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Fabrication of machinable phlogopite–glass composite using microwave processing

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Abstract

Crystallization and sintering of glass–phlogopite composites were studied using microwave and conventional heating. It was found that by increasing phlogopite content the hydroxyl group in phlogopite structure absorbs microwave irradiation and reduces the optimum sintering temperature.

Diopside was the resulting phase after conventional and microwave heating. XRD results showed that the decomposition of phlogopite occurred 100 °C lower when using microwave irradiation instead of conventional heating. The easy machining of the components sintered in microwave was related to the presence of phlogopite phase. The morphology of diopside sintered by microwave was almost fiber like (100 nm in diameter), whereas in conventional method these crystals were plate like (5 μm in diameter).

Keywords: A. Microwave; A. Sintering; B. Composite; D. Glass

1. Introduction

Phlogopite is a flaky mineral with outstanding chemical, thermal, electrical and flame resistance, and tough. These properties make it a candidate for combining it with a polymer [1] or glass [2] to produce a though and reinforce composite, which has the potential for making composites with low heat conduction, good sound proofing, and fire resistance; such composites may find application in low-cost housing, ceiling boards, roof tiles and dry walling.

On the other hand the microwave (MW) sintering of soda-lime glass with added iron [3], magnetite [4] and molybdenum oxide [5] powder has been studied.

It is possible that the interlayer water and magnetite impurity of phlogopite act like the MW absorber and facilitate the MW heating simultaneously [6].

The sintering behavior of soda lime glass and phlogopite mineral mix-powder was studied by conventional method. It was shown that the thermal decomposition of phlogopite at high temperatures causes composite bloating [7].

In this work the crystallization and sintering behavior of phlogopite–glass composites under conventional heating were compared with MW heating.

Considering the high heating rate in MW, it is expected that the sintering time of glass decreases and the decomposition of phlogopite occurs at a high temperature in the phlogopite–glass composite.

2. Experimental procedures

Glass powders used for present investigation were prepared from recycled colorless soda-lime glass cullet supplied by a local glass manufacture (Gazvin Glass Ltd., Iran). Phlogopite was obtained from a type of phlogopite processed in Varom gia. Oromiea from a high purity ore under a trade name, also known as phlogopite mica. The chemical analysis of glass and phlogopite is represented in Table 1.

The glass cullet was first pulverized into fine grains (in the range of 0.2–0.5 mm in diameter) using mechanical pulverization method. The pulverized glass grains were then ground into fine powders by a grinding process of wet ball mill for a period of about 70 h, using corundum grinding media. The glass and phlogopite powders used for sample preparation were mostly in the range of 25–62 μm, respectively. Typical particle size

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Table 1
Chemical composition of glass and phlogopite.

	SiO_2	K_2O	Na_2O	MgO	CaO	Al_2O_3	Fe_2O_3
Glass	73	–	14	3.2	5.9	2.7	–
Phlogopite	37	10	–	25	2.7	15	3.2

distribution of prepared powders which was measured by Fritsch Particle Size Analysis, (analysette 22) is shown in Fig. 1a and b. The XRD pattern of used phlogopite is shown in Fig. 2.

The weight ratios of phlogopite/glass compositions were 20/80, 30/70 and 40/60 denoted as C28, C37 and C46, respectively. For most experimental evaluation purposes, 50 g of composite powder was prepared from appropriate proportions of the recycled glass and phlogopite powders. Mixing of phlogopite and glass powders was carried out for 2 h in an ordinary ball mill with alumina balls using water as a wet media. The obtained mixture was dried at 120 °C and then uniaxially pressed at 70 MPa into circular discs (50 mm diameter and 10 mm thickness). Pressed samples were heated in an electric furnace under atmospheric conditions in the temperature range of 600–1050 °C for 10 min at heating rate of approximately 10 °C/min. Microwave sintering process was conducted using a microwave furnace (2.45 GHz and 900 W); further details have been reported in Ref. [8]. Density of sintered samples was measured according to ASTM-C20.

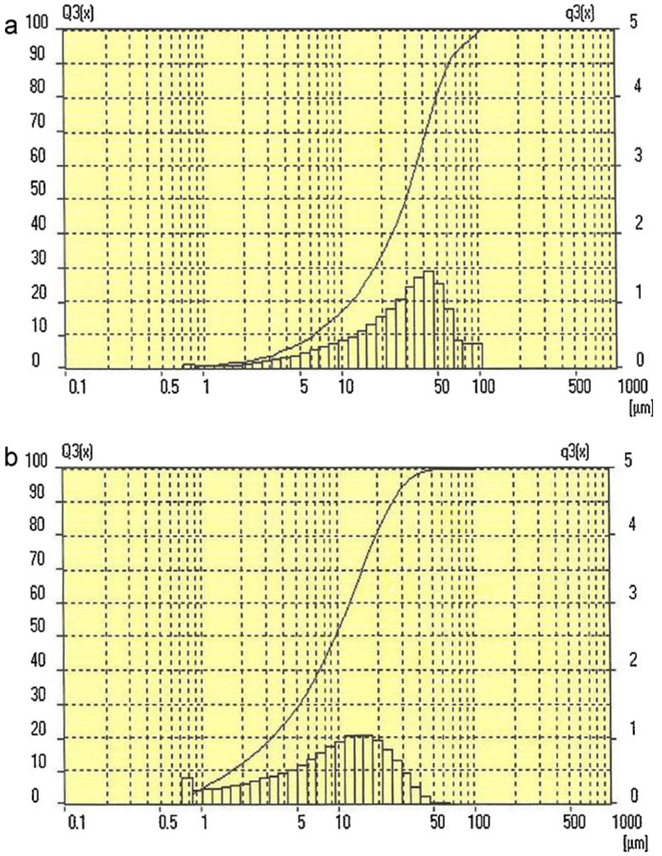

Fig. 1. (a) PSA of glass. (b) PSA of phlogopite.

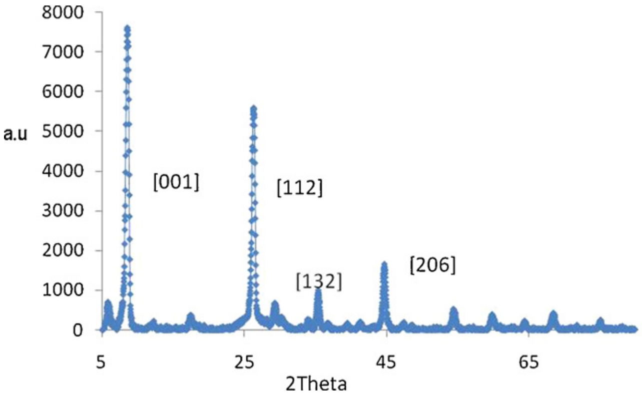

Fig. 2. XRD pattern of phlogopite.

Crystalline phase identification was performed on powder made from ground sintered pellets, using X-ray diffraction (Philips Power Diffractometer 1710) with Ni-filtered Cu K_α radiation and the relevant JCPDS cards (Joint Committee on Powder Diffraction, 1972).

SEM/EDX (JEOL JXA-840) was used in order to observe and analyze the microstructure of sintered samples.

A Fourier transform infrared (FTIR-Bruker-Vector 33) spectroscopy was used for studying water structure.

3. Results and discussion

Thermal analysis corresponding to phlogopite and C37 is presented in Fig. 3. It seems that 4.5 wt% weight loss and endothermic peak appearing at 650 °C are due to the loss of interlayer water in phlogopite. Besides, continuous weight loss of C37 during heating up to 1100 °C is observed. In order to identify the volatile mat, the FT-IR analysis was performed on raw glass and treated glass at 850 °C; these results are presented in Fig. 4.

The intensity of principle peak at 3440/cm related to the molecular water was reduced at 850 °C. This structural water

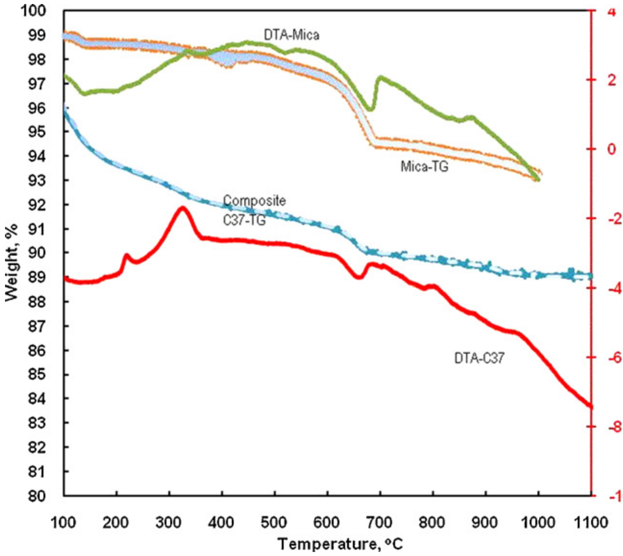

Fig. 3. Thermal analysis of phlogopite and glass composition.

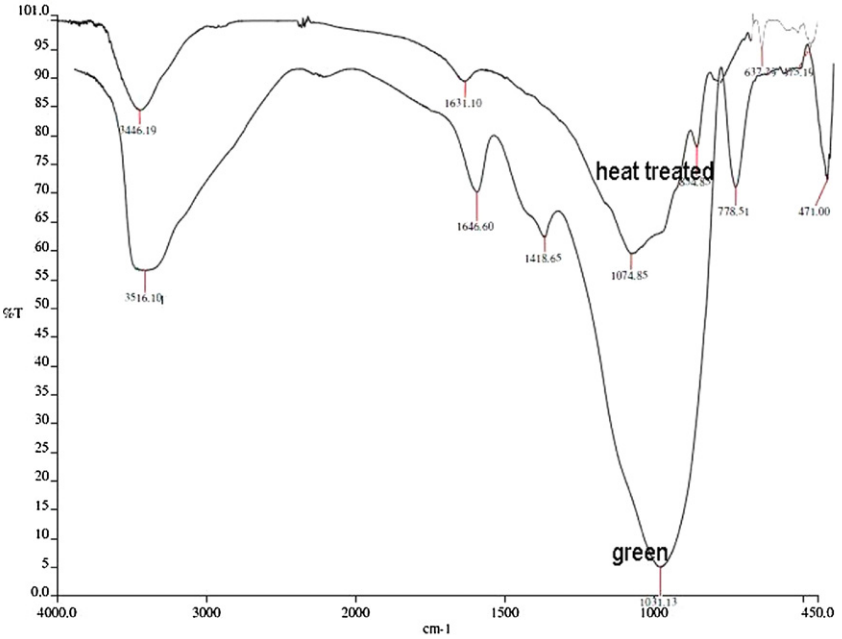

Fig. 4. FT-IR analysis of as received glass and heat treated at 850 °C.

may be originated from raw materials used in the glass melting or wet milling process which is essential for making fine glass powder [9]. According to DTA results, the T_g of glass should be at 560 °C. Therefore, it is assumed that the sintering temperature begins from 600 °C as Fig. 5 confirms. The densification behavior of different samples was studied from 600 to 1050 °C and results are shown in Fig. 5a and b.

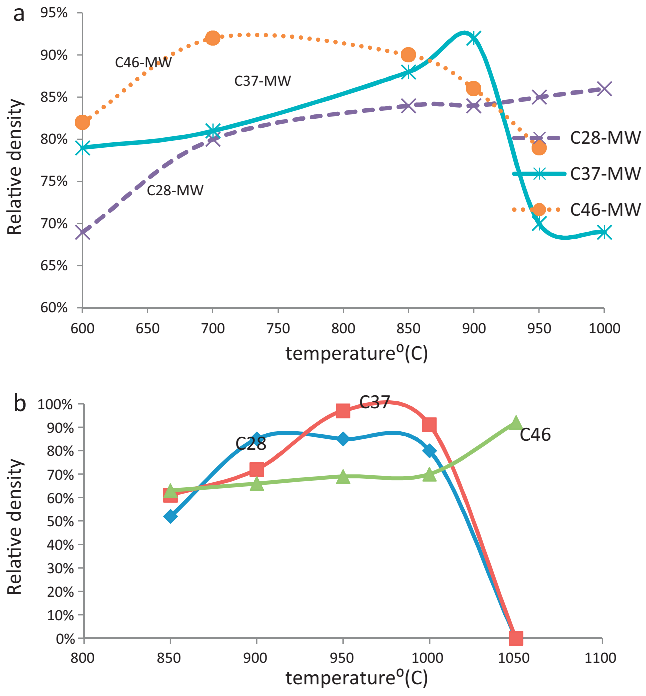

Fig. 5. (a) Composites sintered by MW. (b) Composites sintered by conventional heating.

As Fig. 5a shows, the optimum sintering temperature of composites has been reduced using microwave process. Apparently, the crystallization temperature increased because of the high heating rates in MW process, therefore, the sintering temperature decreased. On the other hand, the rapid sintering without holding in MW process is not sufficient heating conditions for conventional sintering to reach a high density; furthermore, bloating of samples was observed in conventional method (Fig. 5b).

A viscous layer can be formed on the surface of sample as a result of conventional heating which may prevent the extraction of gas molecules and trap them inside the sintered body. In this case, the trapped gas decreases density of samples.

According to Fig. 5a, C46 sample sintered in MW at 700 °C consists of a high amount of muscovite. It can be concluded that the existence of more muscovite phase may cause more MW absorption, and finally increased the resulted density. But diopside phase, which was found in samples heated conventionally in the present work, increases effective viscosity [10] and causes the degradation of sintering.

The hydroxyl group or magnetite (as impurity) in phlogopite can be considered mainly responsible for the interaction between the microwave irradiation and material. X-ray diffraction patterns of samples sintered in MW and furnace are shown in Fig. 6.

As observed, samples sintered at 700 °C in both heating methods (MW and conventional) contain muscovite as the only crystalline phase. Dehydrated muscovite is the transformed structure of phlogopite at high temperature [11].

As mentioned before, C37 was not crystallized at 700 °C by MW heating but obtained a high density (Fig. 6). It is very likely that the OH^- groups of muscovite and glass absorb

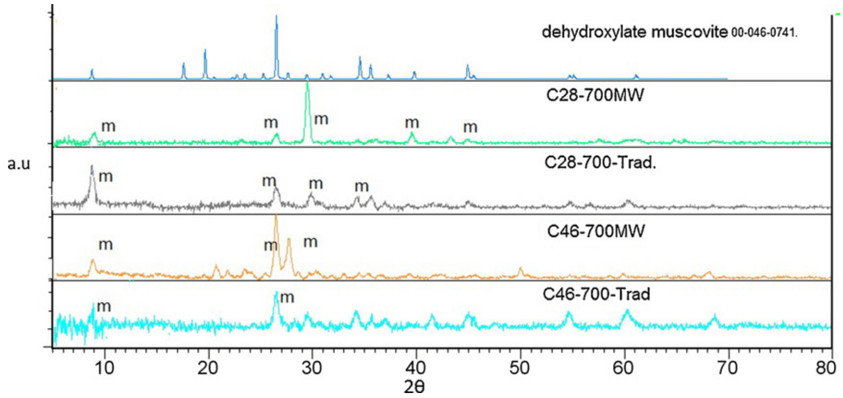

Fig. 6. XRD of C46 and C28 sintered by MW and conventional method at 700 °C.

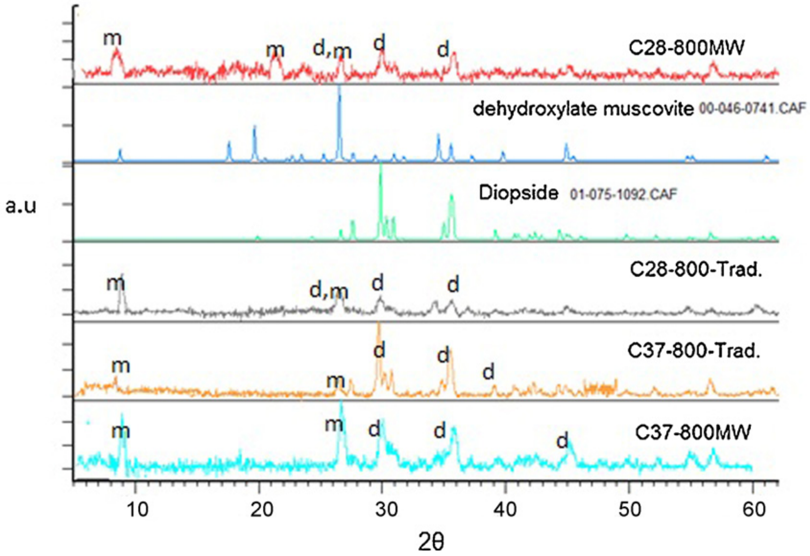

Fig. 7. XRD of C46 and C28 sintered by MW and conventional method at 800 °C.

microwave energy at early stages of heating and increase the temperature. In the case of C28 sample, because of the insufficient, time and temperature of sintering, no increase in densification can be observed.

Diopside was developed in C46 and C28 samples after sintering at 800 °C (Fig. 7), associated with increased viscosity and closed porosity which latter probably reduces the microwave absorption [12].

Fig. 8 shows the crystalline phase-change obtained by heating of samples in MW and furnace. Muscovite was detected in microwave heated samples, whereas at the same conditions, it was not found in specimen heated conventionally which implies that muscovite was dissolved or melted.

Studies on phlogopite thermal behavior have shown that by increasing temperature the phlogopite interlayers will be expanded and, moreover, the weak Van der Waals bonds break and finally the phlogopite melting occurs [13].

By increasing the temperature up to 1000 °C, the peaks of muscovite disappeared in all samples (Fig. 9).

From the above results, it can be postulated that the degradation temperature of muscovite via MW heating is higher than that of conventional heating.

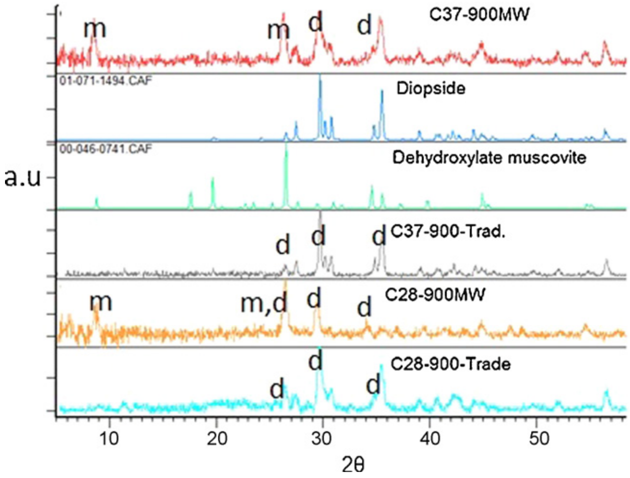

Fig. 8. XRD of C46 and C28 sintered by MW and conventional method at 900 °C.

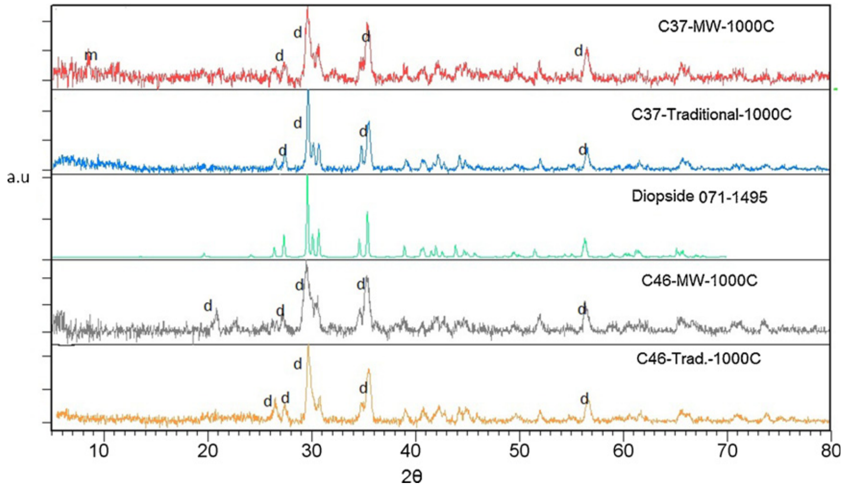

Fig. 9. XRD of C46 and C28 sintered by MW and conventional method at 1000 °C.

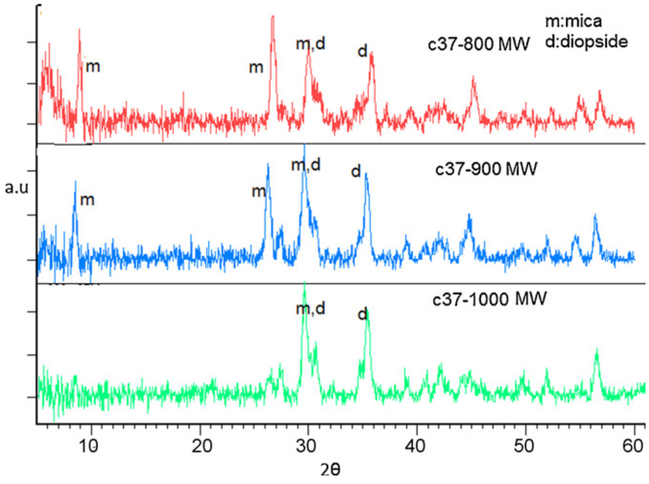

Fig. 10. XRD of C37 heat treated at 800, 900 and 1000 °C by MW heating.

Furthermore, the width and intensity of diopside related peaks were not changed by increasing temperature in C37 at different temperatures (Fig. 10).

On the other hand, according to the XRD results presented in Fig. 10, all the diopside related peaks in MW heated samples are wider than those in conventionally heated ones, indicating the existence of nano-crystalline phase in MW heated samples.

Machinability of sintered samples was also examined and the results for MW sintered samples are presented in Fig. 11. The conventionally sintered specimens were brittle and cracks appeared after machining. Machinability of MW sintered samples can be attributed to the presence of muscovite and diopside phases in glass matrix at high temperatures.

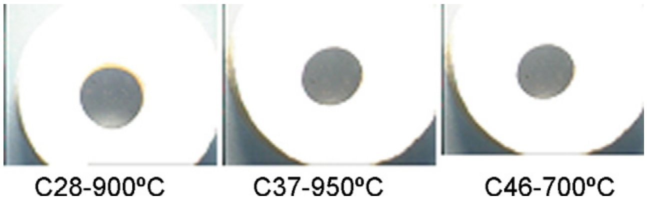
C28-900ºC C37-950ºC C46-700ºC

Fig. 11. Machinable samples sintered by microwave.

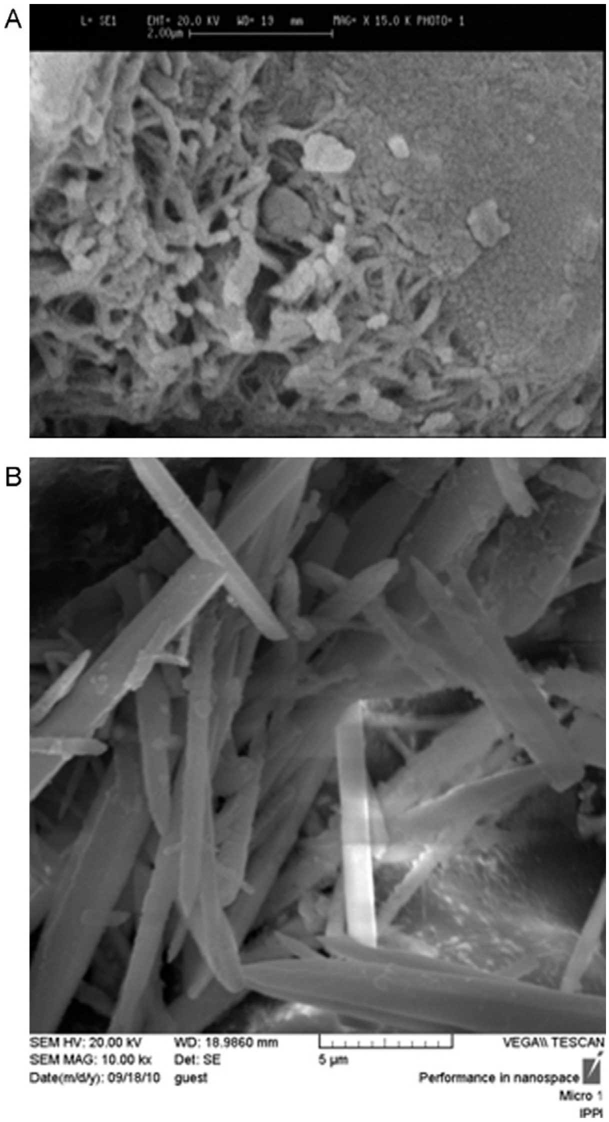

Fig. 12. (a) C37 heat treated at 950 °C by MW. (b) C37 heat treated at 950 °C by conventional method.

As Fig. 12 reveals diopside in MW sintered sample is thinner than conventionally sintered ones. The aspect ratio of crystals in MW sintered samples is almost 10, while in the case of conventional sintered samples, this ratio is 5. It is possible to explain the formation of these crystals by surface-to inside grain growth with glass depletion in the center of the grains which lead to mechanically weak structure [14]. As Fig. 12a shows the fiber-like diopside crystals are approximately 100 nm in diameter precipitated in MW sintered samples; whereas, these crystals are 5 μm in diameter and flake like in conventional sintered samples (Fig. 12b).

4. Conclusion

MW and conventional sintering of phlogopite–glass composites were studied. Results showed that sintering of samples occurred at a lower temperature in microwave heating compared to conventional heating and in the former, crystals were in nanometer size. XRD results also confirmed the diopside as main phase in sintered samples by both methods.

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