UNIVERSITI PUTRA MALAYSIA

MICROSTRUCTURE AND DIELECTRIC PROPERTIES OF YTTRIUM IRON GARNET-STRONTIUM TITANATE BI-PLATES JUNCTIONS AND COMPOSITES JUNCTIONS

WONG SWEE YIN

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By

WONG SWEE YIN

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January 2017

Supervisor : Jumiah Hassan, PhD
Faculty : Science

In this project, the investigation towards morphology and dielectric properties of single sample Strontium Titanate (ST) and Yttrium Iron Garnet (YIG), ST-YIG composite slabs (SYCS) and ST-YIG composites (SYC) were carried out. This is a pioneer work since no reports or studies were found on dielectric-magnetic slabs.

Each slab is made up of ST and YIG sample where for one slab, the samples were prepared via mechanical alloying (MA) method while the other were prepared via conventional solid state (SS) method. X-ray Diffraction (XRD) was employed for phase identification and purity of the samples. Since the ST-YIG composites prepared were multiphase, Rietveld refinement method was used to estimate the phase composition in each sample. MA method successfully reduced the sintering temperature for the reaction to occur at a much lower temperature compare to SS method. The surfaces of the sample were visualized using Field emission scanning electron microscopy (FESEM) and the average grain size was calculated. From FESEM images, MA method produced very fine particles in nano-scale while SS method in micro-scale.

The slabs were sintered at 1300^{o}C with different sintering hours from 10 hours to 20 hours at intervals of 2 hours for SS method whereas for MA method, it was sintered at 1200^{o}C. The samples for the slab were co-joined strongly through diffusional processes where no welding was involved. The junction properties were studied. FESEM images were also taken of the cross-sectional area of the slab and the diffusion of ST into YIG and vice versa can be observed at the junction. This only occurs for the slab prepared via SS method. The diffusion depth of both samples of the composite slab was determined via EDX. The distance at the junction where the ions moved from one compound to the other during the diffusion process is calculated. Movement of ions in the samples is dependent on the different sintering hours. However, for the slab

prepared via MA method, a slight diffusion occurs but the samples do not stick together. This is probably due to the continuous network of grain in YIG which only indicated the initial stage of sintering. The increased on migration of grain boundary increased the driving force for neck growth. Since surface diffusion and lattice diffusion dominated, therefore no interdiffusion process happened on the surface contact of ST and YIG. Thus, interstitial lattice diffusion among ST and YIG were difficult to obtain.

To understand the charge stored and energy dissipated, the dielectric properties of samples were investigated. Since the samples are multi-phase, hence the dielectric constant obtained is a contribution from different phases. Generally, the dielectric constant of the samples increases with the rise in temperature and decreases with frequency. The samples prepared via SS method have the best dielectric properties among the samples possibly due to the fine grain size of ST with large grain size of YIG produced denser grains by increasing the insulating grain boundary volume. For the slabs prepared via SS method, the dielectric constant and dielectric loss factor follows the trend of YIG rather than ST. The slabs produced better dielectric properties with higher value of dielectric constant and lower dielectric loss factor compared with single sample YIG. Therefore, it may be useful in microwave applications such as tunable HTS (high temperature superconducting) microwave filters. Analysis using complex modulus was also done on the samples to verify the polarization mechanism involved in the samples and slabs.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

MIKROSTRUKTUR DAN SIFAT DIELEKTRIK BAGI SIMPANGAN DWI-LAPISAN YTTRIUM IRON GARNET-STRONTIUM TITANATE DAN SIMPANGAN KOMPOSIT

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Dalam projek ini, siasatan terhadap morfologi dan sifat dielektrik kepada Strontium Titanate dan Yttrium Iron Garnet sampel tunggal, papak komposit ST-YIG (SYCS) dan komposit ST-YIG (SYC) telah dijalankan. Ini adalah kerja perintis di mana tiada siasatan pernah dilakukan terhadap papak dielektrik-magnetik.

Setiap papak terdiri daripada ST dan YIG sampel di mana untuk satu papak, sampel telah disediakan melalui kaedah mekanikal pengaloian (MA) manakala yang lain telah disediakan melalui kaedah konvensional keadaan pepejal (SS). X-ray Diffraction (XRD) telah digunakan untuk mengenal pasti fasa dan ketulenan sampel. Oleh kerana sampel-sampel yang disediakan adalah berbilang fasa, maka kaedah penyempurnaan Rietveld telah digunakan untuk menganggarkan komposisi fasa dalam setiap sampel. Kaedah MA berjaya meningkatkan kereaktifan serbuk campuran dengan menurunkan suhu pensinteraan untuk tindak balas berlaku berbanding dengan kaedah SS.

Papak telah disinterkan pada 1300^{o}C dengan masa pensinteran yang berbeza dari 10 jam hingga 20 jam pada selang 2 jam untuk kaedah SS manakala untuk kaedah MA, ia disinterkan pada 1200^{o}C. Sampel papak telah bercantum dengan kuat melalui proses resapan di mana tiada kimpalan terlibat. Sifat simpangan telah dikaji. Imej FESEM juga diambil daripada kawasan keratan rentas papak dan peresapan ST ke YIG dan sebaliknya boleh diperhatikan di persimpangannya. Ini hanya berlaku bagi papak yang disediakan melalui kaedah SS. Kedalaman peresapan bagi kedua-dua sampel papak komposit telah ditentukan melalui EDX. Jarak di persimpangan di mana ion bergerak dari satu kawasan ke kawasan yang lain semasa proses peresapan dapat dikira. Pergerakan ion dalam sampel adalah bergantung kepada masa pensinteran yang berbeza. Walau bagaimanapun, bagi papak yang disediakan melalui kaedah MA, peresapan hanya berlaku sedikit tetapi sampel tidak bercantum bersama-sama. Ini mungkin disebabkan oleh rangkaian berterusan gandum YIG yang hanya menunjukkan peringkat

awal pensinteran. Peningkatan penghijrahan bagi sempadan bijian meningkatkan daya penggerak bagi pertumbuhan leher. Oleh sebab peresapan permukaan dan peresapan kekisi dikuasai, maka tiada proses peresapan dalaman berlaku pada sentuhan permukaan ST dan YIG. Oleh itu, peresapan kekisi pada celahan antara ST dan YIG sukar didapati.

Untuk memahami pengumpulan cas dan tenaga lesapan, sifat-sifat dielektrik sampel telah disiasat. Oleh sebab sampel mengandungi pelbagai fasa, maka pemalar dielektrik yang diperolehi adalah sumbangan daripada fasa yang berbeza. Secara umumnya, pemalar dielektrik sampel meningkat dengan kenaikan suhu dan berkurang dengan kekerapan. Sampel disediakan melalui kaedah SS mempunyai sifat dielektrik yang terbaik di kalangan sampel mungkin disebabkan oleh saiz butiran halus ST dengan saiz butiran besar YIG lalu menghasilkan bijirin padat dengan meningkatkan jumlah sempadan bijian penebat. Bagi papak yang disediakan melalui kaedah SS, pemalar dielektrik dan faktor kehilangan dielektrik mengikuti trend YIG bukannya ST. Papak yang dihasilkan mempunyai sifat dielektrik yang lebih baik dengan nilai yang lebih tinggi daripada pemalar dielektrik dan faktor kehilangan dielektrik yang lebih rendah berbanding dengan sampel YIG tunggal. Oleh itu, ia mungkin berguna dalam aplikasi gelombang mikro seperti HTS merdu (suhu tinggi superkonduktor) penapis gelombang mikro. Analisis terhadap modulus kompleks juga telah dijalankan ke atas sampel untuk mengesahkan mekanisme polarisasi yang terlibat dalam sampel dan papak.

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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LIST OF SYMBOLS

C	Capacitance
E	Electric field
Q	Magnitude of total charge
V	Potential difference
A	Area of each plate or cross-sectional area
d	Separation between the plates
C_o	Capacitance of parallel plates in vacuum
ε_0	Permittivity of free space
ε^*	Complex permittivity
ε	Permittivity of material
ε'_r	Relative permittivity
ε''_r	Relative dielectric loss factor
D	Displacement vector
P	Polarization

LIST OF ABBREVIATIONS

SS	Conventional solid state
MA	Mechanical alloying
ST	Strontium Titanate
YIG	Yttrium Iron Garnet
XRD	X-ray diffraction
FESEM	Field Emission Scanning Electron Microscope
EDX	Energy Dispersive X-ray Spectroscopy
ICSD	Inorganic Crystal Structure Database

CHAPTER 1

INTRODUCTION

1.1 Background of Project

In this project, a new development of diffusional change and the dielectric behavior of dielectric-magnetic composite slabs were studied. The samples were prepared using different preparation methods, conventional solid state method and mechanical alloying method.

Each preparation method produced a series of samples with different sintering hours, 10 hours - 20 hours at 2 hours intervals. The samples were Strontium Titanate (ST) and Yttrium Iron Garnet (YIG). ST was sintered at 1200 ℃ and 1000 ℃ respectively while YIG was sintered at 1400 ℃ and 1200 ℃ respectively for conventional solid state method and mechanical alloying method. Both samples were sandwiched together and sintered at a specific sintering temperature, 1300 ℃ and 1200 ℃ respectively with different sintering hours.

The purification and crystallinity of the samples were characterized using X-ray Diffraction (XRD), while the surface morphology and the diffusion depths were characterized using Field Emission Scanning Electron Microscope (FESEM) and Energy Dispersive X-ray Spectroscopy (EDX). The dielectric behavior of the samples was measured using Agilent High Resolution Analyzer at frequency ranges, 10 mHz - 1 MHz and Agilent 4294A Precision Impedance Analyzer at frequency ranges, 40 Hz - 10 MHz at different measuring temperatures, 30°C, 50°C - 250°C at 25°C intervals.

1.2 Problem Statement

In the past researches, there were some investigations and reports made on the diffusional and dielectric properties at the junction of the dielectric-dielectric composites slab. However, studies on the dielectric-magnetic composite slab have not been reported.

Diffusion process which occurred in solid state materials were studied especially for samples prepared using the sputtering technique, but diffusion process for samples prepared by conventional solid state method has not been investigated.

Dielectric behavior is usually studied in insulating materials, such as ceramics, glass, and etc. Based on past researches, ceramics which have high dielectric constant will also have high dielectric loss factor at high frequency range. Materials which exhibit high dielectric constant and low dielectric loss factor at high frequency range have not been developed.

1.3 Benefit of Research

In this project, there are some new findings which have not been discovered in previous works. Dielectric-magnetic composite slab is a new composite which may produce both dielectric and magnetic behavior at the same time. These new composite slabs were prepared by conventional solid state method and mechanical alloying method.

Grain growth and diffusion process for both dielectric and magnetic composite slab were analyzed at the junction of the composite slab. The dielectric behavior of the single sample and composite slab were analyzed and comparison between their properties was made. The morphological and diffusional changes and the dielectric behavior within the dielectric-magnetic bi-plate junctions were determined.

In addition, the dielectric-magnetic composite slabs are useful especially in electronic industry. It can be used as power insulation and charge storage in industry. Besides, it can also be used in microwave applications such as tunable HTS (high temperature superconducting) microwave filters.

1.4 Conventional Solid State Method (SS)

Conventional solid state method (SS) is a traditional method where two or more different starting materials will react together after heating to form the required products. This method which involves of mixing different powders is common to use to prepare a wide range of ceramic products, such as metal oxides, titanates, ferrites, silicates, and etc (Rahaman, 2003).

Conventional solid state method is a method which is widely used in preparation of polycrystalline ceramics. This method implicates the consolidation of fine powders to form a green body which is then sintered to produce a dense product with heterogeneous microstructure. These can be prepared by pressing it in pellet form. Solid state reaction relies on several parameters, such as size and shape of the particles, uniformity of the mixing, sintering temperature and time, and etc (Rahaman, 2003).

Solid state sintering resulted in atomic diffusion which leads to densification of the grains or coarsening of the microstructure. Changes in density, grain size, and pore size show the behavior on densification of grains. Coarsening process also undergoes the same changes as densification except pore size. Pores shrink during densification, while pores grow during coarsening. Moreover, surface diffusion, volume diffusion, grain boundary diffusion, viscous flow, plastic flow, and vapor transport from solid surfaces also contribute to the sintering process (Rahaman, 2003).

There are advantages and disadvantages using this method. This method can be used to prepare a large amount of the required products. High purification on the starting materials is not required and the cost of production is low (C. Barry Carter, M. Grant Norton, 2007). However, contamination of powder with impurities may happen during

the grinding process. Besides, particle size of powders is hard to control. This may produce chemically inhomogeneous product with complex microstructures. (Rahaman, 2003). Furthermore, high sintering temperature (above 1000 ℃) is required for the materials to undergo chemical reaction among the reactants to produce the product.

1.5 Mechanical Alloying Method (MA)

Mechanical alloying method (MA) is a method which is similar to the conventional solid state method excluding the preparation of a green body. Mechanical alloying is a technique where two or more different compounds are mixed and milled together in a small high-speed shaker in dry condition. The mixture which forms solid solution, intermetallic, or amorphous phase after milling is a homogeneous alloy. These means material transfer is involved in this method (Suryanarayana, 2004).

Besides, this method induces chemical reactions during the milling process, such as mechanochemical reactions. This method can be used to prepare a variety of powders, such as oxides, carbides, nitrides, borides, and silicides (Rahaman, 2003). The important physical parameters required in this method are the raw materials, the mill, and the process variables. Raw materials normally have particle size in the range of 1-200 μm, which should be smaller than the size of the grinding ball. This method is carried out using SPEX SamplePrep 8000D Mixer/Mill, which is able to contain 8-20 g of the powder at a time depending on the size of the vials. The vials are clamped and swung energetically back and forth for several thousand times a minute. The grinding balls which are put together with the powder will collide with the powder and the vials. This technique is considered as high-energy milling due to the high velocities of grinding balls and the high speed of clamp motion.

There are three possibilities based on the mechanism of mechanochemical reaction. The first possibility may be due to the solid state diffusion mechanism. During the milling process, a slightly increase in temperature in the mill lower the activation energy of the mixtures and therefore leads to diffusion process. Since heating of the mill occurs, the sintering temperature for the mixtures is obviously lower than that as compared to solid state method. The second possibility may be due to the local melting during the milling process where exothermic reaction occurs and releases heat to the surrounding. The third possibility may cause by a form of self-propagating process at high temperature. In highly exothermic reaction, the heat that released sustains the reaction.

The advantage of using this method is simple preparation method especially for silicides and carbides which have a narrow compositional range which are hardly to produce and need to sinter at high temperatures. Besides, this method can produce alloy at low temperature without melting them. This method is able to reduce the particle size in large dimension to nanometer. Conversely, contamination may occur due to the integration of impurities from the mill and milling medium into the powders. Besides, the temperature change in the mill during the milling process is hard to control (Rahaman, 2003).

1.6 Scope of Research

Strontium Titanate is one of the famous ferroelectric materials which exhibit large dielectric constant for sintered ceramics while Yttrium Iron Garnet is a kind of soft ferromagnetic materials which exhibit high Q factor in microwave applications. The dielectric-magnetic composite slabs (Strontium Titanate-Yttrium Iron Garnets composite slabs) and dielectric-magnetic composite (Strontium Titanate-Yttrium Iron Garnets composite) were prepared by conventional solid state method and mechanical alloying method. Each composite slabs and composites were sintered at 1300℃ and 1200℃ for 10 hours respectively. The dielectric properties of the samples were measured using Agilent High Resolution Analyzer at frequency range of 10 mHz to 1 MHz and Agilent 4294A Precision Impedance Analyzer at frequency range of 40 Hz to 10 MHz at 30°C, 50°C - 250°C at 25°C intervals. The morphological and diffusional changes and the dielectric behavior within the dielectric-magnetic bi-plate junctions offer the advancement of new knowledge in this multilayer composite.

1.7 Objectives

The objectives of the present study were developed as follows:

1. To prepare Strontium Titanate (ST), Yttrium Iron Garnet (YIG), Strontium Titanate-Yttrium Iron Garnets composites slab (SYCS), and Strontium Titanate-Yttrium Iron Garnets composites (SYC) via conventional solid state method and mechanical alloying method and characterize their crystallinity, surface morphology and diffusional properties of the bi-plate junctions via XRD, FESEM including EDX and other morphological study techniques.
2. To characterize the dielectric behavior for single samples, composite slabs and composites at different measuring temperatures.
3. To understand and explain the effects of morphological and diffusional changes and the dielectric behavior of the junctions in the composite slabs.

Therefore, the present study required to respond to the following research questions:

1. Would ST, YIG, SYCS, and SYC successfully prepared via conventional solid state method and mechanical alloying method?
2. Would XRD, FESEM including EDX and other morphological study techniques analyze the samples' crystallinity, surface morphology and diffusional properties of the bi-plate junctions?
3. Would the dielectric behavior for single samples, composite slabs and composites been analyzed at different measuring temperatures?
4. Would the effects of morphological and diffusional changes and the dielectric behavior of the junctions in the composite slabs been explained?

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