UNIVERSITI PUTRA MALAYSIA

PHYSICAL AND OPTICAL CHARACTERIZATION OF YTTRIUM ALUMINUM MONOCLINIC PREPARED VIA CITRATE-NITRATE SOL-GEL AND MECHANICAL ALLOYING METHOD

MOHD KHAIRUL IKHWAN BIN MOHD ZAWAWI

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MOHD KHAIRUL IKHWAN BIN MOHD ZAWAWI

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirement for the Degree of Master of Science

July 2014

DEDICATIONS

TO

MY FAMILY,

MY TEACHERS

AND

MY FRIENDS

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Master of Science

PHYSICAL AND OPTICAL CHARACTERIZATION OF YTTRIUM ALUMINUM MONOCLINIC PREPARED VIA CITRATE-NITRATE SOL-GEL AND MECHANICAL ALLOYING METHOD

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MOHD KHAIRUL IKHWAN BIN MOHD ZAWAWI

July 2014

Supervisor: Raba'ah Syahidah Azis, PhD
Faculty: Science

Obtaining single-phase polycrystalline YAM (yttrium aluminum monoclinic, $Y_4Al_2O_9$) has been a difficult effort since YAG (yttrium aluminum garnet, $Y_3Al_5O_{12}$) and YAP (yttrium aluminum perovskite, $YAlO_3$) also invariably present as second phase. YAM, unlike the other two compounds, was extensively studied for its application as scintillators, phosphors and as a laser hosts due to possess of garnet and perovskite structure. The investigation on the YAM itself is considered poor and lacking because very few reports have been published regarding this material even though YAM was discovered along with YAG and YAP.

Hence, for the first time sol-gel citrate-nitrate combustion process (SGCNCT) and mechanical alloying (MA) are to be employed in an attempt to achieve the desired pure single phase. The SGCNCT of yttrium and aluminum precursor in a 2/1 ratio directly forms YAM without going through a transition phase. The polycrystalline YAM powders obtained by this process were characterized by DTA/TG, XRD, ^{27}Al NMR, FTIR and UV-VIS spectrometer for the analysis of structural and optical properties.

Fully dispersible, unaggregated polycrystalline YAM powders were obtained after heat treatment at 740 ^{o}C for 10 h. The formation temperature of the YAM single phase was found to be much lower (700 ^{o}C) than that at the conventional solid state reaction which required high temperature around 1917 ^{o}C. In the normal sol-gel (SG) process, citric acid forms numerous tiny enclosures that have the constituent cations at the molecular level, leading to a reduction of the diffusion length and the enhancement of the reactivity of the precursors. The reactivity seems to have been further greatly enhanced with the inclusion of the combustion step in the sol-gel process. Hence the SGCNCT process yields pure YAM while the SG process alone has always yielded mixed phase. However, it is difficult to synthesize the YAM phase at lower temperature by the MA process as the reaction between oxides occurs at higher temperature. From the optical analysis, it was confirmed that the YAM behaves as insulator as it has the energy band gap value higher than 3 eV.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

PENCIRIAN FIZIKAL DAN OPTIK YTTRIUM ALUMINIUM MONOKLINIK DIHASILKAN MELALUI TEKNIK SOL-GEL SITRAT-NITRAT DAN PENGALOIAN MEKANIK

Oleh

MOHD KHAIRUL IKHWAN BIN MOHD ZAWAWI

Julai 2014

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Mendapatkan fasa tunggal polihabluran YAM (yttrium aluminium monoklinik, $Y_4Al_2O_9$) merupakan satu usaha sukar kerana YAG (yttrium aluminium garnet, $Y_3Al_5O_{12}$) dan YAP (perovskit aluminium yttrium, $YAlO_3$) sentiasa hadir sebagai fasa kedua. YAM tidak seperti dua sebatian ; YAG dan YAP yang telah dikaji dengan meluas untuk aplikasi sebagai scintillators, fosfor dan sebagai laser host kerana memiliki struktur garnet dan perovskit. Kajian mengenai YAM dianggap kurang kerana sangat sedikit laporan mengenai bahan ini telah diterbitkan walaupun YAM ditemui bersama-sama dengan YAG dan YAP
.
Oleh itu, buat pertama kali sol-gel sitrat - nitrat proses pembakaran (SGCNCT) dan pengaloian mekanikal (MA) akan digunakan dalam cubaan untuk mendapatkan satu fasa tulen yang dikehendaki. SGCNCT pada yttrium dan aluminium prekursor dalam nisbah 2/1 membentuk YAM secara langsung tanpa melalui fasa peralihan. Serbuk polihabluran YAM diperolehi daripada proses ini telah dicirikan oleh DTA/TG, XRD, NMR, FTIR dan UV-VIS spektrometer untuk analisis struktur dan sifat optik.

Polihabluran serbuk YAM yang sepenuhnya tersebar dan tidak agregat diperolehi selepas pemanasan pada 740 ^{o}C selama 10 j . Suhu pembentukan fasa tunggal YAM didapati (700 ^{o}C) jauh lebih rendah daripada tindak balas keadaan pepejal konvensional yang memerlukan suhu yang tinggi sekitar 1917 ^{o}C. Dalam proses sol -gel (SG) biasa, asid sitrik membentuk banyak kurungan kecil yang mempunyai juzuk kation pada peringkat molekul, yang membawa kepada pengurangan kepanjangan resapan dan peningkatan kereaktifan prekursor. Kereaktifan ini seolah-olah telah dipertingkatkan lagi dengan kemasukan langkah pembakaran dalam proses sol-gel. Dengan itu, proses SGCNCT menghasilkan YAM tulen manakala proses SG sendirian sentiasa menghasilkan fasa campuran. Walau bagaimanapun, adalah amat sukar untuk mensintesis fasa YAM pada suhu rendah melalui proses MA kerana tindak balas antara oksida berlaku pada suhu yang amat tinggi. Daripada analisis optik, ia telah disahkan bahawa YAM bersifat penebat kerana ia mempunyai nilai jalur tenaga lebih tinggi daripada 3 eV.

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I certify that a Thesis Examination Committee has met on 7 July 2014 to conduct the final examination of Mohd Khairul Ikhwan bin Mohd Zawawi on his thesis entitled "Physical and Optical Characterization of Yttrium Aluminum Monoclinic Prepared Via Citrate-Nitrate Sol-Gel and Mechanical Alloying Method" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

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Name of Chairman of Supervisory Committee:	Raba'ah Syahidah Azis	Name of Member of Supervisory Committee:	Mansor Hashim

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

C_Q	Coupling constant
DTA	Differential Thermal Analysis
FT-IR	Fourier transform infrared analysis
HEBM	High Energy Ball Milling
MA	Mechanical Alloying
MAS	Magic angle spinning
MQMAS	Multi quantum magic angle spinning
NMR	Nuclear Magnetic Resonance
SG	Sol-Gel
SGCNCT	Sol-Gel Citrate-Nitrate Combustion Technique
TG	Thermogravimetriy Analysis
V_Q	Larmor frequency
XRD	X-Ray Diffraction
YAG	Yttrium Aluminum Garnet
YAM	Yttrium Aluminum Monoclinic
YAP	Yttrium Aluminum Perovskite
δ_{iso}	Chemical shift
η_Q	Asymmetry parameter

CHAPTER 1

INTRODUCTION

1.1 Introduction

The production of the powders which consists of good quality and composition are crucial in the ceramics manufacturing industry, moreover for the development of the high tech electronics application. In the United States, the total market for powders of advance ceramics (e.g., electronics and structural ceramics) alone is around $1 billion per year (Carter and Norton, 2013a). Nowadays, the use of ceramic materials both natural and fabricated has increased significantly since they have been applied for numerous applications. To tailor ceramics for a wide range of properties required a better understanding of chemical composition, micro-structure as well as processing methods (Marchal, 2008).

Many processing methods are available for preparing ceramic powders which can be divided into just three basic types (Carter and Norton, 2013a):

- Mechanical
- Chemical
- Vapor phase

Mechanical methods are subjected to a series of processes called as comminution, in which solid materials are reduced in size, by crushing, grinding and milling. This powders production method is widely used in the industry due to ability for mass production of the product. Nonetheless, the powders produced by this method have low purity.

Chemical methods such as sol-gel processing always come up with several advantages over the mechanical methods. The ability to produce samples at lower temperature, control particle morphology and purity always make the sol-gel process is the best processing method for structural study of the ceramic materials. Because of this ability, this chemical processes are widely used in the production of advance ceramics materials such as laser, semiconductor, etc.

The other technique used to produce ceramic powders is vapor-phase process. This method offers many advantages, such as the ability to produce high purity powders, discrete and nonaggregated particles, nanoparticles with narrow size distributions and versatility in producing powders of oxides and nonoxides but it tend to be more expensive compared to others.

In the following we first discuss the history of the Y_2O_3-Al_2O_3 system, their potential application and the stability ranges of compounds in the system. Then, we introduce the research questions and the objective of the research. The discussion related to the previous work on the synthesis the compound within the Y_2O_3-Al_2O_3 system, sol-gel citrate-nitrate combustion process and its important parameters and the literature on ^{27}Al NMR in Y_2O_3-Al_2O_3 system presented in chapter two. Materials and methods used in this experiment were discussed in chapter three. Results and discussion were discussed in chapter four. Chapter five summarizes and concludes the research findings, in addition to some suggested recommendations.

1.2 History of Y_2O_3-Al_2O_3 system

According to the literature, there are three important intermediate compounds known to exist in the Y_2O_3-Al_2O_3 system. These compounds correspond to the compositions of 3Y:5Al ($Y_3Al_5O_{12}$), Y: Al ($YAlO_3$) and 2Y: Al ($Y_4Al_2O_9$) (Cockayne, 1985; Abel et al., 1974). The way of nomenclature of these three compounds are a little confusing because they are used to be referred by their crystal structures so the 3Y:5Al and Y: Al becomes yttrium aluminium garnet and yttrium aluminium perovskite with the acronyms YAG and YAP respectively. Then, when it comes to give a name to the 2Y:Al as yttrium aluminium monoclinic, shortened as YAM, it looks inconsistent with the previous two compounds since the perovskite and garnet terms are derived from geological counterparts and nonesuch derivation exists for a monoclinic term (Cockayne, 1985). However those nomenclatures seem to be accepted and repeatedly found in use in the articles.

Ceramics based on Y_2O_3-Al_2O_3 system is a promising material in semiconductor technology and also have been applied for numerous advanced applications (Gowda, 1986). For instance, the YAG doped with Nd is widely used as a lasing medium for solid state laser. In addition, the rare earth doped with YAP is also an excellent material for phosphor use in the development of display devices (You et al., 2012). YAP is extensively used as gain media, scintillator and acousto-optic (Medraj et al., 2006). Finally, the least studied material which is YAM are suggested to have potential applications in the field of advance plasma display panel (Yadav et al., 2009) and as thermal barrier ceramic (Zhan et al., 2012) due to excellent short decay time, photoluminescence and chromaticity and very low theoretical and experimental thermal conductivities.

There are many ways that can be employed to synthesize the material within this ternary system. The most common one is the solid state method. Basically, extensive mechanical mixing and high heating temperature is required in order to form the material since the alumina and yttrium oxides are the materials which have high hardness value and high melting point. Other than conventional process, Czochralski method (produce single crystal YAM) and wet chemical process such as sol-gel, micro emulsion, co-precipitation etc. also can be used to prepare the polycrystalline material.

The stability ranges of compounds found in the Y_2O_3-Al_2O_3 system have been reviewed and elucidated by several groups. Different view and argument have been expressed regarding the stability region. Many phase diagrams have been developed in order to express and explain the stability of these intermediate phases. One of the earliest phase diagrams were reported by Schneider (1961) who showed the presence of three compounds which are YAG and YAM over the temperature range between 1000 ^oC-1800 ^oC and for YAP at temperature above 1800 ^oC. However, Warshaw and Roy (1959) did not include YAP in their diagrams and they are also did not give any distinct comment on the YAP stability. According to Toropov et al., (1964) YAP phase has very limited stability due to formation with YAM and liquid at 1875 ^{o}C and decomposition into YAM and YAG phases at 1835 ^{o}C. Noguchi (1967) suggested that YAP was a congruently melting phase and agree with Toropov et al., (1964) explanation regarding the phase decomposition. The idea about the stable congruent melting on YAP was confirmed by Class (1968) and many other groups who melt growth of a large single crystal using a float-zoning and Czochralski process respectively.

The phase diagram produced by Abel et al., (1974) included the YAP phase, as the single crystal produced was sufficiently stable during cooling to room temperature and within the exacting thermo mechanical environment of a laser cavity. On the contrary with YAP, YAM has been shown on all phase diagrams published for the Y_2O_3-Al_2O_3 system. However, several studies noticed that YAM phases become unstable below 1000 ^{o}C and the single crystal produced cracked destructively when cooling to 1000 ^{o}C. Toropov et al. (1964) suggest that the crack on the single crystal produced due to polymorphic transformation whilst others believe due to the decomposition of YAM into YAG and alumina. The only unambiguously stable phase in the system is the YAG phase. The recent version of phase diagram was produced by Roth (1995) as illustrated in Figure 1.1.

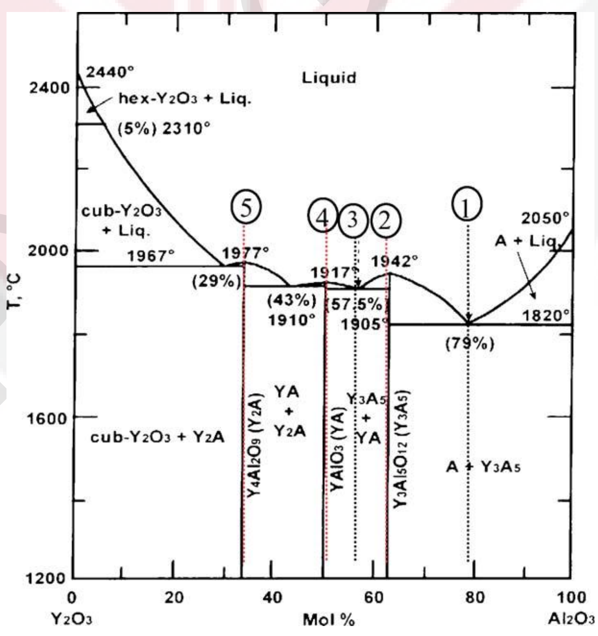

Figure 1.1: Phase diagram of Y_2O_3-Al_2O_3 system (Roth, 1995)

1.3 Scope of Study

The scope of the research focused on preparing high purity YAM powders. Two different synthesized method for preparing the YAM, i.e. SGCNCT and MA which employed in the experiment was explained in this dissertation. This research only investigates the structural and optical behavior of YAM. Apart from that, there were no report on electrical or mechanical properties of YAM because it beyond our scope of study.

1.4 Problems Statement

In Y_2O_3-Al_2O_3 system, there are three crystalline phases: YAG, YAP and YAM. The first two are extensively studied and reported. Instead, YAM phase is less reported and studied due to difficulty in the producing the materials. However, there are several methods available for producing the YAG and YAP phases such as solid-state method, melt growth and chemical method which could be used to fabricate the polycrystalline YAM phase. The question is what would be the best method for producing the pure YAM phase. What would be the optimum temperature to produce pure YAM? Is there any transition phase occurring during the development process as occurred when growing single crystal YAM. Can the method be used to produce single phase YAM? To provide answers to some of these questions, the SGCNCT synthesis, instead of well-known SG synthesis is to be employed, since the normal SG process has not been shown to be able to yield single phase polycrystalline YAM at lower temperature.

1.5 Objectives

The main goal of this research is to obtain single phase of polycrystalline YAM powder. To obtain these materials, the study need to be embarks on the following objectives:

1- To study the structural of YAM prepared by sol-gel citrate-nitrate combustion and mechanical alloying method.
2- To determine the optical behavior of the YAM by measuring the energy band gap

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