Carbonation of Sodium Disilicate During Spray-drying

A. Horvat, J. Golob[*], and R. Gabrovšek[**]

Silkem d.o.o., Kidričevo, Slovenia
[*]University of Ljubljana, Ljubljana, Slovenia.
[**]National Institute of Chemistry, Ljubljana, Slovenia

In memory Prof. Emeritus Vera Johanides

Although drying is generally considered as a separation process, the present contribution reports on drying connected with chemical reaction. During spray-drying of sodium disilicate by the help of directly heated air it was found that the presence of CO_2 influenced the quality of the product, expressed by the percent of insoluble content. Based on the product and process analysis, equipment was modified to avoid too long exposure of sodium disilicate to CO_2 atmosphere. During the shortened exposure time CO_2, transfer from gas to solid was limited to surface chemosorption, and CO_2 solid diffusion was practically eliminated. As a result a commercial product with low insoluble content was obtained.

Key words:
Spray drying, sodium silicate, carbonation, waterglass, carbon dioxide

Introduction

The most frequent product of the dried waterglass family is sodium disilicate $Na_2O \cdot 2SiO_2$, commercially known as NADIS. In the production of dried waterglass both, the synthesis of alkaline and silicate components, as well as drying of colloid solution, are included to obtain a product with specific properties[1]. The analyses of NADIS produced in a directly heated spray dryer have shown extremely different insoluble content of the product, caused by different CO_2 fraction.

The literature review has shown that the mechanism of liquid waterglass carbonation is well known[2]: namely, the reaction with CO_2 represents the basis for the solidification of foundry sand and for the precipitation of SiO_2. However, in relevant literature only the process of the carbonation of waterglass-based coatings is presented[3], while the reaction of waterglass with CO_2 during spray-drying has not been described. It may be expected that the exposure of NADIS to CO_2 atmosphere may damage the product during the drying process. Materials with bulk densities of 100 kg m^{-3} or less, such as NADIS, are rather rare in the industrial practice, because they cause a variety of technical problems and obstacles, particularly with regard to separation processes. There is little evidence of a successful experience in handling and drying of light and ultralight powdery materials. Very high investment costs for the adaptation of the industrial drying plant with a direct heating system to a plant with an indirect system to eliminate CO_2 influence on NADIS properties during drying process, resulted in the search for a suitable technical solution of the existing plant.

The aim of our research program was to find proper conditions for achieving standard quality of the product, obtained by drying process with directly heated air. For this purpose the research was focused on: (i) finding quantitative relation between the CO_2 content and insoluble content of NADIS, (ii) analyzing the product at relevant positions of a dryer and gas/solid separator, and (iii) proposing economically and technically acceptable solutions to produce NADIS with a minimum possible content of insoluble substance.

Relation between CO_2 content and NADIS quality

Literature[3] shows that waterglass reacts with CO_2 from the air and that white powder consists of Na_2CO_3 and precipitated SiO_2 forms. The reaction can be written as:

$$Na_2O \cdot 2SiO_2 + CO_2 \Rightarrow Na_2CO_3 + 2\ SiO_2$$
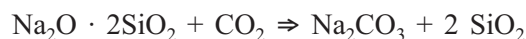
This reaction is the basis for the solidification of foundry sands by CO_2 process. This process also takes place under atmospheric conditions where the presence of CO_2 causes the carbonation of waterglass. However, if carbonation time is considered, the two processes are not comparable: a few minutes under CO_2 versus a few weeks at carbonation

under atmospheric conditions. Assuming that CO_2 reacts completely with the available Na_2O in NADIS, which contains 15% of crystal water, one may find maximum $w = 16.5$ % of CO_2 fraction.

In waterglass SiO_2 is coordinated by Na^+ and OH^- provided that metastable equilibrium conditions exist[1]. In the process of carbonation this equilibrium is interrupted due to a greater affinity of Na_2O to CO_2 than to SiO_2. Carbonation causes steric changes, thus enabling the formation of polysilicate units insoluble in water.

It was found out that during drying of waterglass the quality of the dried product, characterized as insoluble residue, varied greatly. To establish the appropriate quality of the dried product, research was carried out[4] in order to explain the changes of NADIS quality. NADIS was exposed to CO_2 atmosphere ($w = 1.3$ % of CO_2 in air) at 5 different times, the shortest being 2 minutes. The CO_2 content and the insoluble residue were measured. The CO_2 fraction was determined according to ISO 1691 (removal of CO_2 by HCl), while the insoluble residue was determined by the using a method commonly used in the industry of washing powders (5g of sample + 500 g of water (20 °C); 10-minute mixing, filtering through black ribbon filter paper, drying and weighing the residue). Figure 1 presents the CO_2 fraction and the mass percent of the insoluble residue in a dried product.
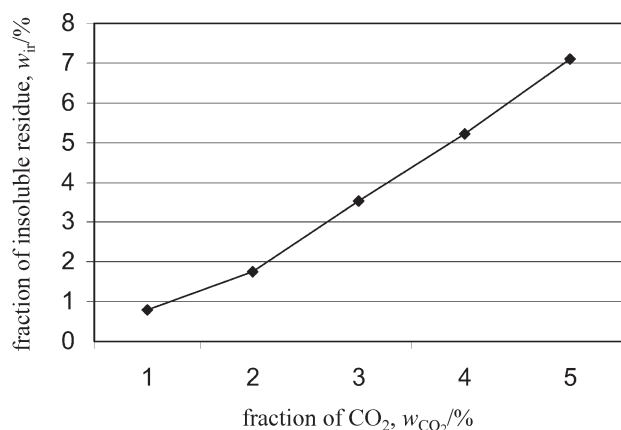

F i g . 1 – *Relation between CO_2 fraction and insoluble residue in NADIS*

It would be necessary to study the kinetics of carbonation separately, however, the present research was limited only to studying the conditions during the spray-drying process which might be relevant for obtaining the expected product quality.

Industrial process

For drying of NADIS a 125 m³ volume spray-dryer with a 24-hole disk with 120 mm diameter was used. Because of the tendency of NADIS to agglomerate at temperatures above 60 °C, it was found necessary to cool down the material immediately after drying. This was carried out during the pneumatic transport. The separation of hot material from the hot air flow was conducted from the drying chamber (I), cyclone, and the bag filter (III), as shown in Figure 2. The separation of cooled product that was suitable for the packing was performed from the transport filter (II). In the process of waterglass drying, products with different bulk densities are formed. This depends on the drying conditions, because of the so-called "ballooning effect" (5, pp. 266). Drying quantities were kept constant during experiments in order to avoid possible changes in the bulk density of material.

It must be borne in mind that the drying of waterglass represents a rather complicated task because of the tendency of the dried material to stick to the walls of the drying chamber, causing the product to form lumps and agglomerates. The reproducibility of the results of bulk density analyses is further hindered by irregular particle shapes.
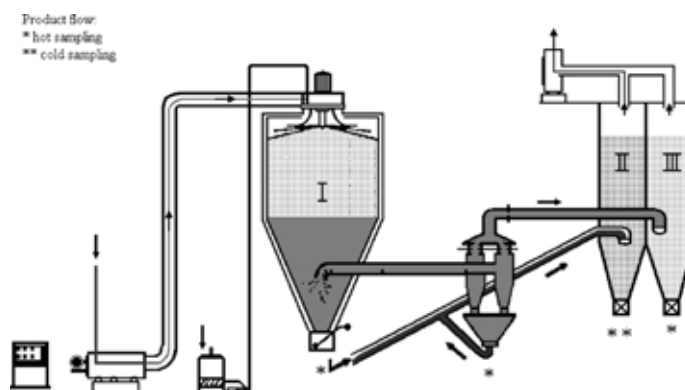

F i g . 2 – *The process of waterglass spray-drying*

The experimental work was focused on determining the degree of carbonation of a dried product at different positions of the drying process. Dried material was collected from the drying chamber-I, the cyclone, the transport filter-II and the bag filter-III, as shown in Figure 2. The solution of sodium silicate, type NaVS48 with a modulus of 2 and with solids concentration of 45 %, was used as a feed. Ambient temperature was 25 °C, targeted material bulk density of material 100 kg m⁻³ and the drying 250 kg h⁻¹. The flow rate of methane used was 22 kg h⁻¹ and the measured fraction of CO_2 in the dryer at the exhaust pipe was 1.2 %, while the calculated fraction of CO_2 in the drying atmosphere was 1.3 %. The difference of 0.1 % of CO_2 between both values was found to be adsorbed by dried material.

T a b l e 1 – *Analyses of NADIS from different positions of the dryer prior to changes of the process system*

No. of sample	Location of sample taken	Fraction of sample population, X / %	Bulk density ρ / kg m^{-3}	Fraction of CO_2 w / %	Fraction of insoluble residue w_r / %
1	Under drying chamber-I	80	105	0.6	0.7
2	Under cyclone-I	15	65	1.8	1.2
3	Filter-II, continuos operation	93	100	0.8	0.6
4	Filter-II, rest	2	24	4.6	8.4
5	Filter-III, continuos operation	4	44	7.7	27.6
6	Filter-III, rest	1	32	13.2	55.5

Other relevant drying quantities:

T_{inlet} = 400 °C
T_{outlet} = 135 °C
Air flow rate through drying chamber Q = 5,400 m^3 h^{-1} at 135 °C or q = 4,600 kg h^{-1}
Atomizer speed = 15,000 rev min^{-1}

Table 1 shows that the fraction of CO_2 greatly depends on the location of the sample and on residence time: the difference between a sample taken during a continuous removal and a sample taken after the completion of the drying process. The material with the shortest residence time in the dryer has the highest bulk density, the lowest CO_2 fraction, and the smallest surface area per gram (Sample 1–200 μm particles). It is well known that the Gaussian particle size distribution of dried materials spans over a wide area, especially when a rotating disk is used. Due to a range of diameters of falling particles, different extent of "ballooning" was noticed which resulted in different bulk densities of the particles formed. Thus the particles possessed different specific surface areas and consequently showed different reactivity which form the basis for the chemisorption of CO_2 onto NADIS. The final consequence is a different insoluble content of the product which indicates uneven product quality.

After the drying process, an amount of 2,000 kg of the product with bulk density of 100 kg m^{-3} and with CO_2 fraction between 0.7 and 2 % was obtained. Additionally, there was 25 kg of material from both filters with a bulk density between 25 and 40 kg m^{-3} and with CO_2 fraction in excess of 5 %. The CO_2 fraction 13.2 % of the sample taken from hot filter (III) indicates intensive carbonation.

Process equipment modification

Based on the analysis of experimental results presented in Table 1 and on industrial run it was found that in order to minimize CO_2 adsorption a shorter residence time in the dryer is necessary.

Changes on the industrial drying system were done by rebuilding the filters with two independent continuously "hot" and "cold" operating filters (II and III). After improving the system, the filters were adapted to operation periods of 2 minutes for sucking, and 2 minutes for emptying a chamber. While one chamber is emptying the product, the other is filtering it. After two minutes the filters take an alternate role. The chambers operate simultaneously by a hydraulically-driven flap in 4-minute cycles for each phase. This method provides mixing of the material from both filters, and the material is left in CO_2 atmosphere for maximum 4 minutes. Figure 3 shows the difference between the old and the improved filter system.
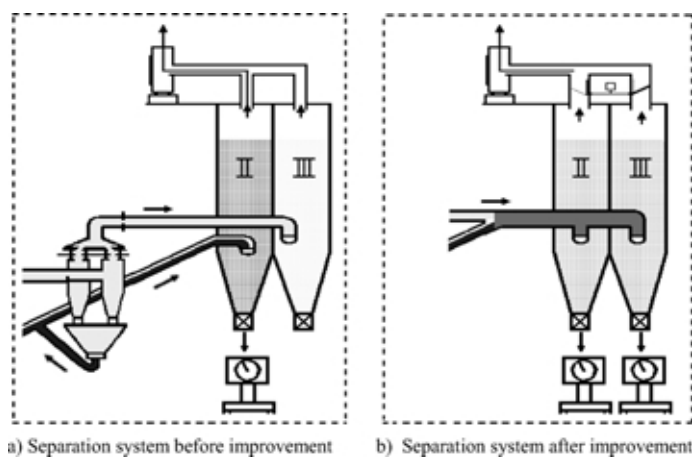

a) Separation system before improvement   b) Separation system after improvement

F i g . 3 – *Basic and improved filter system*

Improved system does not need any cyclone separators, just two filters (Fig. 3b). Some experiments have been carried out using the same drying conditions as prior to adaptation. It was found out that the depositing of materials has not occurred at any other location except the two filters where the sampling was performed. In Table 2 the characteristics of the products after the change of the process are presented.

T a b l e 2 – *Analyses of the NADIS sample from different dryer position after changes in the system*

No. of sample	Location of sample taken	Fraction of sample population, X / %	Bulk density ρ / kg m^{-3}	Fraction of CO_2 w / %	Fraction of insoluble residue w_r / %
7	Filter-II, continuos removal	50	100	0.7–1.2	0.6–0.9
8	Filter-III, continuos removal	50	100	0.7–1.2	0.6–0.9

Conclusion

The improvement of the process allowed significant decrease of exposure time of dried NADIS particles to CO_2 atmosfere. After rebuilding the filter we got a product with CO_2 fraction between 0.6 and 0.9 %, without any further residue on the filter, so the quality of the product increased with a yield of 99.5 %, compared with a previous yield of 93 %. The fraction of insoluble residue after this improvement was kept below 1 %.

We suggest a different process of separation on filters by using two parallel filters. One filter can be switched alternately in the process, while the other filter is emptied, without the use of underpressure. With this filter, the removal of material is possible in short time intervals (less than 4 minutes) which consequently prevents any further carbonation. As a result the product quality and the yield are increased, while undesirable deposition of material is avoided.

List of symbols

Q – volume flow rate, m^3 h^{-1}

q – mass flow rate, kg h^{-1}

T – temperature, °C

w – mass fraction, %

X – number fraction, %

ρ – density, kg m^{-3}

Literature

1. *Vail J. G.,* Soluble Silicates, Reinhold Publishing Corporation, New York, 1960.

2. *Iller R. K.,* The Chemistry of Silica, John Wiley &Sons, Inc., New York, 1979.

3. *Veinot D. E.,* Efflorescence of Soluble Silicate Coatings, Journal of Non Crystalline Solids, 1991, 127, pp. 221–226,

4. *Horvat A.,* Research of CO_2 Influence on Dried Sodium Disilicate Characteristic, Master thesis, University of Ljubljana, 1998.

5. *Masters K.,* Spray Drying Handbook, George Godwin Limited, London, 1985.