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RESEARCH ARTICLE

LIGHTWEIGHT AGGREGATE PRODUCTION by RAPID SINTERING TERRA ROSSA-NaOH MIXTURES in MICROWAVE OVEN

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ABSTRACT

In this study, the use of microwave ovens (MO) in the production of lightweight aggregates (LWA) with rapid sintering method was investigated. Sintering temperatures up to 1200 C° was obtained by using SiC coated microwave kiln in MO. LWA were produced by rapid sintering terra rossa (TR)-NaOH mixtures at MO. Alkali amount (2.5, 5 and 7.5% NaOH by weight of TR), drying type (no drying, drying in laboratory at 20 °C or in oven at 80 °C for one day) and sintering time (8, 12 and 18 min) on the physical, mechanical and microstructure of aggregates were investigated. The aggregate pellets dried in oven were stronger than those dried in the laboratory. Thus the sintering cracks diminished and the compressive strengths were enhanced for aggregates. The highest compressive strength (36.96 kg/cm^2) was obtained in aggregates using 2.5% NaOH by weight of TR, dried in oven and rapid sintered for 18 minutes. When more alkali (5% NaOH) was used, due to the increase in flux content; the aggregate shell formed earlier and had more time to expand up to 127%. Thus the unit weight (0.67 g/cm^3) and compressive strength (4.74 kg/cm^2) of the aggregate were decreased.

Keywords: *Rapid sintering, Microwave, Terra rossa, Lightweight aggregate, Compressive strength*

1. INTRODUCTION

Humans first kneaded and shaped the soil, which can remain intact later, in the Paleotic era, 35,000 years ago [1]. Terra rossa (TR) are reddish, bordeaux and brownish clay and silt-sized soils commonly found in the Mediterranean region. TR consists of clay minerals such as sericite, montmorillonite, illite, chloride, hydromica and silicon compounds, clay minerals rich in Fe2O3, clay minerals, albeit in small quantities, with a fine material content of more than 20%, containing less than 30% Al2O3 [2]. Soft and very fine-grained feldspar with a high amount of clay, aluminum and silica, with a large amount of fine grains, is called a sedimentary mineral formed by the decomposition of mica and other silicates. Clay makes it easier to shape the soil. It is of great importance that the type and amount of clay in the soil is suitable for lightweight expanded clay aggregate (LECA) production [3]. LECA can

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be produced by drying, heating and rapid sintering plastic clay having too low or no lime content in rotary kilns at 1100 to 1300 °C [4]. LECA were commonly used as lightweight filler material in geotechnical works and production of lightweight concrete [5].

At high sintering temperatures, the aggregate pellets become pyroplastic. In the meantime, the gas phase is formed from the combustion of organic materials, water vapor, reduction of iron oxides and decomposition of carbonates and sulfates. In the pyroplastic state, gases trying to escape inflates the closed pores of agregate. Thus increases up to 5-6 times in the volume of agregate can be observed [6, 7, 8, 9, 10,11]. Na_2O can significantly reduce the viscosity of the silicate melt by depolymerizing the silicate networks [12]. Ge et al. [13] observed that increasing the Na_2O contents reduced the viscosity of ceramic foams sintered.

Microwaves are defined as electromagnetic radiations with wavelengths ranging from 100 cm to 1 mm. Microwave ovens operate at frequencies in the range of 950 MHz and 2450 MHz. It became widespread with the use of radar in the second world war. Later, microwave ovens entered the interiors of homes and were used for tasks such as heating food. Microwaves stimulate molecules or ions [14]. It is stated that the sintering time of Nickel-Zinc ferrites has been reduced from 9 hours to 4 hours and less than 69% energy is used by microwave sintering.The sintering time of alumina decreased from 15-20 hours to 6 hours and 65% less energy was consumed by microwave sintering. The pure Al_2O_3 sample from room temperature to 1700°C using 60 GHz radiation, an intensity of 95.7% was achieved over a period of 6 hours.To achieve a density of 95.7% of the same type of samples, conventional sintering requires sintering at 1600 °C for 20 hours. In any case, microwaves have produced fine-grained structures than traditional sintering. As a result, the materials became denser, stronger and more durable. At the same time, they have undergone less deformation and breakage during production. Microwave technology has been shown to significantly shorten the sintering time. The microwave sintering method is most often used in the sintering of ceramics [15]. Silicon carbide (SiC) is high temperature resistant microwave-absorber. Besides it has good chemical stability and mechanical strength and it is readily available [16, 17]. Thus SiC coated microwave kiln was used in this study to reach high sintering temperatures.

A significant part of the area of Kütahya-Turkey was occupied by mountain terrains/high lands, which are meta-carbonate rocks (marbles). The most characteristic types of red soil over carbonate rocks are Terra Rossa. The TR can be suitable as raw material in the brick and roofing tile industry. The present study presents the results of a master thesis [18] that was motivated by a lack of data on the production of aggregates by rapid sintering of TR-NaOH mixtures at microwave oven. This paper aims to determine the contribution of alkali amout, drying type and rapid sintering time to the physical and mechanical properties of aggregates.

2. MATERIALS AND METHOD

Terra rossa (TR) collected from the village of Çöğürler in Kütahya/Turkey was ground by a ringed mill (Fig.1) to increase the surface area up to 2700 cm^2/g. The TR was mainly composed of SiO_2, and Al_2O_3 (Table 1). Aggregates were produced by i) mixing TR with technical grade NaOH (>99% purity obtained from Detsan Chemicals Company in Eskişehir/TURKEY) and 33% water by weight of

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TR+NaOH for 3 minutes and forming pellets by hand ii) drying them for one day or no drying iii) rapid sintering them at microwave oven.

Table 1. Oxide content of TR.

Oxide content	SiO_2	Al_2O_3	Na_2O	K_2O	TiO_2	Fe_2O_3	CaO	MgO	MnO	Cl	SO_3	*LOI
% mass	56.07	15.98	0.14	1.93	0.57	6.52	1.91	1.27	0.11	0.06	0.05	15.25

*LOI: Loss on ignition

A total of 27 series were produced. Variables were alkali amount (2.5, 5 and 7.5% by weight of TR), drying type (drying them at 20±2 °C in laboratory for one day: L or 80 °C in oven (Fig.1) for one day: O or no drying: N) and rapid sintering time (sintering them at microwave oven given in Fig.1 for 6, 12 or 18 mins). As an example series produced with 5% NaOH by weight of TR, dryed for one day at 20±2 °C in laboratory and rapid sintered in microwave oven (MO) for 18 mins was named as NH5-L18.

Household microwave with output power of 700 Watts and microwave frequency of 2450 Megahertz was used.), SiC coated microwave kiln given in Fig. 2 was used to absorb microwaves and reach high sintering temperatures. The temperatures of aggregate shell were determined as 840, 980 and 1200 °C with infrared thermometer after 6, 12 and 18 mins of sintering at highest power.

Figure 1. The photos of (a) ringed mill, (b) blaine device, (c) oven and (d) household microwave oven

Unit weight (UW) of the aggregates were calculated according to TS EN 1097-6 [19]. Fracture load of agregates were determined by one point loading device using aparatus given in Fig. 2 to spread the load uniformly. One point loading device was preferred for precise measurement of the little crushing loads. All the reported results are the means of three samples. The crushing strength (σ, kg/cm^2) for speherical aggregates are calculated by the equation 1 where: P: fracture load (kN), and X: the distance between loading points (mm) [20, 21]. SEM-EDX analyses on core and shell of the aggregates were conducted.

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$$\sigma = ((2.8*1000*P) / (\pi*X^2)) \text{ (Yıldırım \& Özturan, 2013)} \quad (1)$$

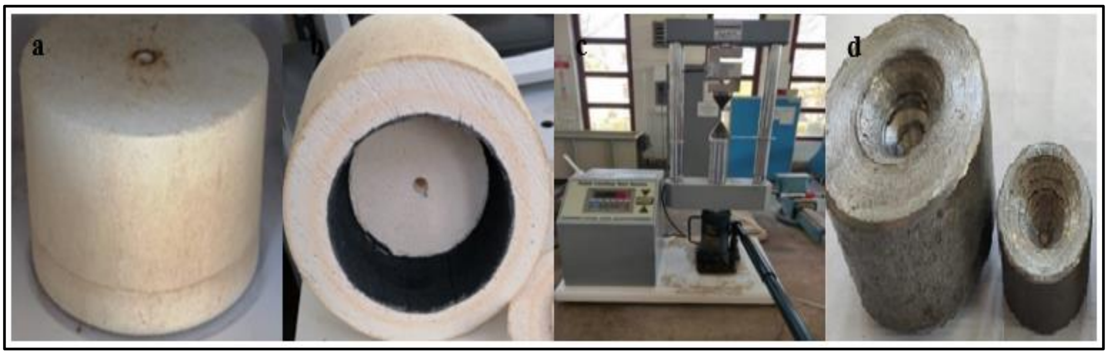

Figure 2. The photos of (a), microwave kiln (b), SiC coating inside the kiln (c) single point loading device (d) apparatus to spread the load uniformly

3. EXPERIMENTAL RESULTS

3.1. Evaluation of Aggregate Photos

The photos of aggregates with different NaOH amounts, drying types and sintering times were shown in Fig. 3-5. Sintering times of 6 minutes was inadequate and caused cracking on the outer shell of aggregates as seen in Fig. 3. Aggregates which were sintered without drying were too week and crushed during sintering. Increasing the amount of NaOH, cracks in the outer shell of aggregates were decreased. The surface of oven dried aggregates were smoother compared to others.

Figure 3. The photos of (a) NH2.5-N6, (b) NH2.5-L6, (c) NH2.5-O6, (d) NH5-N6, (e) NH5-L6, (f) NH5-O6, (g) NH7.5-N6, (h)NH7.5-L6 (ı) NH7.5-O6.

As the MW sintering time increased sintering enhanced. There is a noticeable formation of glassy phase on surface of aggregate on series produced with 5 and 7.5% NaOH by weight of TR (Fig. 4). Noticeable amount of expansion were not seen for sintering of 12 mins.

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Figure 4. The photos of (a) NH2.5-N12, (b) NH2.5-L12, (c) NH2.5-O12, (d) NH5-N12, (e) NH5-L12, (f) NH5-O12, (g) NH7.5-N12, (h)NH7.5-L12 (ı) NH7.5-O12.

As a result of the visual examinations in Fig 5, the colour of aggregates got darker with increasing sintering time. After sintering the iron element in oxidation state II oxidised to state III could be responsible from darker colours of aggregates [22, 23]. Glassy phase enhanced in the aggregates sintered for 18 minutes.

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Figure 5. The photos of (a) NH2.5-N18, (b) NH2.5-L18, (c) NH2.5-O18, (d) NH5-N18, (e) NH5-L18, (f) NH5-O18, (g) NH7.5-N18, (h)NH7.5-L18 (ı) NH7.5-O18.

3.2. Evaluation of Physical and Mechanical Properties

The aggregates produced with 5% NaOH by weight of TR and 18 mins of sintering time showed the highest expansion rates. After rapid sintering in microwave oven (MO) shrinkage up to 31.4% (NH7.5-N6) and expansion up to 127% (NH5-L18) were observed in aggregates. NaOH accumulated on the aggregate shell by diffusion of alkali solution during drying. By the aid of fluxing alkalis, the shell formation accelerated. Since sintering time is constant for all aggregates, there was more time for expansion of aggregate pellets produced with more NaOH.

Drying at laboratory also supported the expansion of aggregate by holding more water in aggregate pellet. Escaping of the water vapour during sintering blowed up the aggregate. The lowest unit weight (0.67 gr/cm^3) was obtained for NH5-L18. The highest unit weight was 2.13 g/cm^3 for NH2.5-O18. Drying in oven at 80 °C excessively decreased the water content of aggregate pellets. Thus there were not enough water steam that will blow the aggregate during sintering.

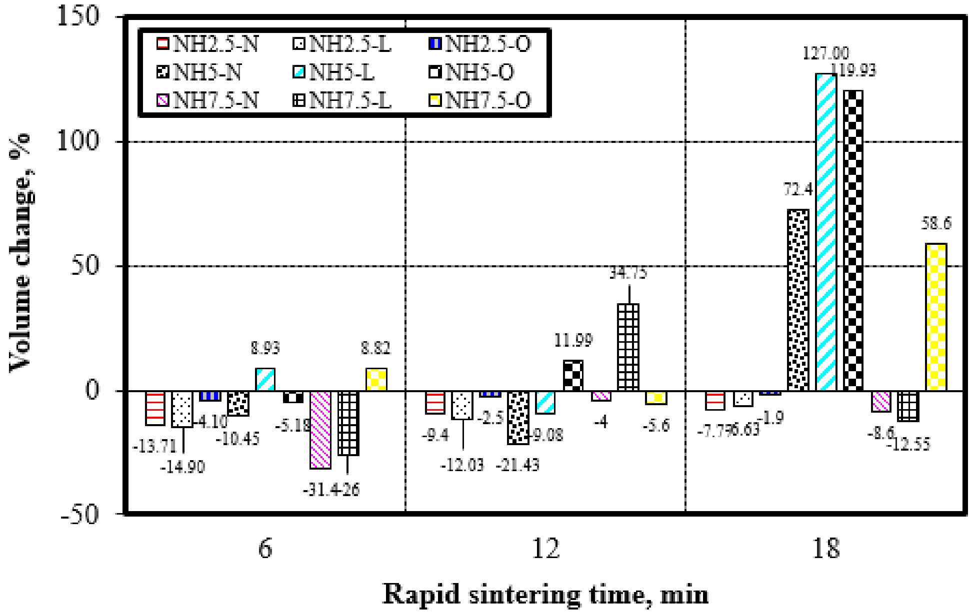

Figure 6. Volume change of the aggregates.

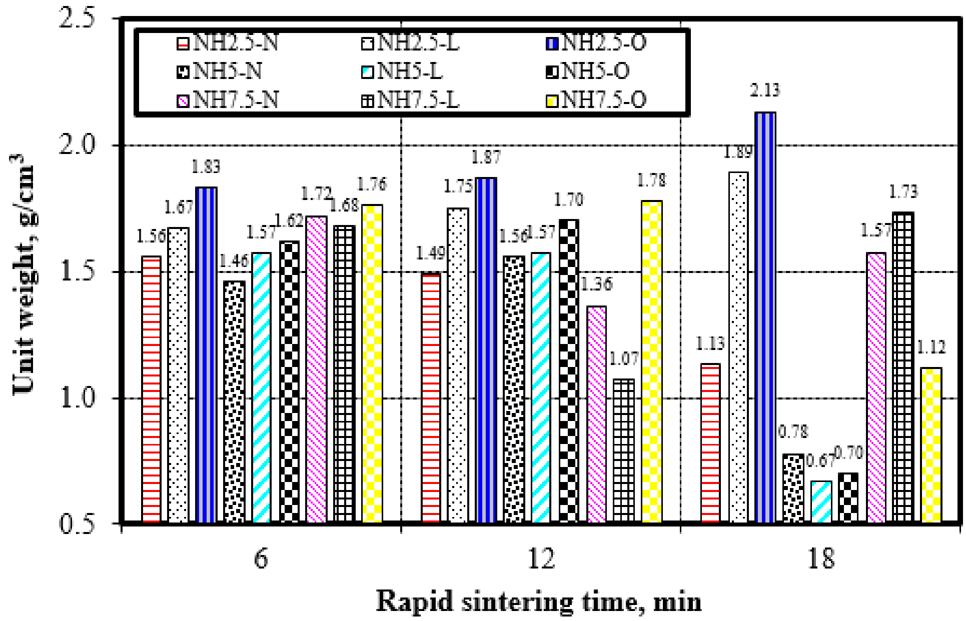

Figure 7. Unit weight of the aggregates.

Drying process significantly affected the water absorption by weight (WA) of aggregates. The highest WA were obtained for aggregates that were not dryed before sintering. The lowest values were obtained for aggregates sintered for 18 mins. Thus drying before sintering decreased the WA.

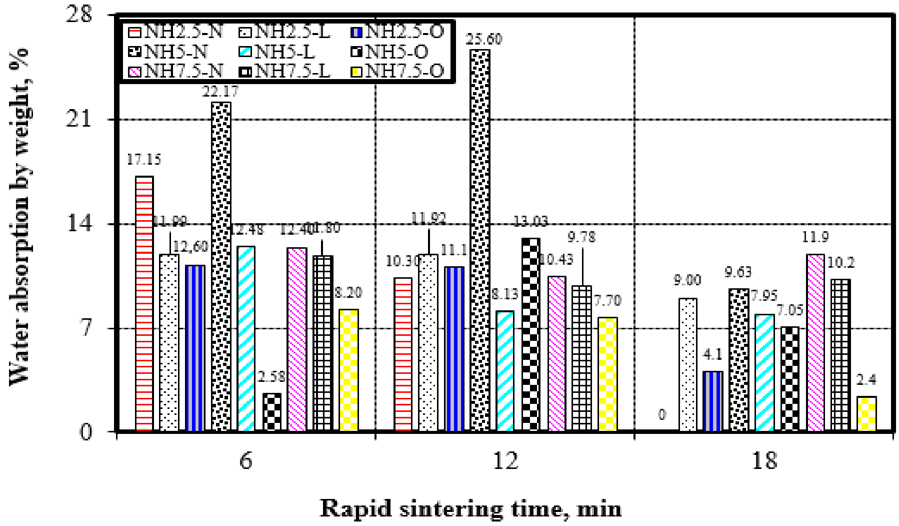

Figure 8. Water absorption of the aggregates.

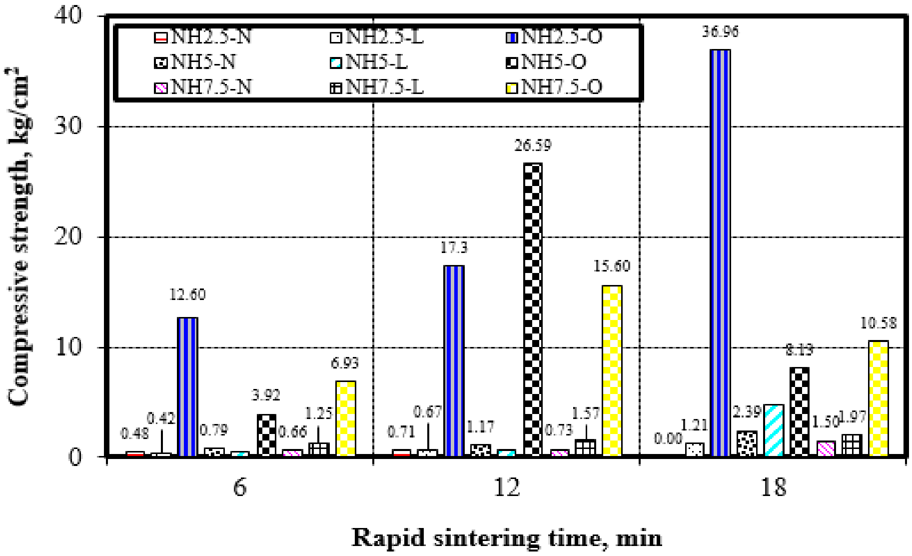

Figure 9. Compressive strength of the aggregates.

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The compressive strength of NH2.5-018 (36.96 kg/cm^2) is much higher than that of NH5-L18 (4.7 kg/cm^2). It is clearly seen from aggregate photos for sintering time of 18 mins (Fig.5) that the NH2.5-O18 forms a thicker and stronger shell than NH5-L18. This situation coincides with SEM analysis. It was revealed that aggregates can be produced by rapid sintering TR-NaOH mixtures in MO.

3.3. Evaluation of SEM-EDX Analysis

Most of the ceramics are transparent to microwaves at 2.45 GHz for low temperatures. Susceptors absorbing the microwaves and transferring the heat to the material were used for microwave sintering of ceramics. Until the critical temperature, susceptors heat the material from surface to core. Then the material starts to absorb the microwaves and participate to sintering process [24, 25]. Lyra et al. produced light weight aggregates (LWA) by microwave oven using silicon carbide as susceptor or electric oven for sintering red clay-sugarcane bagasse ash mixtures. They indicated with SEM observations that the pores were closed and homogeneously distributed when sintered with microwave oven. Lower water absorption and higher compressive strength values were obtained when compared with electric oven sintering [26]. A significant expansion was seen in NH5-L18 from the SEM images compared to NH2.5-O18. NH5-L18 expanded 127% by volume, while reducing the unit weight of aggregate to 0.67 g/cm^3. In NH5-L18, amount and diameter of pores were bigger. Low water absorption of NH2.5-O18 could be linked to it's lower porosity than NH5-L18.

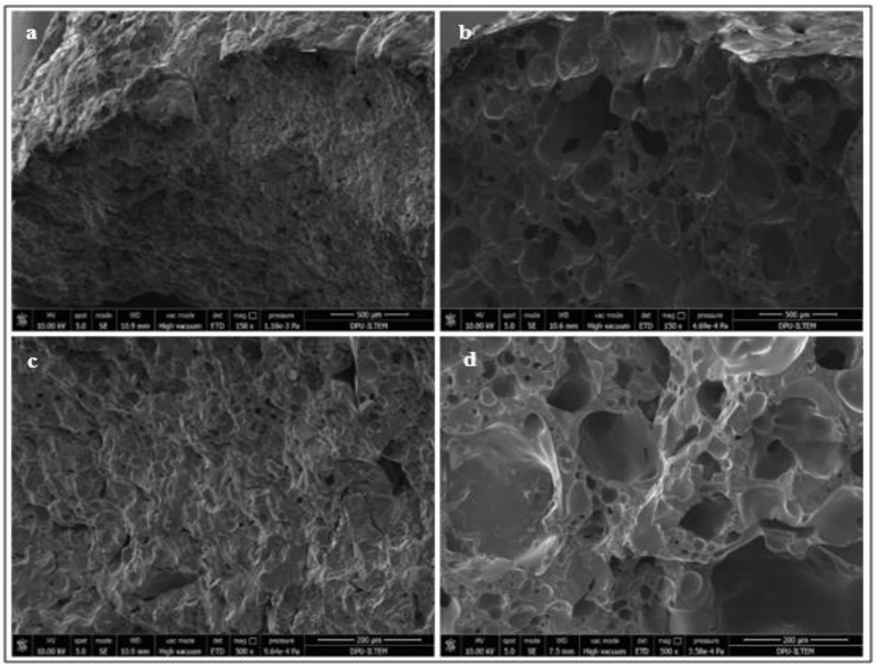

Figure10. SEM photo (150X) of aggregate shell (a) NH2.5-O18, (b) NH5-L18 and (500X) of aggregate core (c) NH2.5-O18, (d) NH5-L18.

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NH2.5-O18 have a homogeneous structure with small cracks and pores. It is seen in EDX data (full area 1) that the amount of alkali is significantly lower than NH5-L18 (selected area 2). NH5-L18 aggregate appears to contain quartz crystals in the glassy structure, indicated by a SiO_2 ratio of 93% (selected area 1). This structure was also seen as white grains in aggregate photos (Fig 3-5). NaOH accumulated on the aggregate shell by diffusion of alkali solution during drying. By the aid of fluxing alkalis, formation of the aggregate shell accelerated. Since sintering time is constant for all aggregates, there was more time for expansion of aggregate pellets produced with more NaOH. Drying at laboratory also supported the expansion of aggregate by holding more water in aggregate pellet.

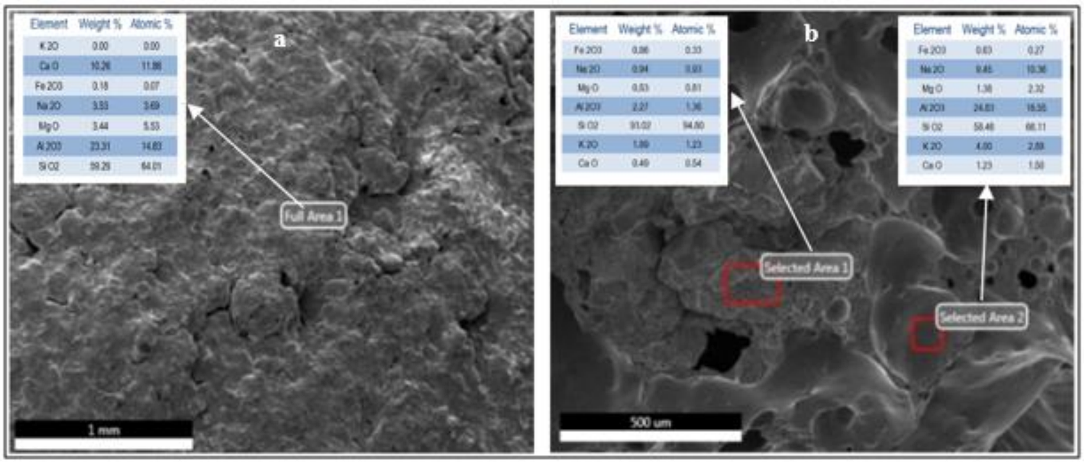

Figure 11. SEM-EDX analysis of (a) NH2.5-O18, (b) NH5-L18.

4. CONCLUSIONS

Lightweight expanded clay aggregates are produced by rapid sintering in tunnel kilns all over the world. This study revealed that lightweight aggregates can also be produced by rapid sintering in household microwave oven. Industrial microwave ovens can be used for commercial production and optimization of the process. Cost analyses should also be performed for commercial use. As the sintering time increased colours of the aggregates got darker. The rapid sintering time of 6 mins was very short to reach adequate sintering temperature for Terra rossa-NaOH mixtures. Aggregate pellets produced with more NaOH expanded more. Drying at laboratory also supported the expansion of aggregate by holding more water in aggregate pellet. The lowest unit weight (0.67 gr/cm^3) was obtained for NH5-L18. The highest unit weight was 2.13 g/cm^3 for NH2.5-O18. The aggregate pellets dried in oven were stronger than those dried in the laboratory. The highest compressive strength (36.9 kg/cm^2) was obtained for NH2.5-L18. For future studies, minerals containing higher alkali content can be effective raw materials for lightweight aggregate production.

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