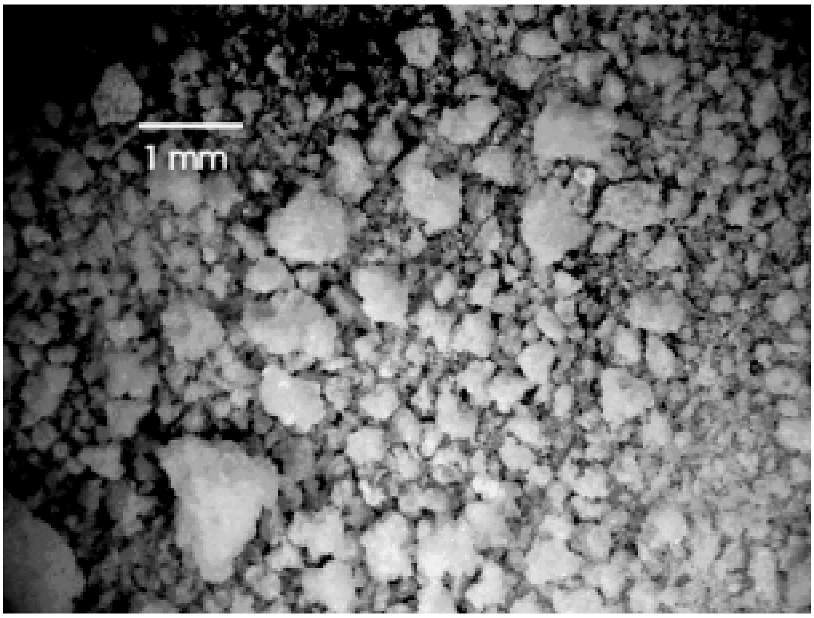

Fig. 13c. Optical micrographs of the sodium bicarbonate, after 70 s microwave irradiation (Initial moisture: 7 %, bed thickness: 5 mm, microwave power: 440 W)

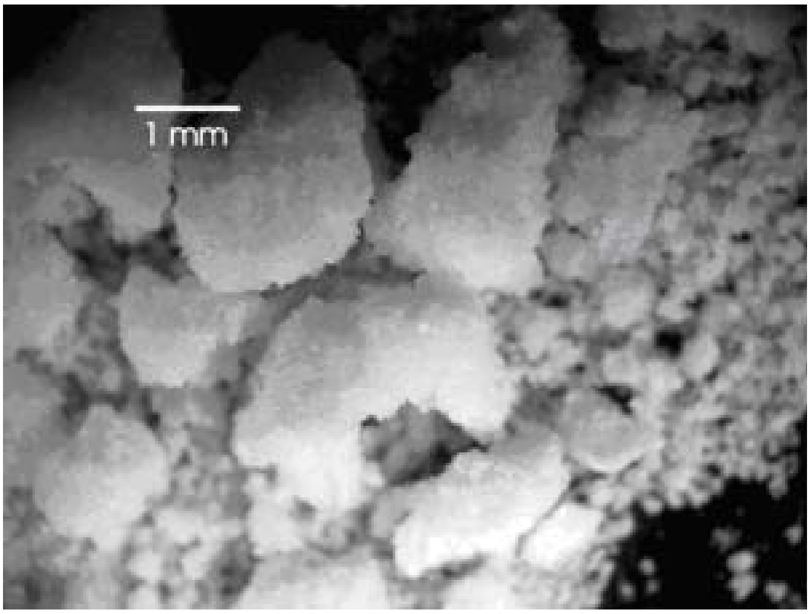

Fig. 13d. Optical micrographs of the sodium bicarbonate, showing caking after 70 s microwave irradiation. (Initial moisture: 7 %, bed thickness: 10 mm, microwave power: 440 W)

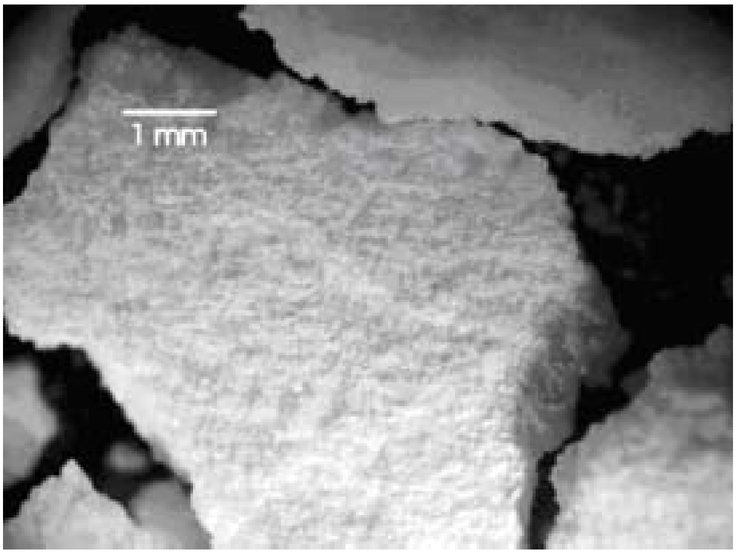

Fig. 13e. Optical micrographs of the sodium bicarbonate, showing caking after 70 s microwave irradiation. (Initial moisture: 15 %, bed thickness: 2 mm, microwave power: 440 W)

Conclusion

In this investigation, sodium bicarbonate powder containing 7 and 15 % moisture was dried using microwave powers ranging from 136 to 800 W at 2450 MHz frequency. The results show that the sodium bicarbonate ($NaHCO_3$) powder can be successfully dried using microwave energy to remove the excess moisture from the sodium bicarbonate powder. Sodium bicarbonate samples with 7 % initial moisture and about 2 mm sample thickness were dried successfully at all powers with no decomposition or disintegration of the crystalline molecules. The results of particle size analysis showed that the average particle size and distribution remain the same and dusting does not occur in the case of 2 mm sample bed.

Decomposition and caking of the product started to occur at 15 % initial moisture content and increased sample bed thicknesses (> 2 mm) at all power levels used.

At higher moisture contents precautions need to be taken during drying of the samples to prevent decomposition and caking. For example, use of shorter heating intervals and decreased sample thickness is recommended.

REFERENCES

1. D.E. Garrett, Natural Soda Ash-Occurrences, Processing and Use, Van Nostrand Reinhold, New York (1992).
2. H. Haynes, *In Situ*, **21**, p. 357 (1997).
3. D.S. Kostick, Soda Ash: U.S. Geological Survey, Mineral Commodity Summaries, pp. 152-153 (January 2007); Available: http://minerals.usgs.gov/minerals/pubs/commodity/soda_ash/sodaamcs07.pdf

4. C. Thieme, Sodium Carbonates, Ullmann's Encyclopedia of Industrial Chemistry, Vol. A24, pp. 299-316 (1992).
5. P.L. Jones, Drying'92, Pt. A., A.S. Mujumdar, ed., Elsevier, Amsterdam, pp. 114-136 (1992).
6. S. Kocakusak, H.J. Koroglu and R. Tolun, *Chem. Eng. Process.*, **37**, 197 (1998).
7. M. Maskan, *J. Food Eng.*, **44**, 71 (2000).
8. R.F. Schiffmann, in ed.: A.S. Mujumdar, Handbook of Industrial Drying, Marker Dekker Inc., New York, Vol. 1, edn. 2, pp. 345-372 (1995).
9. D.A. Jones, S.D. Lelyveld, S.D. Mavrofidis, S.W. Kingman and N.J. Miles, *Resour. Conserv. Recycl.*, **34**, 75 (2002).
10. M.A.M. Khraisheh, T.J.R. Cooper and T.R.A. Magee, *J. Food Eng.*, **33**, 207 (1997).
11. S.W. Kingman, W. Vorster and N.A. Rowson, *Min. Eng.*, **13**, 313 (2000).
12. S.W. Kingman, K. Jackson, A. Cumbane, S.M. Bradshaw, N.A. Rowson and R. Greenwood, *Int. J. Min. Process.*, **74**, 71 (2004).
13. C.Y. Cha and C.T. Carlisle, US Patent No: US 6,046,376 (2000).
14. A. Seyrankaya and B. Ozalp, *Thermochim. Acta*, **448**, 31 (2006).
15. W.C. McCrone, Kirk-Othmer Encyclopedia of Chemical Technology, Vol. 16, pp. 672-700 (1995).
16. S.W. Kingman and N.A. Rowson, *Min. Eng.*, **11**, 1081 (1998).
17. K.E. Haque, *Int. J. Min. Process.*, **57**, 1 (1999).
18. M.R. Sanio and P.S. Schmidt, *Mat. Res. Soc. Symp.*, **124**, 337 (1988).
19. E. Koksal, Trona and Synthetic Soda Ash Symposium, TMMOB, Chamber of Mining Engineers, Ankara, Turkey, pp. 73-77 (2002) (in Turkish).
20. P.D. Ramesh, D. Brandon and L. Schächter, *Mater. Sci. Eng.*, **A266**, 211 (1999).
21. W. Kuu, R. Chilamkurti and C. Chen, *Int. J. Pharmaceut.*, **166**, 167 (1998).
22. D.M. Himmelblau and A.L. Babb, *AIChE J.*, **4**, 143 (1958).
23. M.C. Ball, C.M. Snelling, A.N. Strachan and R.M. Strachan, *J. Chem. Soc., Faraday Trans. I*, **82**, 3709 (1986).

(*Received*: 19 July 2008; *Accepted*: 21 January 2009) AJC-7167