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Polymer-Based Approach in Ceramic Materials Processing for Energy Device Applications

Mahendra Rao Somalu[1]*, Abdullah Abdul Samat[1], Andanastuti Muchtar[1,2] and Nafisah Osman[3]

[1]*Fuel Cell Institute, Universiti Kebangsaan Malaysia, Malaysia*

[2]*Department of Engineering and Built Environment, Centre for Materials Engineering and Smart Manufacturing (MERCU), Universiti Kebangsaan Malaysia, Malaysia*

[3]*Faculty of Applied Sciences, Universiti Teknologi MARA, Malaysia*

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***Corresponding author:** Mahendra Rao Somalu, Fuel Cell Institute, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia, Tel: +6 03 8911 8522; Fax: +6 03 8911 8530; Email: mahen@ukm.edu.my

Abstract

Polymer-based approach such as sol-gel method is a well-known method to produce ceramics materials with excellent properties for a better performance of solid oxide fuel cell. The properties of the materials are generally controlled by chemical agents used in this method. The roles of the chemical agents including chelating agent, polymerization or esterification agent and surfactant are presented and briefly discussed in this mini review paper.

Keywords: Sol-gel method; Polymerization agent; Surfactant; Electrolyte; Anode; Cathode; Solid oxide fuel cell; Microstructure

Abbreviations: SOFCs: Solid Oxide Fuel Cells; SSR: Solid-State Reaction; WCMs: Wet Chemical Methods; CA: Citric Acid; EDTA: Ethylene Diamine Tetra-Acetic Acid; EG: Ethylene Glycol; TETA: Triethylenetetramine; PEG: Poly-Ethylene Glycol; PVA: Polyvinyl Alcohol; PVP: Polyvinyl Pyrrolidone; YSZ: Yttria-Stabilized Zirconia; AC: Activated Carbon

Introduction

High temperature perovskite-type oxide conductive ceramics have attracted great attention worldwide due to the fact that these materials have a great potential to be used as electrolyte and cathode components in solid oxide fuel cells (SOFCs). SOFC is currently deemed as one of the most promising future power generation devices due to its high energy conversion efficiency, less/zero pollutant emission and able to operate on various fuels. Two major concerns that limited the performance of the current developed SOFC systems are low electrolyte conductivity and high electrode polarization resistance [1,2]. Controlling and modifying the microstructural properties of the ceramics components of SOFC is a promising way to tackle the concerns and could be achieved by selecting suitable ceramics processing routes as they greatly affect the microstructure properties of the produced ceramics materials [3]. Traditionally, a Simple Solid-State Reaction (SSR) method is used to prepare the perovskite-type oxide ceramics materials [4-7]. However, this method resulted in a poor microstructural property of the produced powders due to high temperature of treatment (> 1400 °C) and the produced powders are frequently contaminated [8,9]. Hence, Wet Chemical Methods (WCMs) are introduced to overcome the drawbacks of the SSR method. The WCMs are able to produce fine powders with high purity and good homogeneity at lower processing temperature than that of the SSR method [3,10]. One the most popular WCMs is a sol-gel method. The preparation of materials through this method is thoroughly discussed in the following section.

Sol-gel Method

A sol-gel method has been introduced in the 1800s to produce inorganic ceramics and glass materials [11]. It is a process to form oxide linkages via inorganic polymerization reaction. It starts with a reaction between molecular precursor and solvent to form metal organic complexes. The complexes will undergo polymerization process to yield colloid or sol followed by hydrolysis to form a gel. Then, the gel will turn into ceramics powder after drying and sintering processes [12]. A simple illustration of the sol-gel processes is shown in Figure 1.

There are two basic chemical reactions involve in sol-gel method which preserve the homogeneity of the metal salts (precursor materials) in the solution into gel. The first one is the complexation between metal ions and chelating agent such as Citric Acid (CA) and Ethylenediaminetetra-Acetic Acid (EDTA) which provides a stable metal-chelate in the solution by preserving atomic scale homogeneity. Another one is polymerization of the complexes with polymerizing agent or surfactant such as Ethylene Glycol (EG) which forms three-dimensional structures that hinder ion mobility and segregation. A simple illustration of the chemical reactions which involves in this method is shown in Figure 2 [13].

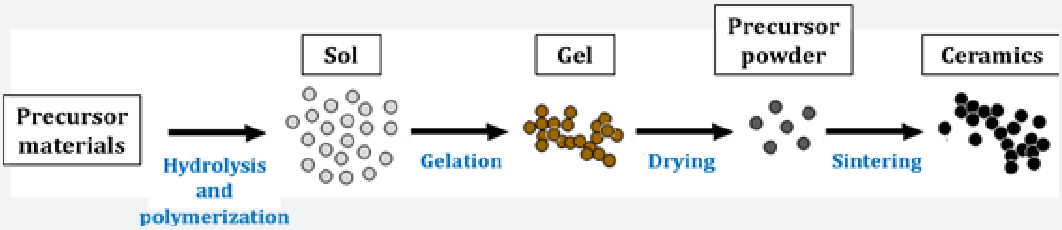

Figure 1: Process in producing ceramics powder via a sol-gel method. Adapted from [12].

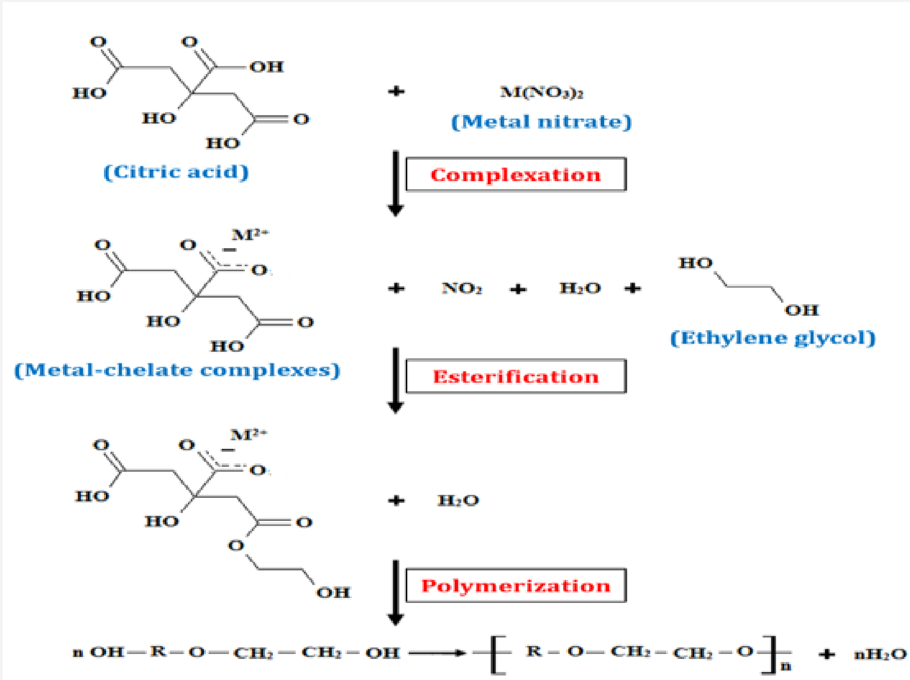

Figure 2: Chemical reactions in a sol-gel method. Adapted from [13].

Roles of Chemical Agents

Generally, a chelating agent acts to bind all the metal cations of the precursor materials to form a stable metal complexes in a precursor materials solution with homogenous distribution at atomic or molecular level. The addition of chelating agent is able to control the rate of hydrolysis reaction, phase transition, particle size and powder morphology [14]. CA and EDTA are the most conventional chelating agents used to produce various ceramics materials for SOFC application. They can be used separately as a single (CA/EDTA) chelating agent [15-17] or together as a combined (CA-EDTA) chelating agent [18-20]. A combined chelating agent is better than a single chelating agent because it is able to bind almost all metal cations and form more stable complexes than a single chelating agent. Moreover, it is less sensitive to pH value of the complexes solution and able to reduce the temperature required for single phase powder formation [21,22]. Besides, non-conventional chelating agents such as Triethylenetetramine (TETA), glycolic acid, nitriloacetic acid and tartaric acid have been also used to improve the properties of $BaCe_{0.54}Zr_{0.36}Y_{0.1}O_{2.95}$ electrolyte material for proton-conducting SOFC application [23].

Ethylene Glycol (EG) is a conventional solvent used in a sol-gel method. It acts as polymerization agent or esterification agent to produce cathode and electrolyte materials such $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ [20], $BaCe_{0.54}Zr_{0.36}Y_{0.1}O_{2.95}$ [24], Sm1-xCaxFeO3 [25] and SrCox [13] for SOFC applications. EG aids to form a stable polymer resin of metal-chelate complexes. The formation of polymer resin hinders the formation of particle agglomeration by forming rigid network which controls the movement of metal cations in the complex solution of precursor materials during heat treatment process [26-28]. For better properties of the produced powder, the amount of chelating agent and polymerization agent need to be optimized and controlled.

In a modified sol-gel method, a surface-active agent or simply known as surfactant is used to replace EG as a solvent. Surfactant is an amphiphilic compound consists of hydrophilic polar group and hydrophobic non-polar group. The polar and

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non-polar groups are the head and tail of a surfactant monomer, respectively. Surfactant is classified into three groups based on the charge of the polar group which are ionic (anionic or cationic), non-ionic and dipolar or zwitterionic. The non-polar group is made up of long-chain hydrocarbon or siloxane chain [29]. A simple diagram of a surfactant monomer is shown in Figure 3.

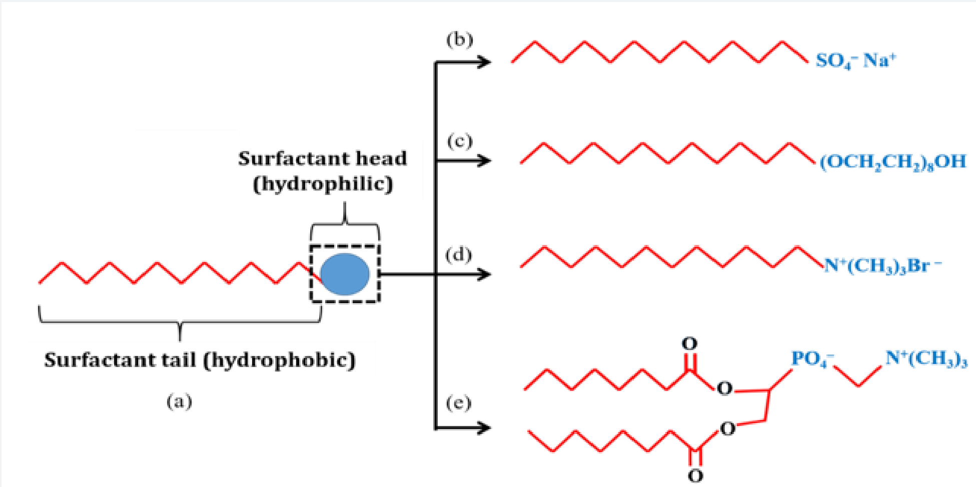

Figure 3: A simple diagram of (a) surfactant monomer, (b) anionic surfactant, (c) non-ionic surfactant, (d) cationic surfactant and (e) zwitterionic surfactant.

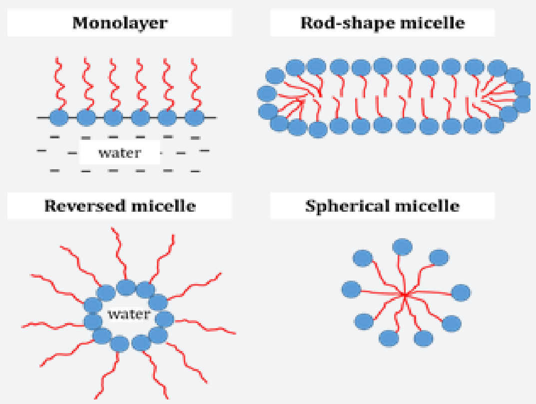

Figure 4: Various shapes of micelle in an aqueous solution. Adapted from [33].

A surfactant can increase miscibility, colloidal stabilization and particle dispersion in a material with various components because of its unique properties of self-assembly. These properties help to reduce the tension of two or more components in a solution system and change the properties of the surface of the solution and increase the compatibility between the particles with different properties in the solution [30,31]. In addition, a surfactant can control the shape and particle size of the produced ceramics materials. It forms a cluster of thermodynamically stable supramolecular known as micelle or microemulsion [32]. Micelle can form in various shapes in an aqueous solution as shown in Figure 4. It is dependent on temperature, surfactant concentration, surfactant composition and pH [29]. The variation in the micelle shapes is one of the factors that affects the particle shape and size of the produced powder [24].

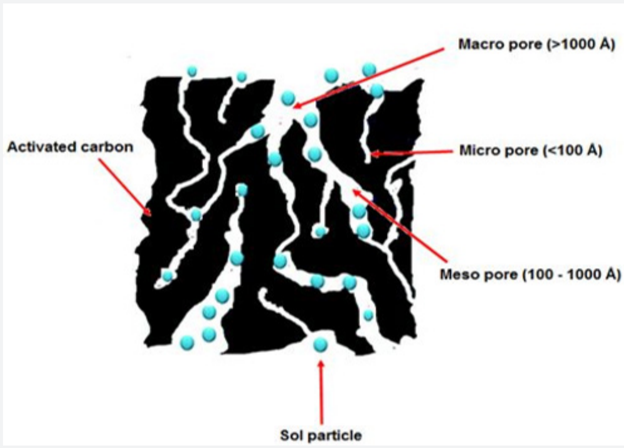

Figure 5: A schematic diagram of an activated carbon. Adapted from [44].

There are many surfactants which have been used to produce ceramic materials for SOFC components via a sol-gel method [33]. Pluronic F127 (tri-block copolymer) and Triton-X-100 were used to synthesis $La_{0.58}Sr_{0.4}Fe_{0.8}Co_{0.2}O_{3-\delta}$ [34,35], and Poly-Ethylene Glycol (PEG) was used to synthesize $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ cathode materials. Polyvinyl Alcohol (PVA) and Polyvinyl Pyrrolidone (PVP) were used to synthesize $La_{0.8}Sr_{0.2}MnO_{3-\delta}$ cathode material [36]. All of these surfactants helped to reduce the particle size from micro to nano, increase the surface area and homogeneity of the produced powders. The same results were also reported for the synthesis of electrolyte materials of Yttria-Stabilized Zirconia (YSZ) [37], $Ce_{0.9}Gd_{0.1}O_{1.95}$ [38], $Ce_{0.8}Sm_{0.2}O_{2-\delta}$ [39], and $BaCe_{0.54}Zr_{0.36}Y_{0.1}O_{2.95}$ [24], using various surfactants. Additionally, surfactant also helps to reduce the temperature required to produce single phase powder and high-density pellet [38,40].

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In addition, Activated Carbon (AC) has been introduced as a dispersing agent in a modified sol-gel method to produce perovskite-type oxide conductive ceramics for SOFC application. The use of AC as a dispersing agent is quite a new invention in sol-gel method. AC, which is a treated form of carbon with high degree of microporosity, surface area (300-2000m2g-1) and well adsorption ability, is used to replace the conventional solvent or surfactant in the conventional sol-gel method. Like the other chemical agents, AC is also responsible for controlling the nucleation process, phase development and particle growth during the synthesis process. However, it has different reaction mechanism as compared with the other chemical agents. AC traps the metal complexes in precursor solution in its highly porous microstructure through van der Waals attraction, creating well-dispersed particles of the produced ceramic materials after calcination process (Figure 5). Its potential in modifying microstructure of perovskite-type ceramic materials with good electrochemical performance particularly for SOFC application have been reported by Ismail et al. [41] and Abdul S et al. [42,43].

Table 1: Properties of ceramic materials produced by sol-gel and other methods for SOFC application.

Ceramic Material	Synthesis Method	Chemical agent	Particle size (nm)	SBET (m^2g^{-1})	Reference
$La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$	Sol-gel	CA-EDTA and EG	34	12	[41]
		CA-EDTA and AC	36	11	
	Microwave-assisted combustion	-	113	12	[45]
	Citrate-hydrothermal		975	5	[46]
	SSR			4-13	[47]
$La_{0.6}Sr_{0.4}CoO_{3-\delta}$	Sol-gel	CA		14	[48]
		CA and EG	50-200		[49]
		CA-EDTA and AC	219-221	10	[42]
		CA-EDTA and EG	130-260		[20]
		CA-ETDA and PEG	60	7	[50]
$BaCe_{0.54}Zr_{0.36}Y0.1O_{3-\delta}$	Sol-gel	CA, EG and Brij97	20-80		[24]
	Sol-gel	CA-EDTA and TETA	342-396		[51]
	Supercritical fluid		531 - 712		
	Sol-gel assisted supercritical fluid		255 - 295		
$Ce_{0.8}Sm_{0.2}Ba_{0.8}Y_{0.2}O_{3-\delta}$	Co-precipitation		60		[52]
	Sol-gel	CA	67		
	Ball milling		75		
$Gd_{0.1}Ce_{0.9}O_{1.95}$	Sol-gel	EG		16	[53]
		Tartaric acid		13	
		Glycerol		16	
	Co-precipitation			105	[54]
CeO_2 doped with Gd, Nd and Sm	Combustion		5-8	29-36	[55]
	Hydrothermal		4-8	43-57	
	SSR		90-400	2-3	
	Co-precipitation		3-5	88-123	

The improved properties of the produced ceramics materials by sol-gel method are very important for better electrochemical performance of the SOFC components [44-48]. The properties i.e. particle size and BET specific surface area (SBET) of some common ceramic materials for SOFC application produced by different chemical agents via sol-gel method and other synthesis methods are tabulated in Table 1. It can be seen that the properties of the produced ceramic materials are different. It is due to the different chemical agents used in the sol-gel method and the different in the method used to produce the ceramic materials [49-52]. The discrepancy in the mentioned properties might also be contributed by the different in the heat treatment process and synthesis parameters applied to obtain a single perovskite phase of the ceramic materials [53-55].

Conclusion

A sol-gel method is regarded as a promising synthesis route to produce better properties of ceramics materials for SOFC electrolyte and cathode components. The chemical agents used in this method have been proved to significantly affect the properties of the produced materials such as particle size and SBET. Given that research on the sol-gel method has been

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continuously improving with time, many other materials can be used as chemical agents. Additional studies of basic understanding on how the chemical agents work and what are the condition they work at the best, including composition, pH, concentration and processing temperatures much be investigated in detail.

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