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ZIRCONIA MIXED METAL OXIDE NANOPARTICLES FOR THERMAL BARRIER COATINGS: SYNTHESIS AND CHARACTERIZATION: A REVIEW

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ABSTRACT

For the synthesis of zirconia mixed metal oxide nanoparticles, various synthetic routes have been used. The various synthetic methods, as well as other parameters such as concentration, pH, and the type of precursor used, aid in the creation of zirconia mixed metal oxide nanoparticles with various physicochemical properties. This paper discusses the various synthetic routes of sol-gel, hydrothermal, and co precipitation for the formation of zirconia mixed metal oxide nanoparticles from zirconia and other metal oxides, as well as the physicochemical properties and characterization techniques of the synthesised zirconia mixed metal oxide nanoparticles.

KEYWORDS: *Synthesis of Zirconia, Metal Oxides, Metal Oxide Nanoparticles*

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INTRODUCTION

Mixed metal oxide nanoparticles, also known as heterometal mixed oxide nanoparticles, are created by combining two or more metal oxides to improve the properties of synthesized products. Numerous metals with various oxidation states can be combined in a variety of ratios to form mixed metal oxide nanoparticles. The physical, chemical, and morphological properties of the synthesized mixed metal oxide nanoparticles vary, and they are used in a variety of fields **[1]**. Since the invention of nanomaterials, it has been well established that the nanoparticles' small size is responsible for a variety of their unique properties **[2]**. This has ushered in a plethora of research studies spanning various fields such as chemistry, physics, and medicine. Nanoparticles' high surface energy, large area-to-volume ratio, and small size in comparison to bulk materials **[3]** have enabled them to denote unique catalytic **[4]**, thermal **[5]**, optical **[6]**, electrical **[7]**, and biological application **[8]** properties that are being used in a variety of fields.

Due to its high strength, fracture toughness, and hardness, zirconia nanoparticles are used in a variety of syntheses. Depending on the temperature, pure zirconia can take three different shapes: monoclinic, tetragonal, and cubic. At room temperature, the monoclinic form is thermodynamically stable, but at temperatures above 1170oC, it transforms into the tetragonal form, and at 2370oC, it transforms into the cubic form. The hydrothermal process **[9]**, biological synthesis **[10]**, co-precipitation **[11]**, solid state reaction **[12]**, microwave synthesis **[13]**, and sol-gel method **[14]** have all been used to synthesize zirconia nanoparticles. However, due to agglomeration, the majority of those nanoparticles are poorly crystalline or have a broad particle size distribution **[15, 16]**. Zirconia has been combined with other diverse metal oxides to create zirconia mixed metal oxide nanoparticles to address the issue of agglomeration that occurs during the synthesis of single metal oxide nanoparticles and to stabilize the higher temperature polymorphs (tetragonal and cubic) to enhance their application in various fields **[17, 18]**. Sol-gel, hydrothermal, co-precipitation, chemical polymerization, stir casting technique, thermal casting, and biological synthesis are among the various synthetic routes for the preparation of zirconia mixed metal oxide nanoparticles discussed in this review. In addition, the various characterization techniques used will be discussed. Finally, during this research path, some future considerations and advances are highlighted.

Synthesis of Zirconia Mixed Metal Oxide Nanoparticles

Metal oxide and mixed metal oxide nanoparticles can be made using a variety of different synthetic routes. Each of the synthetic routes has been categorised according to the technical approach used. The various technical approaches could also be in accordance with the type of growth media: vapour phase growth, liquid phase growth, solid phase formation, and hybrid growth, as well as the state of the products: nanoparticles via colloidal processing, nanorods and nanowires via template-based electroplating, thin films via molecular beam epitaxy, and nanostructured bulk materials. The top-down approach entails physically reducing a bulk material to nanoscale dimensions, whereas the bottom-up approach entails constructing a structure from the unit building blocks **[19]**.

Thermodynamic Method

The hydrothermal method is an example of a solvothermal synthetic route that is used to make a variety of nanomaterials by dispersing the starting material in a suitable solvent and subjecting it to moderately elevated temperature and pressure conditions, resulting in nanoparticle formation. The method is known as hydrothermal synthesis when water is used as the reaction's solvent. Chemical parameters such as reactant composition and concentration, solvent or reducing agent ratio, and thermodynamic parameters such as temperature, pressure, and time all play a role in the formation of nanoparticles. Machmudah et al used hydrothermal synthesis to make ceria-zirconia mixed oxide and found that the size of the particles formed is temperature dependent **[20]**. J. R. Kim et al. used supercritical water as a catalyst to investigate Ceria-Zirconia mixed oxide prepared by continuous hydrothermal synthesis. As a result of its sparsely agglomerated morphology, supercritical synthesis has been reported to result in Ceria-Zirconia mixed oxides with higher thermal stability and better oxygen storage capacity, with potential application as a catalyst **[21]**. According to Piticescuet. al, the solubilization-reprecipitation process results in the formation of a stable cubic phase, with the crystallinity of the synthesised nanoparticles increasing with time and temperature of the hydrothermal treatment **[22]**. X. Wang et al investigated the photocatalytic activity of ZrO2-CeO2 synthesised by calcinations of the precursor prepared by a one-step hydrothermal method in the light region. According to the findings, when compared to the monocomponents of ZrO2 and CeO2, ZrO2/CeO2 nanocomposite showed enhanced photocatalytic activity **[23]**. In addition to the above, other synthetic routes have been used to synthesise zirconia in combination with other compounds or elements for a variety of applications.

Patoliya et al. investigated the preparation and characterization of Zirconia reinforced aluminium metal matrix composites using the stir casting technique and found that increasing the weight fraction of Zirconia particles within the aluminium matrix improved mechanical properties such as hardness and impact strength **[24]**. A. Sultan et al. used a chemical polymerization method to study the synthesis, characterization, and electrical properties of poly pyrrole/zirconia nanocomposites and their application as an ethane gas sensor **[25]**. H. Tu et al. investigated the synthesis and characterization of Scandia-Ceria stabilised Zirconia powders prepared using a polymeric precursor method for integration into anode-supported solid oxide fuel cells **[26]**, while Mudila et al. investigated the electrochemical performance of zirconia/graphene oxide nanocomposites cathode designed for enhanced power density super capacitor and found that the nanocomposite David et al. investigated the synthesis and characterization of Co3O4-ZnO-ZrO2 ternary nanoparticles and found that as the concentration of the precursors increases, so does the size of the nanoparticles due to agglomeration of the tiny metal oxide nanoparticle **[28]**. Tsai et al investigated the reactive oxygen species scavenging properties of ZrO2-CeO2 solid solution nanoparticles and found that the ceria-zirconia nanoparticles are highly crystalline in nature and will be dispersed in sodium citrate buffer at pH 7.4 where the reactive oxygen species scavenging activity of CeO2 nanoparticles was increased fourfold by incorporating zirconium into its crystal structure. The amount of oxygen vacancies within the lattice correlates with the scavenging activity of these nanoparticles **[29]**.

Co-Precipitation

Using a precipitating medium, the oxo-hydroxide form of a salt precursor (for example, chlorides and nitrates metal salts) is precipitated from a liquid of the salt precursor in a solvent (e.g., H2O or NaOH). When the critical concentration of the specie within the solution is reached, the nucleation process is followed by the growth phase. Arsent'ev et al. synthesize ZrO_2-CeO_2 nanoparticles using coprecipitation method **[30]** and Rossignol et al prepared ZrO_2-CeO_2 materials by using sol-gel and coprecipitation methods. It has been reported that both the structure and therefore the texture of the solids obtained depend upon the syntheses route and also the precursor used **[31]**. Kumar et al investigated the preparation, characterization and antibacterial application of MgO-ZrO_2 mixed oxide nanoparticles. After the experiment, the antibacterial study was reported to demonstrate that the mixed nanoparticles are going to be employed to treat infectious diseases caused by E.coli **[32]** while M Liu et al. administered the investigation of (CeO_2) x (Sc_2O_3) (0.11-x) $(ZrO_2)_{0.89}$ (x=0.01-0.1) electrolyte materials for intermediate temperature solid oxide cell **[33]** and Maridurai et al. reported the synthesis and characterization of yttrium stabilized zirconia nanoparticles **[34]**. Glushkova et al investigated the nanostructure evolution in partially stabilized Zirconia- based solid solutions prepared by coprecipitation and to assess the efficiency of freeze drying in preventing agglomeration of the powders **[35]**. Thirupathy et al. reported that Yttria stabilised zirconia nanoparticles were successfully synthesised using the co-precipitation method. The XRD pattern was used to confirm the structure. SEM and TEM analyses were used to determine the sizes of the synthesised samples, and the sizes calculated were in nanometers. The PL spectrum was used to investigate the optical properties. The dielectric constant, dielectric loss, and AC conductivity of YSZ nanoparticles were investigated as a function of frequency and temperature. Dielectric studies revealed that as frequency increased, both the dielectric constant and the dielectric loss decreased **[34]**.

Sol-Gel Technique

The sol-gel method entails hydrolysis of a metal-organic compound precursor to produce oxo-hydroxide, followed by condensation and polymerization to form a network of metal hydroxide and a porous gel, respectively, with drying and heating of the gel resulting in nanoparticle formation **[35]**. Various zirconia mixed metal oxide nanoparticles have been synthesised using this method. Morteza Hajizadeh-Oghaz, et al. investigate whether the calcining temperature should be above 600 °C in order to obtain pure CYSZ powders that will not decompose at high temperatures. Above 650 °C, the thermal decomposition of the gel into oxide was almost complete **[36]**. Using the sol-gel method, alumina-zirconia powders were synthesised and characterized. The effect of the solvent and the rate of water addition. Following the experiment, different solvents result in different textural properties, while different water addition rates result in different structural properties of the synthesised material **[37]**. Tayseir, M, et al. investigated the sol-gel synthesis and characterization of zirconia containing hydrophobic silica nanoparticles in 2017. The structural and textural properties of pure silica and silica zirconia were clearly different depending on the zirconium percentage, as revealed by characterization techniques. The characteristic result revealed that the zirconia content had an impact on the structure and morphology of synthesised silica **[38]**. Bashir et al. used a sol-gel method to make ZrO2-ZnO nanoparticles and found that smaller ZrO2-ZnO nanoparticles could only be made after doping zirconia with zinc oxide **[18]**. Rossignol et al. used sol-gel and co precipitation methods to make ZrO2-CeO2 materials. The structure and texture of the solids obtained are said to be dependent on the synthetic route and the precursor used **[31]**. The catalytic activity of CeO2-ZrO2 mixed oxide catalysts prepared using the sol-gel technique and tested for CO oxidation was investigated. Under the conditions of the study, highly uniform nanosize solution particles of ceria-zirconia have been reported. The addition of zirconium to the catalysts helped to stabilise them. The CO oxidation activity of mixed oxides was discovered to be dependent on the Ce/Zr ratio, which is related to reducibility. With a lower Ce/Zr ratio, the catalytic activity of CO oxidation decreases **[39]**. Abd El Hakam et al. investigated the structural, photo-catalytic, and antibacterial activity of ZnO and ZrO2 doped ZnO nanoparticles and found that undoped ZnO is more photo-catalytically active than ZrO2 doped ZnO, with ZrO2 doped ZnO having better antimicrobial activity **[40]**. ZrO2-CrO2 **[41]**, ZrO2-TiO2**[42]**, and ZrO2-GO **[43]** are examples of other sol-gel synthesis.

Zro 2 Mixed Metal Oxide Nanoparticles Characterization

Transmission electron microscopy (TEM), X-ray diffraction (XRD), and scanning electron microscopy were used to characterize the dimension, crystal structure, and morphology of the synthesised zirconia mixed metal oxide nanoparticles (SEM). Different researchers have used different synthetic methods to synthesize and characterize ZrO2- CeO2 nanoparticles. From the results of the SEM analysis, Hou et al. synthesize zirconia-ceria by chemical precipitation to obtain irregular spheroids uniformly coated by cubic fluorite of CeO2 nanoparticles. According to the TEM analysis, the dimensions of the mixed composites increased by about 5nm compared to pure ZrO2 nanoparticles. As a result, the CeO2 shell was formed on the ZrO2 surface, with the PH value having a significant impact on coating tetragonal phase ZrO2 nanoparticles **[44]**. Rossignol et al. synthesised ZrO2-CeO2 using the sol-gel and co-precipitation methods, obtaining cubic and orthorhombic structures, respectively, for the sol-gel and co-precipitation methods. Various ZrO2-CeO2 systems with various structures can be made using various precursors. The nanoparticles' structure and texture are determined by the synthetic method and, as a result, the precursor used **[32]**. Table 1 shows some examples of various synthetic processes and their characterization techniques:

Table 1: Some Examples of Various Synthetic Processes and Their Characterization

Method	Morphology (SEM)	Size (TEM)	Crystal Structure (XRD)	Reference
Hydrothermal	Smooth surface	35,61, 31 nm	Sphere shape	Machmudah, *et al.*, 2015 [20]
Hydrothermal	Rough surface	300 nm	Tetragonal, Monoclinic	X. Wang, *et al.*,2013 [23]
Reversemicelle method	Not Investigated	3.7 nm	Cubic fluorite structure	Tsai, *et al.*, 2008 [29]
Chemical precipitation	Rough appearance	Increased by 5nm of the size of ZrO2	Irregular spheroids	Hou, *et al.*, 2015 [44]
Coprecipitation	Rough appearance	5-8 nm	Monoclinic, Tetragonal	Glushkova, et al., 2006 [45].

Machmudah et al. used a hydrothermal method to make CeO2-ZrO2 nanoparticles. The diameter of the nanoparticles at different temperatures was determined using a particle size analyzer that was shaped like a sphere with a smooth surface morphology. Increasing the temperature in a hydrothermal process has a positive effect on structural characteristics and can improve particle nucleation and linear growth rates **[20]**. Arsent'ev et al. reported diameters of 10-12 nm for ZrO2-CeO2 synthesized by chemical precipitation, with the phase dependent on the percentage composition of each precursor present in the reaction **[30]**.

CONCLUSIONS

The literatures cited in this review demonstrate how zirconia mixed metal oxide nanoparticles can be made using various synthetic methods. A variety of parameters, such as the synthetic method, precursor, temperature, and the type of solvent used, have regulated and controlled the sizes, shapes, and thus the desired applications of the synthesised nanomaterial. To analyse the formed nanomaterial, various characterization techniques were used, with TEM being used in the majority of the literatures to define particle size. Although zirconia has been synthesised with other metal oxides for various applications **[27, 30, 32, 40]**, there have been few reports on the thermal barrier coating of zirconia mixed metal oxide nanoparticles. As zirconia nanoparticles have been synthesised with other metal oxides, more work needs to be done to investigate the combination of zirconia with other metal oxide to obtain improved thermal barrier coatings.

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CONFLICTS OF INTEREST

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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