

Structural Evolution of YIG during Microwave and Conventional Sintering at 1000 °C

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ABSTRACT. Microwave heating (MH) has emerged in recent years as a new method for sintering a variety of materials against conventional sintering (CH) procedure. However, the phenomenon of microwave-materials interaction is still under debate. In this work, the effect of microwave heating on YIG phase formation was investigated at 1000 °C under the electromagnetic field (2.4 GHz). The phase stability fired samples were qualitative and quantitative analyzed using X-ray diffraction (XRD) techniques. The obtained data were compared with CH to track the phase evolution. XRD revealed MH has produced YIG with 94% faster than CS. The calculated activation energy at this temperature via diffusion control model shows that MH (39.20 kJ/mol) can effectively promote the phase growth rather than CH (90.96 kJ/mol).

Keywords: Microwave heating, Reaction kinetic, Diffusion controlled, Refinement method;

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1. INTRODUCTION

Garnet ceramics with a chemical formula of $A_3B_5O_{12}$ have gained much attention because of their wide range of applications. These include YIG, YAG, and LLZ ceramics which have been investigated in a solid-state laser, wireless communication, storage devices and solar cell applications, recently [1-3]. Owing to their remarkable properties, various synthesizing techniques such as sol-gel route, hydrothermal, chemical vapor deposition (CVD), self-combustion, mechanochemical milling and co-precipitation techniques have been improvised to ensure the final product is in high purity and density. Compared to the above-mentioned techniques, the microwave heating (MH) has been found to be able to boost the production rate at a shorter time (10 times faster than conventional heating, CH) with low energy consumption without deteriorating materials' properties. It is also reported that MH has been effectively densified materials up to 99% [4]. There are many reports on the usage of MH to enhance the garnet properties [3,5-7]. However, due to ponderomotive effect and local thermal effect, most of the final product synthesized via MH was end up with the presence of "unwanted" phases (e.g., secondary phase) [8,9]. These phases in garnet ceramics are

detrimental because they can degrade the device's performance. Therefore, it is important to understand the reaction mechanism and mass transport of these materials so that the presence of any unwanted phases can be prevented. However, the reaction mechanism and mass transport of these materials under MH has not been well clarified in the literature. The study is of necessity as fine garnet's structure with high purity and density can be synthesized quickly.

There are several diffusion models that could explain the kinetic behavior or reactivity of the materials at the solid-state regime. Ginstling-Brounshtein, Dunwald-Wagner, Jander, Valensi-Carter models are among them [10,11]. These models with the aid of Arrhenius plot could portray the cation transport through the reacted layer. In MH, the cation transport is assumed increases since the reaction is induced by the electromagnetic field (EMF). The reaction is related to the ease of moving atoms from their normal sites to neighbouring sites in order to reach a state of chemical equilibrium which in return, resulting in the formation of a new compound [12,13]. For that reason, some insight on diffusion mechanism in MH can be derived by correlating garnet phases and reaction kinetic.

The purpose of this work is to investigate the enhancement of the reaction using microwave heating and measure the conversion rates for YIG at various reaction temperatures and times. The first series of investigation is to determine the mass fraction of the reacted layer followed by developing multi-step reaction model based on the kinetics rate of garnet formation and its disassociation reactions under microwave field.

2. MATERIALS AND METHODS

A standard stoichiometry formulation of garnet was applied; 3:5 of A to B ratio based on $A_3B_5O_{12}$ garnet chemical formula. A mixture of garnet starting precursors was mixed in ethanol- reagent grade (C_2H_6O) to enhance the particle surface contact. The process was assisted by the addition of the cylindrical zirconia ball at 1:10 powder to ball weight ratio. The mixing process has taken place for 8 hours at 25 rpm using Heidoph Overhead Mixer (Reax 2) to form a homogeneous slurry before dried in an oven (Memmert Oven-UM 400) at 80 °C for 24 hours to produce a dried cake. The cake was crushed using agate mortar and later, uniaxially pressed into 13 mm cylindrical pellets.

For reaction kinetic and mass transfer investigation, the compacted pellets were heated from 800 °C to 1250 °C using DAWNYX microwave furnace and were held at various heating rates; 15, 30, 45, 60 min, respectively. The samples were placed in Al_2O_3 crucible while the temperature of the samples was measured using a Pt-Rh thermocouple (B-type thermocouple)-direct touching that is installed and placed in contact with the pellets surface. The characterization of fired samples was carried out using X-ray diffraction analyses (XRD) to study the phase changes. Rietveld refinement method was performed using the Expert Highscore Plus program and the reliability of the refinement result was judged by the goodness of fit (GOF) and the weighted pattern R factor (Rwp).

3. RESULTS AND DISCUSSION

Fig. 1 and Fig. 2 showed the qualitative and quantitative analyses of phases formed at different heating; microwave heating (MH) and conventional heating (CH). All phases presence are matched and corresponded well with ICSD number of 98-000-6274 (Fe_2O_3 , hematite), 98-000-8038 ($YFeO_3$, YIP), 98-005-3764 ($Y_3Fe_5O_{12}$, YIG) and 98-002-3761 (Y_2O_3 , Yttria), respectively. It is worth to note the goodness of fit (GOF) for quantitative analyses as per Fig. 1(b) and Fig. 2(b) obtained are approximate ~1.5 (closer to 1 is an ideal fitting).

It can be seen that YIG phase presence after 10 minutes under microwave heating whereas none of YIG phase was found even after 60 minutes of firing under conventional heating. It has been found that phase

transformation of $5\text{Fe}_2\text{O}_3 + 3\text{Y}_2\text{O}_3 \rightarrow 2\text{Y}_3\text{Fe}_5\text{O}_{12}$ is relatively quick in MH compared to CH. This showed that MH can promote phase transformation at a lower temperature. It was worth noting that this phenomenon was quite different from the conventional heating. In microwave heating, the electromagnetic waves propagate within the sample. The heat generation has caused dipole interaction between Fe_2O_3 and Y_2O_3 to heat and react. This phenomenon is called absorption resonance. This promising result is believed to be the reason behind the accelerated diffusion growth of the phases with the application of the electromagnetic field.

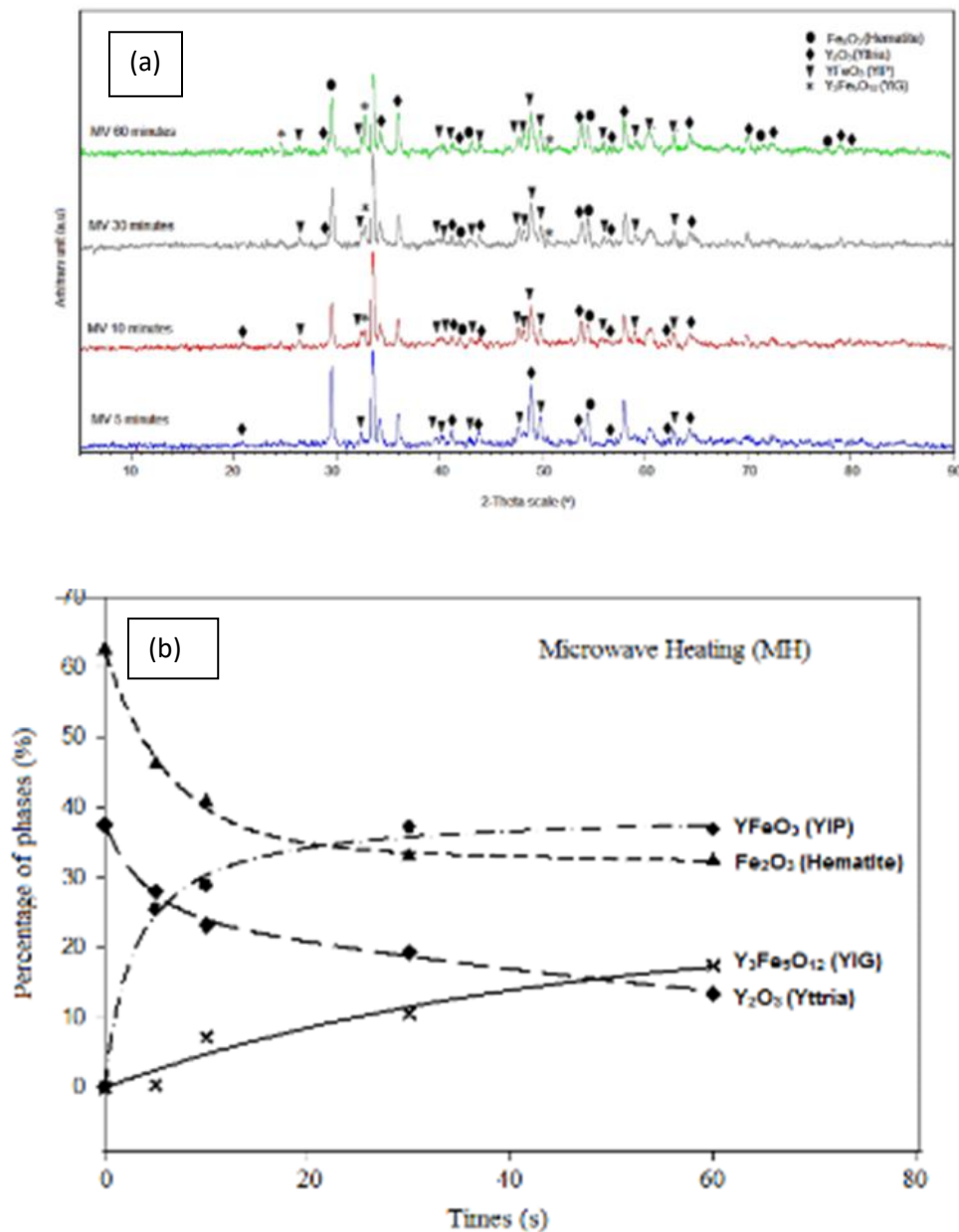


Fig. 1 (a) XRD analysis $\text{Fe}_2\text{O}_3\text{-Y}_2\text{O}_3$ calcined via microwave heating $1000\text{ }^\circ\text{C}$ and (b) quantitative analysis using Rietveld refinement method

Fig. 2 shows that, the less energy surmounted for YIG formation is because of dipolar loss from Y_2O_3 . The oscillating electric fields create agitation of molecular dipoles and have caused the changes in orientations of molecules; subsequently, increases molecular kinetic energy and resulted in the bonding reorientation that caused the formation of YIP and YIG. According to Mehra et al. [2], the formation of unwanted phase; YIP may due to coarser particle size ($>100 \mu m$) of Fe_2O_3 that offers non-uniform heating characteristic than fine particles. In this works, the initial starting raw materials for both oxides are less than $50 \mu m$ which are contradicted with their finding.

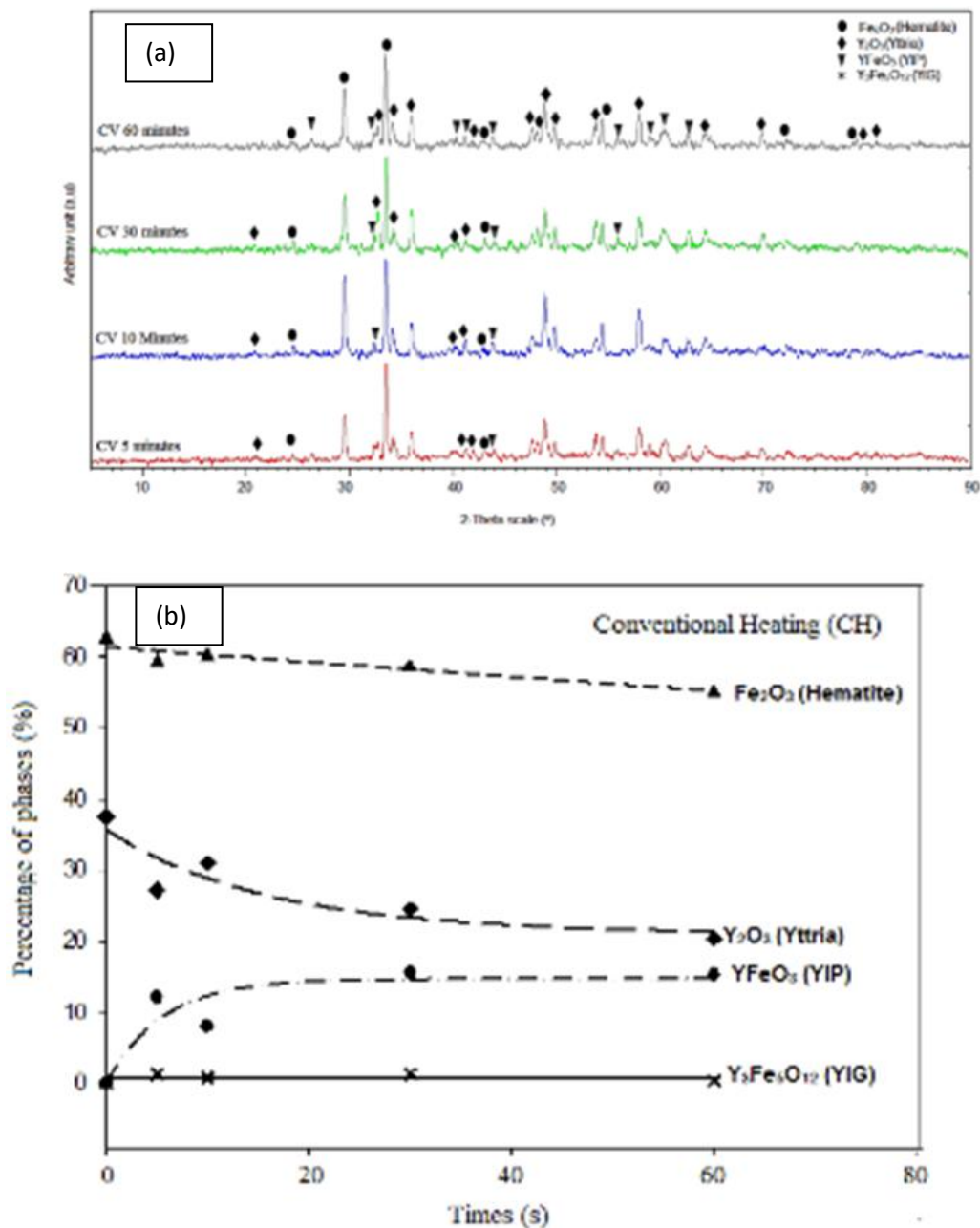


Fig. 2 (a) XRD analysis Fe_2O_3 - Y_2O_3 calcined via conventional heating $1000 \text{ }^\circ\text{C}$ and (b) quantitative analysis using Rietveld refinement method

It is suggested that the formation of YIG is obeying the core-shrinking rule. The reaction between Fe_2O_3 and Y_2O_3 has created an outer layer called YIP. The depth of this layer is assumed to be increased with reaction times and temperature which later inhibit the heat transfer for further reaction with Fe_2O_3 particle core, thus makes YIG difficult to form under conventional heating. This thermally-unstable condition has resulted in the major formation of YIP. Unlike microwave heating, the electromagnetic waves can penetrate YIP layer to enhance the reaction. At each moment of the heating process, the microwave power is absorbed in the samples and increase its core temperature and compensate the concurrent heat losses. It is believed that this microwave effect may also lead to the formation of single phase YIG only if the microwave irradiation is property controlled and well-tailored. The microwave energy has overcome the energy barrier to promote atoms to diffuse.

4. SUMMARY

In the present works, YIG formation mechanism at 1000 °C has been investigated under both microwave heating (MH) and conventional heating (CH). It is found that electromagnetic field has affected the diffusion rate of YIG formation. The calculated activation energy at this temperature via diffusion control model shows that MH (39.20 kJ/mol) can effectively promote the phase growth rather than CH (90.96 kJ/mol). The observed effect is explained by both preferential microwave absorption and enhanced mass transport between Fe_2O_3 and Y_2O_3 particles.

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