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Production of Sodium Silicate Cullets by Using Trona

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This study focuses on production and characterization of sodium silicate cullet by using trona. Sodium silicate cullets are generally produced by direct fusion of precisely measured portions of soda ash (Na_2CO_3) and quartz (SiO_2). In this study trona ($Na_2CO_3 \cdot NaHCO_3 \cdot 2H_2O$) was used as a source of soda ash. For this purpose trona sample was first characterized by X-ray diffraction, scanning electron microscopy, differential thermal analysis/thermal gravimetry and chemical analyses. Different batches were prepared and fused to produce sodium silicates varying in ratio $Na_2O \cdot nSiO_2$. Produced samples were evaluated by X-ray diffraction and scanning electron microscopy. The results showed that trona can be used instead of soda ash to produce sodium silicate cullets.

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1. Introduction

Trona is a complex salt of sodium carbonate and sodium bicarbonate [1] and composed of approximately 46% sodium carbonate (Na_2CO_3) and 36% sodium bicarbonate ($NaHCO_3$), which is relatively abundant in nature [2]. Trona is a natural soda ore, and it is one of the main sources of the alkaline solutions. It has the form of $Na_2CO_3 \cdot NaHCO_3 \cdot 2H_2O$ [3]. Sodium silicate is a colorless compound of oxides of sodium and silica. It has a range of chemical formula varying in sodium oxide (Na_2O) and silicon dioxide or silica (SiO_2) contents or ratios. Sodium silicates are produced in the form of a variety of compounds ranging from $Na_2O \cdot 4SiO_2$ to $2Na_2O \cdot SiO_2$ by properly proportioning the reactants. Sodium silicates varying in ratio from $Na_2O \cdot 1.6SiO_2$ to $Na_2O \cdot 4SiO_2$ are known as colloidal silicates. These are sold as 20% to 50% aqueous solutions called water glass [4]. Sodium silicate cullets are produced by the direct fusion of precisely measured portions of pure silica sand (SiO_2) and soda ash (Na_2CO_3) in oil, gas or electrically fired furnaces at temperatures above 1100 °C according to the following reaction [5–7]:

$$Na_2CO_3 + xSiO_2 \rightarrow Na_2O \cdot xSiO_2 + CO_2. \quad (1)$$

Studies in the literature point out that the production of sodium silicates can be performed by using different raw materials. But, production of sodium silicate cullets by using trona has not been studied. To be able to fill this gap in the literature, the present study was carried out to: (i) determine the characteristics of trona and (ii) investigate the production of sodium silicate cullets by using trona.

2. Materials and method

In this research trona sample was obtained from Eti Soda Plant that was located in Beypazarı, Ankara. Quartz was commercially obtained from MATEL Raw Materials Industry and Trade Company. The chemical composition of trona sample was analyzed by inductively coupled plasma mass spectrometry (ICP-MS). The phases present in the samples were identified by X-ray diffraction (XRD) using a Rigaku D/Max-2200/PC model diffractometer. The XRD patterns were obtained with the diffractometer at 40 kV and 36 mA using Cu K_α radiation. Thermal analyses of the samples were done by using a Shimadzu DTG-60 H model DTA/TG. For morphological characterization of trona powder and formed glass cullets, a scanning electron microscope (SEM, JEOL-JSM 6060) was used. The SEM was operated in the secondary electron (SE) mode with 10 kV and 20 kV acceleration voltage levels. Chemical make-up of samples was determined by an energy dispersive X-ray spectrometer; EDS (IXRF System 500) attached to the SEM. To produce of sodium silicate cullets, trona and quartz powders were used. The batches with different SiO_2:Na_2O molar ratios were prepared and mixed according to the equations below

$$2Na_2CO_3NaHCO_3 \cdot 2H_2O + 3SiO_2 \rightarrow 3Na_2OSiO_2 + 2CO_2 + 5H_2O, \quad (2)$$

$$2Na_2CO_3NaHCO_3 \cdot 2H_2O + 6SiO_2 \rightarrow 3Na_2O2SiO_2 + 2CO_2 + 5H_2O, \quad (3)$$

$$2Na_2CO_3NaHCO_3 \cdot 2H_2O + 9SiO_2 \rightarrow 3Na_2O3SiO_2 + 2CO_2 + 5H_2O, \quad (4)$$

$$2Na_2CO_3NaHCO_3 \cdot 2H_2O + 12SiO_2 \rightarrow 3Na_2O4SiO_2 + 2CO_2 + 5H_2O. \quad (5)$$

Powders mixtures were placed in a porcelain crucible and separately fired at 1150 °C and 1200 °C for 2 h. These fused melts were drawn from the kiln and poured into a water to solidify. The resulting products (cullets) were characterized by using SEM and XRD analysis.

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3. Results and discussion

3.1. Characterization of trona

Table I lists the chemical analysis results of the trona powder. Its high Na_2O and loss of ignition content indicates the purity of the ore. XRD analysis of the trona powder can be seen in Fig. 1. According to the XRD data base, the powder sample contains trona, natrite, natron and nahcolite phases. The index number given in the parenthesis in Fig. 1 shows the XRD pdf card numbers according to the JCPDS-ICDD formats.

TABLE I

Chemical analysis results of the trona powder.

Oxides [wt%]	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	MnO	LOI
Trona	0.16	0.03	<0.04	0.18	0.17	49.75	<0.01	<0.01	42.70

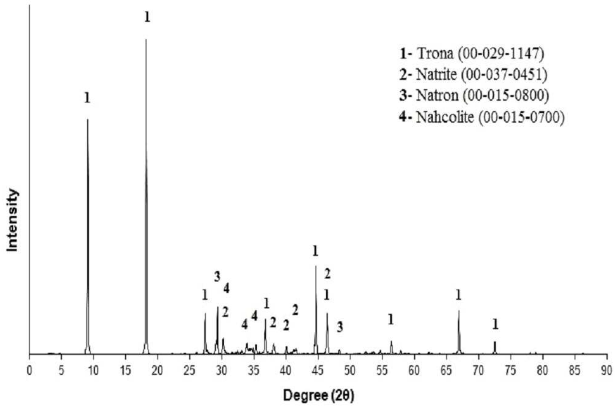

Fig. 1. XRD analysis of the trona powder.

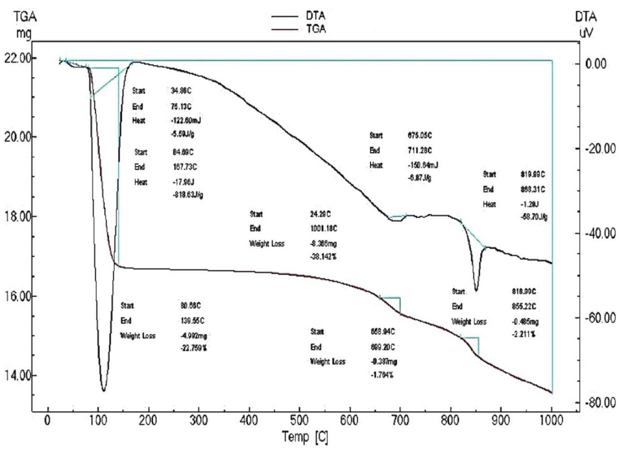

Fig. 2. Thermogravimetric analysis of the trona powder.

Figure 2 shows the differential thermal/thermal gravimetric analyses (DTA/TGA) of the trona powder. DTA analysis indicates that there are exothermic reactions. The exothermic reaction that takes place between 35 °C and 75 °C and 84 °C–167 °C shows that trona create intermediates such as wegschiderite ($Na_2CO_3 \cdot 3NaHCO_3$) and sodium monohydrate ($Na_2CO_3 \cdot H_2O$). Above 160 °C, trona decomposes to sodium carbonate [2]. Also another exothermic reaction is observed between 819 °C and 868 °C. According to the TGA analysis, the trona powder shows a sharp weight loss after approximately 85 °C due to the exothermic reactions explained above.

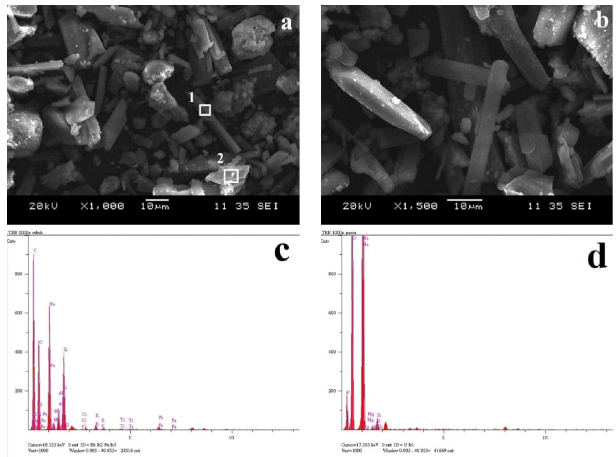

Fig. 3. (a, b) SEM images of the trona powder, (c) EDS analysis graph of area 1 and (d) EDS analysis graph of area 2.

Figure 3a,b shows SEM images of the trona powder. Here, powder crystals with different geometric shapes and dimensions in approximately 10 μm were observed. Figure 3c,d shows EDS graphs of the areas indicated as 1 and 2. EDS analysis of the white square area (indicated as number 1) showed that the composition of the area contained 42.265% C, 31.512% O, 14.467% Na, 0.834% Mg, 1.978% Al, 7.043% Si, 0.083% Cl, 0.533% K, 0.207% Ti, and 1.079% Fe (by weight). Energy dispersive spectrometry (EDS) analysis of the white square area (indicated as number 2) showed that the composition of the area contained 12.681% C, 41.405% O, 44.242% Na, 0.187% Mg, 0.355% Al, 1.131% Si (by weight).

3.2. Production of sodium silicate cullets

Table II lists the recipes of different sodium silicate cullets prepared according to Eqs. (2)–(5). The composition of the recipes and melting temperature played a great role on the viscosities of the sodium silicate melts. The viscosity of the melt became higher with increasing quartz amounts. SSR4 could not be poured due to its high viscosity at 1150 °C. Therefore, it was not possible to get samples from this recipe at 1150 °C.

The XRD mineralogical characterization (Fig. 4) showed the cullets to be an amorphous material. The analyses showed that crystalline phases began to form with increasing temperature and SiO_2 ratios. SiO_2, Na_2SiO_3, $Na_2SiO_3 \cdot 5H_2O$, $Na_2SiO_3 \cdot 9H_2O$ and NaH_3SiO_4 phases were identified at the sample melted at 1200 °C.

TABLE II

Sodium silicate cullet recipes.

Raw materials	SSR1	SSR2	SSR2	SSR4
Trona [wt%]	71.52	55.67	45.56	38.57
Quartz [wt%]	28.48	44.33	54.44	61.43

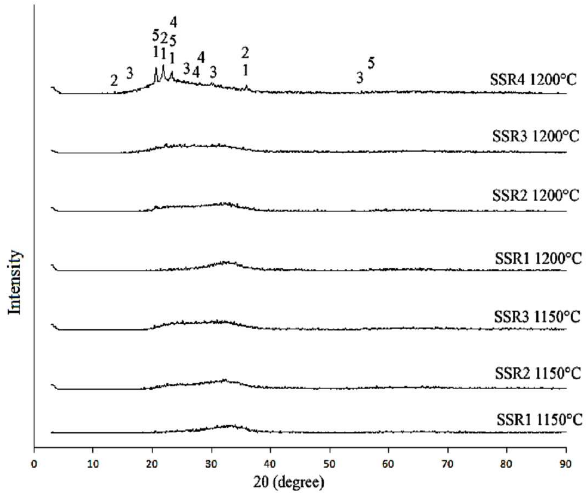

Fig. 4. XRD 2 theta scans of the samples obtained by melting at 1150 °C and 1200 °C (1: SiO_2, 2: $Na_2SiO_3 \cdot 5H_2O$, 3: Na_2SiO_3, 4: $Na_2SiO_3{\cdot}9H_2O$ and 5: NaH_3SiO_4).

Microstructures of sodium silicate cullets that were obtained by melting at 1150 °C can be seen in Fig. 5. EDS analysis of the white square area in (b) showed that the composition of the area contained 10.358% Na, 66.656% Si and 22.986% O (by weight).

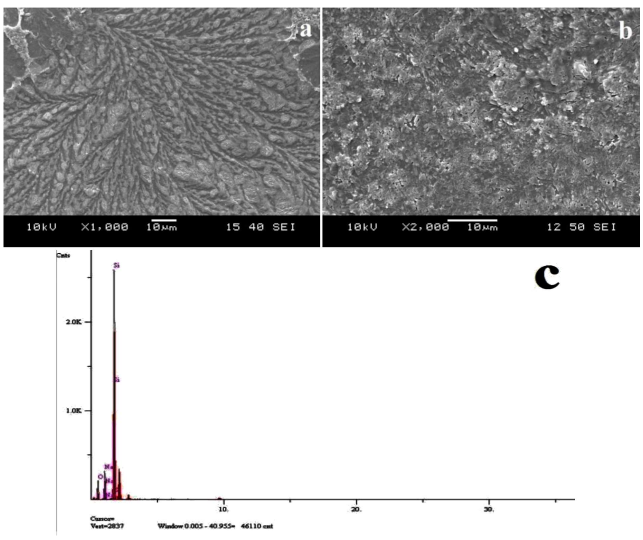

Fig. 5. Microstructures of the samples melted at 1150 °C (a) SSR2, (b) SSR3 and (c) EDS analysis graph of the white square area in (b).

Figure 6a,b shows the SEM images of the sodium silicate cullets obtained by melting at 1200 °C. Figure 6c–f shows EDS graphs of the areas in Fig. 6a,b. EDS analysis of the white square area (indicated as number 1) in (a) showed that the composition of the area contained 17.019% Na, 54.070% Si and 28.911% O (by weight). EDS analysis of the white square area (indicated as number 2) in (a) showed that the composition of the area contained 15.829% Na, 54.550% Si and 29.621% O (by weight). EDS analysis of the white square area (indicated as number 1) in (b) showed that the composition of the area contained 23.459% Na, 49.264% Si and 27.277% O (by weight). EDS analysis of the white square area (indicated as number 2) in (b) showed that the composition of the area contained 8.626% Na, 69.420% Si and 21.954% O (by weight).

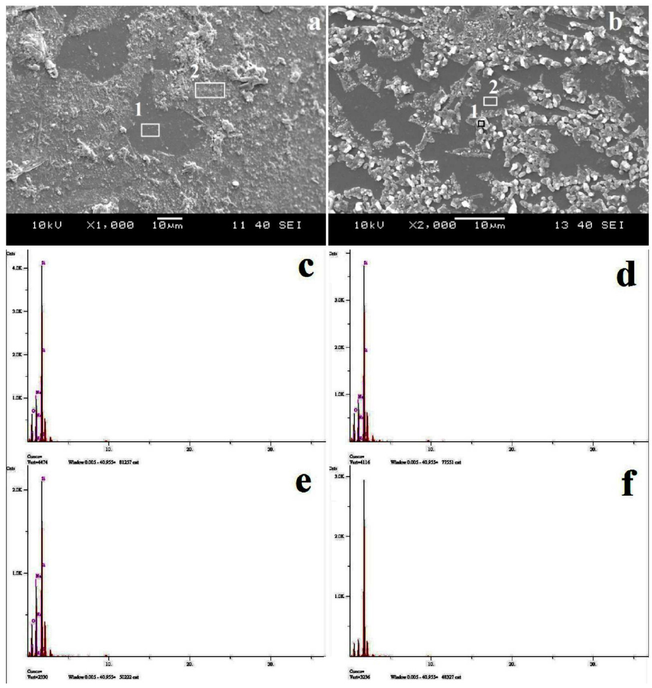

Fig. 6. Microstructures of the samples melted at 1200 °C (a) SSR2 and (b) SSR3, EDS analysis graphs for (c) the white square area (indicated as number 1) in (a), (d) the white area (indicated as number 2) in (a), (e) the black square area (indicated as number 1) in (b) and (f) the white area (indicated as number 2) in (b).

4. Conclusion

In this paper, characterization of trona and its usage in the production of sodium silicate cullets were studied. Chemical analysis results of the trona powder showed that it has high Na_2O and loss of ignition content indicates that purity of the ore. XRD analysis revealed that trona powder sample contained trona, natrite, natron and nahcolite phases. According to SEM images, crystals with different geometric shapes and dimensions in approximately 10 μm were observed in trona powder sample. After melting the trona–quartz mixtures at 1150 and 1200 °C, sodium silicate cullets were obtained. The results showed that trona can be used instead of soda ash to produce sodium silicate cullets. Although the results of sodium silicate cullet production studies were largely positive, there are other controlling factors such as melting temperature and composition of the recipes, which usually influences the quality of the finished product. In this regard, more detailed studies with different compositions and melting temperatures are recommended.

Acknowledgments

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