# SOL-GEL PREPARATION OF LITHIUM ALUMINOSILICATE POWDER

## (FUMED SILICA AS SILICA SOURCE)

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Penyediaan serbuk litium aluminium silikat dengan menggunakan kaedah sol-gel
(fumed silika sebagai sumber silika)

## ABSTRAK

Fumed silika telah digunakan sebagai sumber silika untuk sintesiskan serbuk litium aluminium silikat dalam proses sol-gel. Hasilan serbuk gel telah dikalsinat pada suhu 400 °C, 600 °C, 800 °C and 1000 °C. Selepas kalsinasi, serbuk litium aluminium silikat yang diperlukan telah dianalisis dengan menggunakan Fourier transfom inframerah (FTIR) spektroskopi dan pengimbasan mikroskop elektron (SEM). Kaedah penyediaan bi-komponen and tri-komponen telah digunakan untuk menghasilkan serbuk litium aluminium silikat. Berdasarkan keputusan FTIR, ikatan Si-O-Si telah muncul di rantau 1083.95 cm$^{-1}$ bagi sampel yang telah dikalsinat pada suhu 400 °C. Sampel-sampel yang telah dikalsinat pada suhu 600 °C, 800 °C dan 1000 °C menunjukkan pembentukan ikatan Si-O-Al. Berdasarkan empat IR spektrum, β-eucryptite telah muncul di antara rantau 1005cm$^{-1}$, 750 cm$^{-1}$ dan 670cm$^{-1}$. Berdasarkan keputusan SEM, sampel litium aluminium silikat menjadi lebih padat dan bergabung bersama apabila suhu menaik. Phenomenan ini disebabkan oleh penyingkiran air.

# ABSTRACT

Fumed silica ($SiO_2$) was used as silica source to synthesize lithium aluminosilicate powders (LAS) in sol-gel process. The gel-powders obtained were calcined at 400 °C, 600 °C, 800 °C and 1000 °C. After calcination, the respective lithium aluminosilicate powder was subjected into Fourier Transform Infrared Spectroscopy (FTIR) and Scanning Electron Microscope (SEM) analysis. Preparation of bi-component and tri-component was done to prepare lithium aluminosilicate powders. Based on FTIR studies, Si-O-Si bonds were appeared in the region of 1083.95 $cm^{-1}$ for sample calcined at 400 °C. For sample calcined at 600 °C , 800 °C and 1000 °C , the band shifted to the right which led to the formation of Si-O-Al bonds. By observing the four IR spectra, β-eucryptite was appeared in the region around 1005 $cm^{-1}$, 750 $cm^{-1}$ and 670 $cm^{-1}$. Based on SEM studies, the samples became denser and packed together as temperature gets higher due to the removal of water.

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# LIST OF SYMBOL AND ABBREVIATION

| | |
|---|---|
| °C | Degree Celcius |
| % | Percent |
| K | Kelvin |
| nm | Nanometer |
| atm | atmospheric pressure |
| g | Gram |
| M | Molar |
| ml | Milliliter |
| CRH | Constant rate heating |
| $NH_4OH$ | Ammonia Hydroxide |
| FTIR | Fourier Transform Infrared Spectroscopy |
| SEM | Scanning Electron Microscope |
| XRD | X-Ray Diffraction |

# CHAPTER 1

# INTRODUCTION

## 1.1 Glass Ceramic

Lithium aluminosilicate (LAS) is a type of glass ceramics. The subject of ceramic involves a wide range of materials. Glass ceramic materials are polycrystalline solids prepared by controlled crystallization of shaped glass objected (Arnault *et al.,* 2000). The invention of glass ceramics are the result of two independent line of research. The first glass ceramic produced industrially was introduced in the 1950s by Stookey (Hou *et al.,* 2006). Stookey who performed his research on controlled glass crystallization process had investigated the nucleation of glasses. The other investigation of glass ceramic had performed by Hummel in 1951. The discovery of Hummel showed the negative volume expansion of crystalline of β-eucryptite ($Li_2O$-$Al_2O_3$-$2SiO_2$). The glass ceramics approach has two major advantages over the ceramic approach: (a) very fined-grained microstructures can be produced; (b) high speed glass manufacturing processes can be used (Bach & Krause, 2005).

The dominant crystalline phase in LAS glass ceramics is either β-eucryptite ($Li_2O.Al_2O_3.2SiO_2$) or β-spodumene ($Li_2O.Al_2O_3.4SiO_2$) solid solution (Naskar & Chatterjee, 2005). According to Muller (1979), the structure of β-eucryptite with its site occupancy Li ions will displays order-disorder phenomena according to its Si/Al distribution. Besides, the physical properties of β-eucryptite are unsual. For β-spodumene, it is a rock forming mineral that bears other lithium minerals. β-spodumene has a very low thermal expansion coefficient , but its dielectric constant was found to be unacceptably high at approximately nine (Wessel, 2004).

Based on Bach and Krause (2005), glass ceramics based on two mutually development steps; first, it is includes the development of a base glass composition. Second, it is includes the ceramization process of the glass ceramic by which the base glass is transformed due to specified material properties. In principle, there are two options for the development of low-expansion glass ceramics. One option is based on a crystalline phase which itself has a coefficient of thermal expansion (CTE) closed to zero in the required temperature interval; in this case a corresponding composition has to be found and a ceramization process has to be developed so that the base glass transformed to almost 100% into designed crystalline phase. The second option is based on the crystalline phase which itself has a negative CTE in required temperature interval; in this case, the glass ceramic has to be designed in such a way that it consists partly of that crystalline phase with negative CTE and partly of glass phase (called residual glass phase) with positive CTE; the CTEs and the fractional volumes of both phases are adjusted to each other so that the CTE of the glass ceramic is close to zero.

In the development of low-expansion LAS (LAS=$Li_2O$-$Al_2O_3$-$SiO_2$) glass ceramics the second approach has always been favored. The requirement for malting a base of high quality as well as those for fine turning the CTE during ceramization can be met easily by this second approach (Bach & Krause, 2005).

Lately, sol-gel method is widely used in preparing the glass ceramics. In this case of study, fumed silica is used as the major silica in preparing the lithium aluminosilicate (LAS) powder by using sol- gel technology. The well known of sol gel process in making glass ceramics because of the use of low temperature without melting in the process. This technique is applicable to single-component oxide glasses as well as multi-component oxide glasses using metal alkoxides as precursors. The metal alkoxides from gels through hydrolysis accompanied by polycondensation reactions and then gels are converted to the oxide glasses by heat treatments (Lee *et al.,* 1994).

In this case of study, fumed silica ($SiO_2$) is used to be the major silica source to prepare the lithium aluminosilicate (LAS) powders. Generally, fumed silica also called pyrogenic silica, fumed silicon dioxide, and Aerosil fumed silica in Europe and Cab-O-Sil fumed silica in the United States. Fumed silica has been produced by combustion of silicon tetrachloride ($SiCl_4$) in hydrogen-oxygen flame. Fumed silica is an amorphous, synthetic, stable, high-purity chemical substance. Although purity of fumed silica is very high, greater than 99.8%, it nonetheless contains small amounts of trace metallic impurities, mainly Al, B, Fe, Mg, Ni, and Ti as well as a small amount of residual hydrochloric acid (less than 0.025%) from the manufacturing process (Bogdan & Kulmala, 2002).

Fumed silica is widely used in industrial scale. Due to its properties, fumed silica posses many unique functions, which include thickening and thixotropy, free flow and anticaking, suspension aid, reinforcement, gloss reduction, anti-slip, and anti-blocking.

Once the lithium aluminosilicate (LAS) powders are prepare, further characterization processes are need to carry out. During the experiment, Fourier Transformed Infrared (FTIR) spectroscopy and Scanning Electron Microscopy (SEM) are used to characterize the lithium aluminosilicate (LAS) powders.

## 1.2 Objective of study

The objectives s of this study are as follow:

i. To prepare the LAS powders by using sol-gel method with fumed silica as silica source.
ii. To characterize the LAS powders at different calcination temperatures by using Fourier transformed infrared (FTIR) spectroscopy and scanning electron microscopy (SEM).

## 1.3 Scope of study

This study focus on the preparation of lithium aluminosilicate (LAS) powders by using sol gel method. In this study, fumed silica ($SiO_2$) is chosen to be the silica source to produce the LAS powder. The LAS powders formed in this experiment are characterized by using Fourier Transformed Infrared (FTIR) spectroscopy and Scanning Electron Microscopy (SEM).

# CHAPTER 2

# LITERATURE REVIEW

## 2.1 Sol-gel

### 2.1.1 Definition of sol-gel

Sol-gel process has been widely known as an important new process for the manufacture of glasses and ceramics. Actually sol-gel process is built up by the term "sol" and "gel". The term "sol" refers to a colloidal suspension of solid in a liquid. While the term "gel" refer to a substance that contains a continuous solid skeleton enclosing a continuous liquid phase (Brinker & Scherer, 1990).

A colloid is a suspension in which the dispersed phase is so small (~1-1000nm) that gravitational forces are negligible and interactions are dominated by the short-range forces; such as Van der Waals attraction and surface charges. For a stable colloidal suspension, the particles are usually about 500nm or less in size. By modifying the condition of the solvent or suspension, the stability of colloidal suspension will be affected. Once the equilibrium of the colloid is disturbed, the sol will be transformed to either peptised particles, aggregated particles or gels.

### 2.1.2 Beginning of sol-gel science

The earliest use of colloids to prepare functional materials is seen in the cave paintings at Lascaux in France, dating back 17 000 years. The pigments used were ground into fine powder and graded by sedimentation and dispersed in water by using natural oils as surface active stabilizers. This decorative method is closely related to more recent ceramic decoration methods in construction and other practical applications field.

Firing technique with the addition to simple grinding to alter the chemistry of the mineral precursors is the second major development after the ceramic methods. During the firing technique, the sol-gel ideas are start to initiate. The general idea of sol-gel is about the fine powdered or colloidal material in suspension was moulded and then dried and densified by chemical action or by firing at high temperature (Wright & Sommerdijik, 2001).

Many early technologies have no longer exists in the Dark Ages after the decline of the Roman Empire. Discovery of "water glass" by Von Helmont in year 1644 became a major development after the lost of earlier technologies. Von Helmont had dissolved silicate materials such as stones, sand, flint and etc. in alkali and found that on the acidification a precipitate of silica equal in weight to the original silicate materials was obtained. In 1779, Bergman reported that if the correct amount of dilute acid was used the mixture gelled on acidification. This preparation of a silica gel led to a series of applications remarkable similar to those of today's sol-gel chemistry (Wright & Sommerdijik, 2001).

In year 1845, the formation of a transparent material as a result of slow hydrolysis of an ester of the silicic acid was reported by Ebelman. While during year 1930, sol-gel process for oxide layers on the industrial glasses with the use of metal-containing precursor by deep-coating had been proposed by Geffcen and Berger. After that D. Roy and R. Roy proposed a method for preparation of more homogenous melts and glasses using a sol-gel process (Dimitriev *et al.*, 2008).

According to the studies of Dislich, the synthesis of ceramic material and quartz glass from gel precursors have been proceed. Dislich had synthesized borosilicate glass by heating bathes of oxide powders at low temperature by using sol-gel process. In this case, Dislich compressed his first sol-gel Duran glass granulates at 550 °C and 2800 atm, yielding a solid borosilicate glass, in order to prove the identity with melted Duran. Figure 2.1 shows that the borosilicate glass from hydrolysed alkoxide (Dislich, 1985).

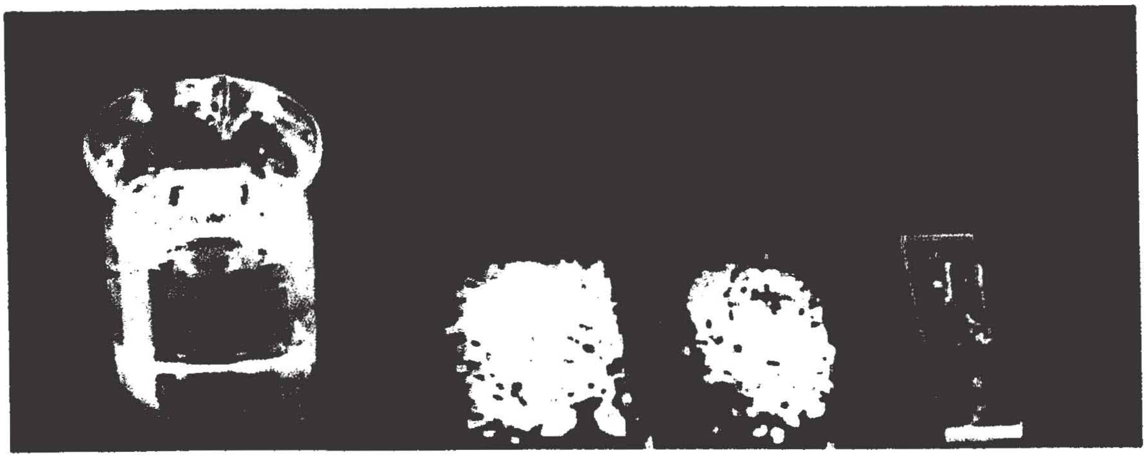

**Figure 2.1** Borosilicate glass from hydrolysed alkoxides.

According to Sakka, who is the pioneers in this new trend of technological development, the sol-gel technology is a typical nanotechnology because all gel products may contain nanoparticles or are nanocomposites (Dimitriev *et al.*, 2008).

Sol-gel processing has been used commercially for over fifty years by one glass company to produce thin film coatings with specialized optical or protective properties, but it has not widespread acceptance, either by industry or by university researchers. However, in last thirty years sol-gel process show a resurge of academic interest. This condition led to the consideration of sol-gel economic and technical viability from other companies.

The glass industry is interest in sol-gel technology for two reasons: to make existing products better and more cheaply; and to imagine new products accessible only through this technology. Hence, the direction of future sol-gel research and development will be determined by product price, quality, and novelty (Wenzel, 1985).

### 2.1.3 Basic principle of sol-gel technology

Sol-gel processing for the synthesis of glasses, glass ceramics and ceramics become more and more important regard to scientific and application purposes. There are two important sol-gel process; alkoxide method and colloidal method. Alkoxide method is based on the polymerization of alkoxysilane while colloidal method is based on the gelling colloidal sol. In the sol-gel process, the precursors adopted can be inorganic salts, colloidal solutions or metal-organic precursor in water or organic solvent. But generally, the most common route in sol-gel process is the use of alkoxide method.

The alkoxide method is based on the polymerization of alkoxysilane with metal alkoxides or metal salts to produce multicomponent noncrystalline inorganic gels with network structures (Mukherjee, 1980). During the alkoxide method, the precursor use is metal alkoxide. The use of metal alkoxide in the sol-gel synthesis will provide a high-purity glasses and crystalline ceramics.

The general formula for metal alkoxide is $M(OR)_n$. It can considered to be derivatives of either an alcohol, ROH where R is an alkyl chain, in which the hydroxyl proton is replaced by a metal M, or of a metal hydroxide, $M(OH)_n$. Alkoxy derivatives exhibit a large variety of physical properties, ranging from alkoxides of strongly electropositive metals through alkoxides of the heavier multivalent elements to the alkoxides of the lighter transition elements.

To have better understanding to the properties of alkoxides, few aspects must be take in account, such as (i) the percentage of ionic character of the M-O bond due to the electronegativity differences between oxygen and the M element; (ii) the electronic effect of the alkyl (or aryl) R group on the oxygen atom and (iii) the formation of oligomers due to the expansion of the metal coordination sphere by means of dative intermolecular bonds with donor atoms of neighbouring alkoxide groups (Guglielmi and Carturan, 1988).

According to Guglielmi and Carturan (1988), among the many physical properties, The most interesting properties which can be discuss is (i) the degree of oligomerization or polymerization; (ii) volatility and (iii) viscosity. The degree of oligomerization or polymerization can directly induce the differences in the sol-gel process and the resulting structures of multi-component systems. It may affect the homogeneity at the molecular level and indirectly induces the changes of other parameters.

For the second parameter, high volatility of some alkoxides precursor may allow the possibilities to easily achieve pure precursors by distillation. This parameter must be taken into account during sol-gel process in order to avoid unwanted changes of composition in multi-component solutions. Actually, the volatility of alkoxides may be related to the molecular size and shape of alkoxide group, thus it may affect the polarization of M-O-C bond, the degree of oligomerization and the intermolecular forces.

The third parameter of alkoxides is viscosity. Viscosity available is also affected by the chain length and branching of alkyl groups and by the degree of molecular association. In the sol-gel process, alkoxides are dissolved in solvents, thus the viscosity of the solution is depends on concentration of the solution.

In the sol-gel process, preparation of silica is important during the early research. In this process $Si(OH)_4$ molecules will be undergo condensation to form a siloxane network. In this case, the soluble silicate usually sodium silicate will be chosen to use as the starting material. The use of alkoxysilanes $Si(OR)_4$, with R equals to alkyl group such as $CH_3$, $C_2H_5$ or $C_3H_7$ become more and more significant in today research.

In addition, sol-gel processing is used not only for the preparation of silica gels, but also for the production of ceramic products, which ranging from thin films and coatings over porous membranes to composite bodies. This broad success of the method is due to its ability to form pure and homogenous products at very low temperature. Thus, the sol-gel technology is replacing the millennia old ceramics fabrication processes in which powders are shaped into objects and subsequently densified at temperatures close to their liquids. This allowed transformation of ceramics and glasses from ' stone age materials' to 'space age materials' (Vansant *et al.*, 1995).

### 2.1.4 Chemistry of sol-gel process

The most common precursors use to start the sol-gel processes are by using tetramethoxysilane (TMOS) and tetraethoxysilane (TEOS). Both of these two starting material are types of alkyoxysilane which using alkoxide route.

Actually sol-gel process is a type of wet chemical method which involves certain reaction such as hydrolysis, condensation, gelation, aging, drying, and stabilization or consolidation. Figure 2.2 shows that the simplified chart for the sol-gel routes (Pierre, 1998).

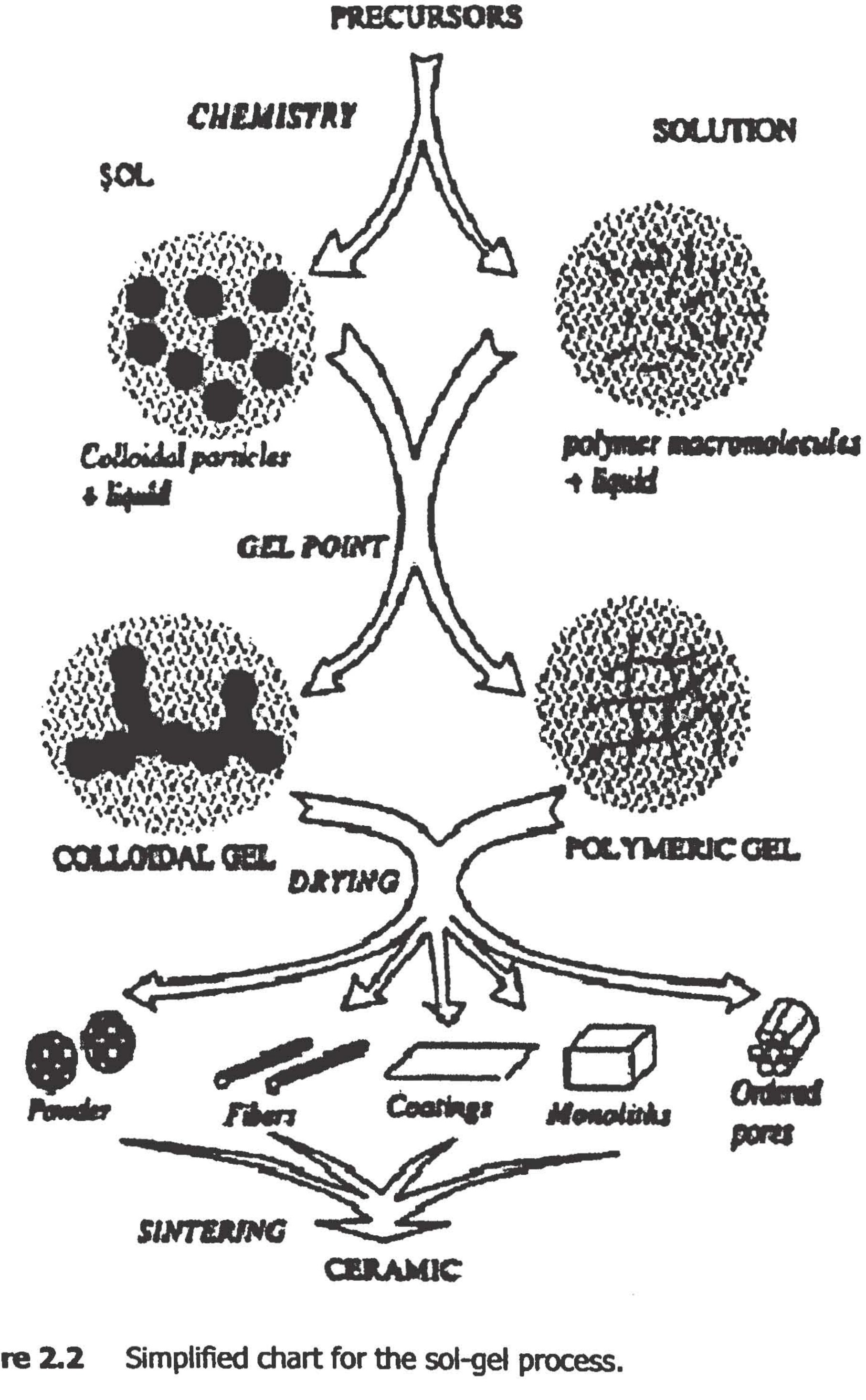

**Figure 2.2** Simplified chart for the sol-gel process.

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