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A Review of Advanced Mullite Ceramics

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Abstract

Mullite is becoming one of the most extensive oxide ceramic materials for advanced structural and functional ceramics because of its remarkable properties. Such properties are low density, low thermal expansion, excellent creep resistance, low thermal conductivity, excellent strength at high temperatures, and good chemical stability. Nowadays, mullite has vast areas of application in various fields such as structural, electronic, optical, and high-temperature, *etc*. Mullite exists in an orthorhombic crystal structure with the stoichiometric composition of $3Al_2O_3{\cdot}2SiO_2$. This study provides an overview of the mullite's structure, properties, synthesis routes, various modern applications. Following a short introduction, this review paper focuses on the basic crystal structure of mullite. Secondly, this paper deals with various properties and application areas of mullite ceramic, Thirdly, the authors have listed differently stating raw materials and various synthesis routes to fabricate mullite ceramic in table format and try to compile the research outcome by other researchers. Finally, the last part of the study is the various applications of mullite ceramics, mullite synthesis challenges, and waste material utilization.

Keywords: Chemical vapor deposition; Mullite; Mullitisation temperature; Sol-gel; Spray pyrolysis.
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1. Introduction

The mineral mullite is an alumina (Al_2O_3) and silica (SiO_2) compound extensively used in traditional and as well as technical applications.[1] Mullite shows excellent properties such as low density, superior strength, low conductivity, low thermal expansion, high thermal stability, excellent thermal shock behavior, and good fracture toughness. Mullite also shows excellent chemical resistance and low gas permeability in severe atmospheres. The mineral mullite has $3Al_2O_3{\cdot}2SiO_2$ as its stoichiometric composition, found very rare in natural rocks. Natural mullite was found deposited on the Isle of Mull in U. K. (United Kingdom), and it was named "mullite".[2] The formation of this natural mullite on the Isle of Mull is due to volcanic activity through contact with super-heated lava and produces a high-temperature mullite phase. Natural mullite can also form by the lightning actions into the alumina-rich quartz sands.[3] The phase diagram of the alumina and silica system is present in Fig. 1.

The alumina-silica phase diagram indicating mullite and other phases is shown in Fig. 1. Mullites phase boundary joint in 1800 °C with 77.2 wt.% of Al_2O_3 composition corresponds to $2Al_2O_3{\cdot}SiO_2$ and this mullite composition are called 2/1 mullite. Suppose the mullite composition decreases from 1800 °C to below 1600 °C, the mean Al_2O_3 content shifts towards lower, near about < 73 wt.%. It corresponds to a composition of about $3Al_2O_3{\cdot}2SiO_2$, and this composition of mullite is called 3/2 mullite or stoichiometric mullite.[4]

Mullite is becoming an essential material because of its suitability for optical, electronic, and high-temperature structural applications. Classical mullite ceramics are broadly used for porcelain, pottery, whitewares, cement manufacturing, glass production, refractories, kiln slabs, lining for high-temperature reactors, *etc*.[5] Advanced or engineering mullite ceramics have gradually increased an essential role during the last two decades. The advanced mullite is used as a high-temperature composite matrix material development, protective coating, *etc*. The latest synthesis processing methods have often produced advanced mullite ceramics for the purity and homogeneity of mullite microstructure. Modern mullite-based ceramics applications such as; Mullite fibers: mullite fiber, shows an excellent high-temperature insulation characteristic at very high temperatures, such as kiln seals and electrical insulation. Diesel emission filters are also using mullite fiber fabrics.[6-9] Mullite whiskers: mullite fiber bundles, short and continuous fibers are using as reinforcement in ceramics or metals.[10] Monolithic mullite

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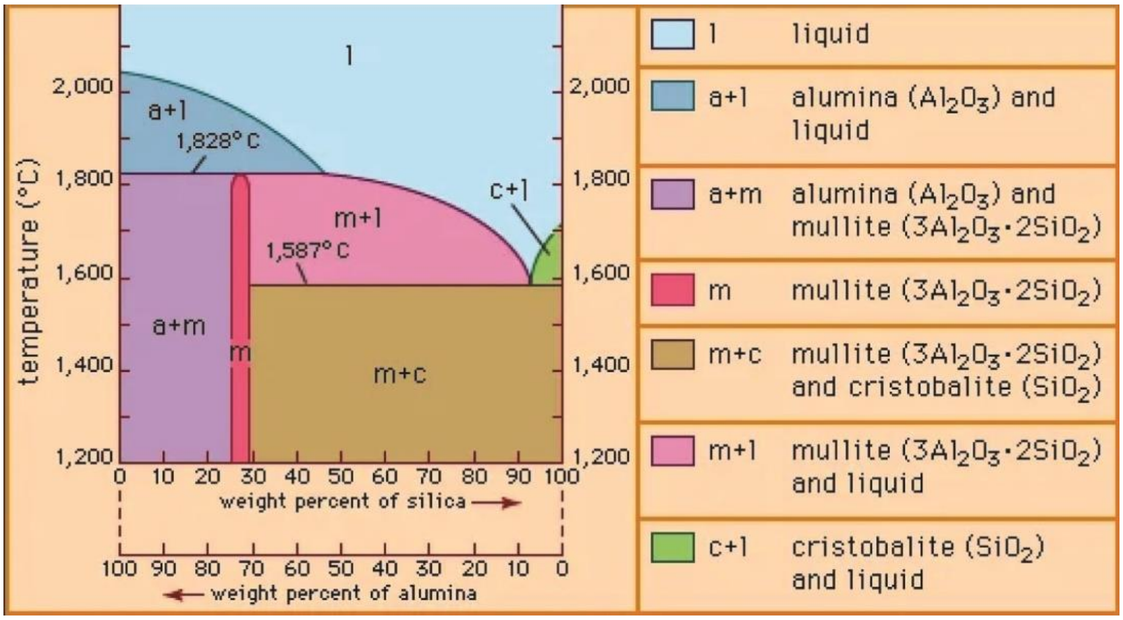

Fig. 1 Mullite part in the Al_2O_3-SiO_2 phase diagram. Reproduced with the permission form [4], Copyright 2013 International Scholarly and Scientific Research & Innovation.

ceramics: the properties of monolithic mullite are depended on the shape, size, amount, and porosity. Monolithic mullite ceramics are used in refractories, crucibles, thermocouple tubes, heat exchangers, substrates for silicon solar cell devices, dental ceramic components, hot gas filters, electronic packaging, *etc*.[3,11-16] Mullite coatings: mullite coatings are used to avoid oxidation of materials, and a thin coating of mullite is deposited using the chemical vapor deposition (CVD) method. Mullite coatings are also used on substrates for oily water cleaning and hot gas.[17-19] Mullite coatings show good corrosion resistance and excellent creep resistance at high temperatures.[20] Mullite composites: mullite has excellent oxidation resistance, and for this property, mullite is used as a matrix composite and fibers. Due to their superior thermal and mechanical properties, various non-oxide composites are now replacing mullite composites.[21,22]

The application of mullite in various fields increases interest in the researchers for an extensive study of mullite and mullite-based composite by utilizing multiple synthesis methods. Mullites are synthesis artificially because it is rarely found in nature. Different processing methods are available to synthesize mullites, such as sinter mullite, fused mullite, and reaction mullite. Many researchers have studied mullite chemistry and its properties.[23] The present review paper focuses on the mullite structure, its properties, different synthesis methods, various researchers' work, classical and modern applications, synthesis challenges, and waste materials utilization.

2. Structure of mullite

2.1 Crystal of mullite

The structures of mullite were characterized and classified by Fischer and Schneider.[24,25] It was reported that the mullite crystal structure's fundamental building unit is the chain of edge-sharing octahedra in a tetragonal arrangement in its highest topological symmetry. The crystal structure of mullite is typically an orthorhombic aluminosilicate system, with the general composition of $Al_2(Al_{2+2x}Si_{2-2x})O_{10-x}$. The basic building block of the mullite structure consists of chains of AlO_6 octahedra at the edges and center of the unit cell, which is run parallel to the crystallographic c-axis. These octahedra chains are joined together by (Al, Si) O_4 tetrahedra double chains, which run parallel to the c-axis.[24] Projection [001] of a unit cell of mullite crystal is shown in Fig. 2.

Mullite holds a defective crystal structure consisting of Al-O octahedral chains and runs parallel to the c-axis of the orthorhombic unit cell. These warp chains in the crystal structure are connected by Al-O and Si-O tetrahedra's double chains with Al and Si atoms. This defective structure formed new stoichiometry, and the composition of mullite may change from $3Al_2O_3{\cdot}2SiO_2$ to $3Al_2O_3{\cdot}SiO_2$. The compositional variation in mullite is achieved by the substitution of a Si^{4+} and removal of an oxygen ion from an (Al, Si) O_4 tetrahedron, leaving a vacancy of oxygen. The changes in the position of the cations in partially occupied are shown in Fig. 2.[22,23,25,26]

2.2 Chemistry of mullite

Mullite is known for its stoichiometry $3Al_2O_3{\cdot}2SiO_2$. Nowadays, commercially manufactured mullite called 3/2 mullite generally consists of a solid solution of 71 to 76 wt.% Al_2O_3 and 23 to 24 wt.% SiO_2. A few sintering aids, such as TiO_2, Fe_2O_3, CaO, and MgO, are added to control the sintering temperature. As stoichiometric composition moves away, the glassy grain boundary forms and reduces the mechanical properties.[26] 3/2 mullite (stoichiometric mullite) is produced without a glassy (quartz) grain boundary phase, which may give more extraordinary mechanical properties at very high temperatures.[27]

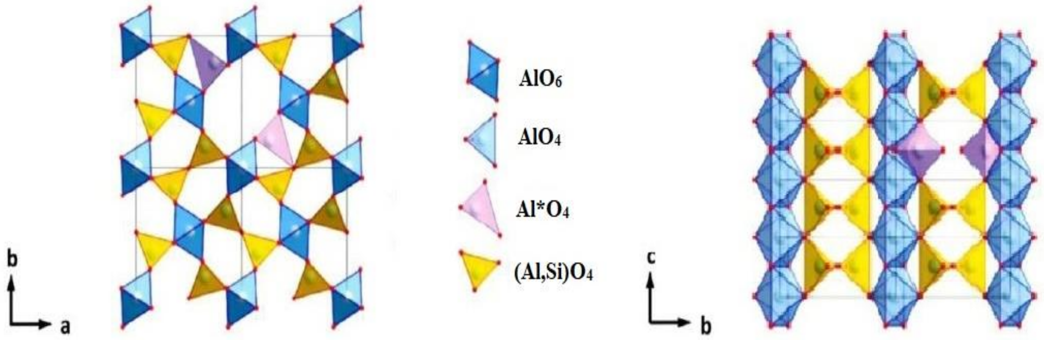

Fig. 2 Ideal orthorhombic unit cell of mullite [001] plane. Reproduced with permission from [27], Copyright 2020 Coatings.

2.3 Microstructure of mullite

The processing methods strongly influence mullite development mechanisms, and a substantial difference has been identified while mixing on the atomic scale or molecular range. The microstructure and properties of mullite are depended on its processing methods. Mullite ceramics should have tiny crystals and a small number of glassy phases.[28] Mullite has to be free from the glass phase in high-temperature structural applications, and grater crystal size is favored. The liquid silica phase present in the microstructure has prismatic mullite grains.[29,30]

3. Basic properties of mullite ceramics

3.1 Strength

Mullite melting point is too high; hence, it uses high-temperature exposed parts and has adequate strength and creep resistance at elevated temperatures. In mullite, creep resistance, bending strength and are affected by the presence of quartz or glass in grain boundaries, but in the absence of glass in the grain boundary, mullite ceramics shows excellent strength at room temperatures and up to 1400 °C. It also exhibits low heat conductivity and excellent thermal shock resistance, respectively. High-temperature mullite can also be formed by reducing the glass phase from its grain boundary. It is difficult to achieve because of mullite's low bulk and surface diffusivity property. Therefore, the mullite processing route plays a vital role in improving the mechanical property by removing the glassy phase. There are some processing methods by which we can minimize glassy phase regions from grain boundaries and the processes discussed in the synthesis section.[31]

3.2 Microhardness

Various authors have studied the microhardness of mullite up to 1400 °C.[31-33] Kriven *et al.* measured microhardness under different temperatures range, which is shown as follows;[33] From room temperature to ≈300 °C, it is shown that there is a minor reduction in the microhardness from ≈16 to ≈13 GPa, which corresponds to ≈0.01 GPa/°C. From 300 °C to 1000 °C, a slight decrement in the value of microhardness from ≈13 to ≈10 GPa, corresponds to ≈0.004 GPa/°C. From 1000 °C to 1400 °C with a relatively substantial decrement in the value of microhardness on an average value of 6 GPa at 1400 °C which corresponds to ≈0.01 GPa/°C.

3.3 Thermal expansion

Thermal expansion behavior is one of the significant properties of mullite ceramic, and severe work has been done to quantify the thermal expansion behavior. Kriven *et al.* studied the thermal expansion of mullite up to the temperature of 1400 °C.[34] Table 1 gives the mean thermal expansion value from 20 °C to 1400 °C.

3.4 Heat capacity (C_p)

It is an extensive property of a material, which may define the amount of heat required to raise the temperature of a mass of a substance by a unit degree Celsius. Pankrats *et al.*, Gierszewski *et al.*, and Hildmann *et al.*[35-37] determine the C_p value of mullite in the range from -125 °C to 1400 °C. The result exhibits a parabolic curve with values of about 0.415 J g^{-1} K^{-1} at -125 °C, 0.78 J g^{-1} K^{-1} under room temperature, and 1.25 J g^{-1} K^{-1} at 1000 °C. Temperature above 1100 °C a weak through reversible step-like C_p irregularity is seen.

3.5 Thermal conductivity

It measures its ability to transfer the rate of heat through a unit cross-section area. This property plays a crucial factor for films or coatings as a heat resistance system for mullite's case. Hildmann *et al.*[37] analyzed the thermal conductivity of mullite, where the temperature range was up to 1400 °C. With the increase in temperature, the thermal conductivity decreases potentially. The thermal conductivity of the mullite ceramics temperature range from 100 °C to 1400 °C shown in Table 1.

3.6 Dielectric strength

Mullite is used as an electrical insulator at ambient temperature; it is suitable for making electronic devices. Roy *et al.* investigated the conductivity of mullites doped with Co^{2+}, Ni^{2+}, and Cu^{2+};[38] it is observed that the resistivity of mullite is increased with an increase in doping quantity. Mullite ceramics doped with Ni^{2+} shows the lowest dielectric constant. Table 1 shows some properties of mullite.

Table 1. Different properties of mullite ceramics.[29,39]

Properties	Units of Measure	SI/Metric
Mechanical		
Density	gm/cc	≈3.2
Porosity	%	0 to > 4%
Color	-	off-white
Flexural Strength	MPa	≈190
Tensile Strength	MPa	≈132
Elastic Modulus	GPa	91-220
Compressive Strength	MPa	550-1320
Hardness (Knoop)	-	≈1450
Fracture Toughness KIC	MPa $m^{1/2}$	≈2.5
Maximum Use Temperature	°C	≈1650
Thermal		
Thermal Conductivity		
100 °C	$(W\ m^{-1}\ K^{-1})$	≈6.07
600 °C		≈4.31
1000 °C		≈3.98
1400 °C		≈3.89
Linear thermal expansion 20-1400 °C	$(\times 10^{-6}\ °C^{-1})$	≈4.5
Specific Heat	J/Kg °K	≈250
Electrical		
Dielectric Strength	ac-kV/mm	≈9.8
Dielectric Constant	@ 1 MHz	≈6.5
Dissipation Factor	@ 1 kHz	≈0.003
Volume Resistivity	Ohm-cm	>1013

4. Mullite synthesis

Mullite ceramic's purity, homogeneity, multistation temperature, densification, and other mechanical, electronic, and thermal properties, *etc*., mainly depend on the synthesis method. The temperature plays an essential role in the mullite fabrication process, leading to mullite formation. The following ways we can synthesize mullites:

1. Sintered Mullite
2. Fused Mullite
3. Reaction Mullite

4.1. Sintered mullite

In the conventional processing method, sintered mullite is usually prepared by mixing clay minerals, alumina (Al_2O_3), Silica (SiO_2), china, *etc*. The name sintered mullite describes the mullite, which has been synthesis by the solid-state diffusion-controlled reaction, where the raw materials are diffused by high temperatures up to 1600 °C. The sintering temperature is influenced by the alumina content described in the introduction (3/2 and 2/1 mullite). If the Al_2O_3 content reduces from 77 wt.% to near about 73 wt.%, the sintering temperature also reduces from 1800 °C to ≈1600 °C.[40,41]

Mullitisation occurs during the starting materials' solid or liquid phase reactions (*e.g.*, clay minerals, alumina, and silica). Mullitisation temperature can be reduced by using very fine raw materials and also by better mixing. Pure alumina and pure silica are the most commonly used starting material for the synthesis of mullite ceramics. Clay minerals such as china clay or kaolin, pyrophyllite, polymorphs sillimanite, andalusite, kyanite, *etc*., can be used as a starting material to fabricate mullite. This route uses synthesis mullite primarily for refractories and crucibles, plates, bushes, tubes, *etc*., used in high-temperature applications. Sintered mullite generally contains few impurities and those impurities that may minimize the reactivity of the raw material, which may decrease the quality of sintered mullite. Hamano *et al*. used high-purity kaolin to overcome this problem.[42] The impurity level reduces up to several hundred ppm (parts per million) only and uses grinded raw materials.[42,43] Low impurity content in raw materials may reduce glass content at the grain boundaries and increase its mechanical property.

4.2 Fused mullite

In this fabrication process, mullite has been synthesized by melting the raw materials in an electric furnace above the raw material's melting point (more than 2000 °C). The melted raw materials were cast into molds and kept at room temperature for cooling.[44] The raw materials used to make fused mullite are Bayer alumina, fused silica, quartz sands, rock crystals, *etc*. The chemical composition of fused mullite is around 83 wt.% (73 mol%, which is near about 2/1 mullite) of alumina has been reported, which is much more alumina content as compared to sintered mullite (about 73 wt.%) obtained by solid-state reaction. Due to the raw material's low impurity level, a better microstructure can be observed in the final product. Fused mullite is used for many applications such as shell building, glass contact refractories, kiln furniture, *etc*.[45-47] Various process parameters such as sintering temperature, duration, and composition of raw materials are vital for mullite crystal size but infused mullite, cooling condition plays an important role.[48]

4.3 Reaction mullite

Mullite coatings, films, and powder are prepared using advanced preparation techniques, such as solution-sol-gel, spray pyrolysis, chemical vapor deposition (CVD), *etc*. These processes are designated as reaction mullite or chemical mullite. The schematic microstructural model for particles is shown in Fig. 3, representing the mixing of Al^{3+} and Si^{4+} on a scale between microns to the atomic level.[49] The following section discussed the basics of each processing method.

4.3.1 Solution sol-gel method

The solution-sol-gel method utilizes atomic, molecular, or nanoscale mixing of the raw materials. This processing method is widely used to fabricate mullite powders, fibers, and dense samples at relatively low temperatures and short duration. Mixing raw materials at the atomic or molecular level increases the reaction rate by shortening the particle-to-particle or diffusion distance.[50]

Mullite prepared by this method, the homogeneous sub-micronic microstructure can be achieved. The particles in the solution-sol-gel method range nanometer range (nm), but in the sintered and fused synthesis method, the particles are in the micrometer range (μm). Hence nullification occurs at low temperatures in the range of 1200 °C to 1300 °C. This process comes under advanced ceramic processing techniques, and since the 1980s, the industry has adopted this processing technique rapidly.[51,52]

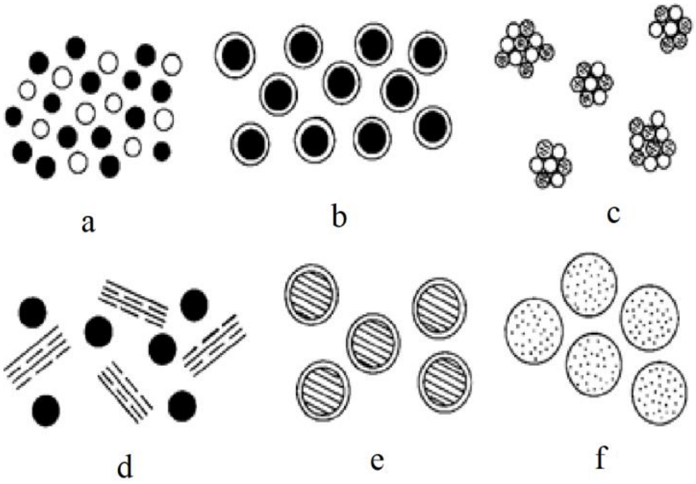

Fig. 3 Schematic microstructural model for mullite starting materials fabricated by different methods are (a) by sol mixing; (b) composite particles; (c) by hydrolysis of alkoxides; (d) conventional mixing of clay minerals; (e) by co-precipitation; and (f) by spray pyrolysis method. Reproduced with permission from [49], Copyright 2005 Teknik Mesin.

4.3.2 Spray pyrolysis method

The spray pyrolysis method consists of the atomization of droplets deposition from a solution. After deposition, it was dried and calcined in the furnace. Fig. 4 shows a schematic illustration of spray pyrolysis equipment. The reaction in this method is the evaporation of solvents, precipitations of compounds, and thermal decomposition; for that, this preparation method is considered appropriate for the preparation of mullite.[53]

The powder produced by this method has been analyzed in spherical shapes with a sub-micrometric to micrometer range. Droplets are prepared mainly in two different ways, using atomizers and ultrasonicator, respectively.[54]

4.3.3 The chemical vapor deposition method

The Chemical Vapor Deposition Method is an essential method used to synthesize mullite powder, films, and coatings. This method uses the vapor phase process to fabricate mullite. Few researchers studied the synthesis of mullite by using starting raw materials such as aluminum chlorides and silicon, where the chlorides are evaporated separately and transported with the help of nitrogen into the mixing area.[55,56] The materials were heat-treated by using hydrogen and oxygen combustion flame, and the estimated temperature was 1900 °C. In the reaction zone, the temperature reduces gradually, and the exit on 900 °C. The processing time during the deposition was nearly 65 milliseconds. After deposit, a spherical powder with a size of about 40 to 70 nm was obtained. A glassy phase along with small amounts of mullites was also observed in the specimen. Mullitisation occurs after firing the sample at 1000 °C for 10 min, and it requires 1500 °C for complete nullification. Various preparation routes used with different raw materials to synthesis mullite by other researchers are summarized in Table 2.

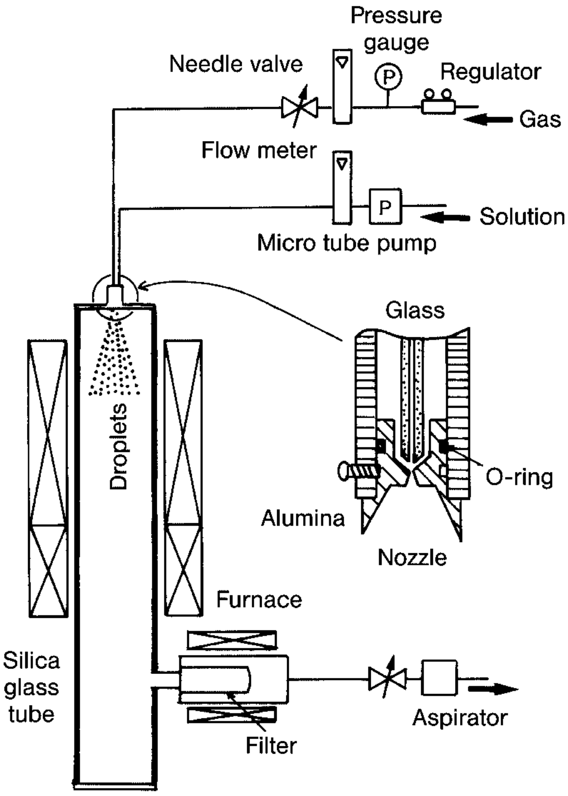

Fig. 4 Schematic diagram of spray pyrolysis equipment. Reproduced with permission from [53], Copyright 1990 Transaction British Ceramic Society.

5. Mullite as an essential oxide ceramic

5.1. Mullite applications

Mullite becoming very important for its outstanding properties were recognized in the 1970s.[57] Nowadays, mullite is one of the essential phases in modern oxide ceramics for its exceptional properties. In the beginning, mullite was used for pottery, whiteware, porcelain, refractories for metallurgical industries, kiln seals, temperature reactors, glass manufacturing, cement additives, chemical industry, tiles, pipe, *etc.*[5] However, over time researchers continuously explored its structure, properties, and various synthesis routes. As a result, during the last 10 to 15 years, the spectrum of mullite applications dramatically changes. Modern mullite ceramics applications are used to develop high-temperature composite

Table 2. Fabrication of mullite by using different synthesis parameters and their observations.

Researcher	Raw materials	Synthesis Route	Temperature	Observations
L. N. Bowen *et al.*[58] 1924)	Pure alumina and pure silica	Fusion process	1500 °C to 2000 °C	Mullites were prepared under the temperature of 1600 °C to 1700 °C, which consists of the stoichiometry of $3Al_2O_3{\cdot}2SiO_2$.
S. Prochazaka *et al.*[59] (1983)	Mullite powder and ZrO_2	Sintering process	1610 °C	High-density (3.77 g/cm^3) mullite bodies were prepared from mullite powders at 1610 °C.
K. C. Song *et al.*[60] (1998)	Tetraethyl orthosilicate and aluminum alkoxide	Sol-Gel method	600 °C to 1200 °C	A homogeneous solution of Al-Si gives mullite at 1000 °C.
Chang-Whan Won a *et al.*[61] (1998)	Silica and alumina	Sol-Gel method	600 °C to 1400 °C	Sol-gel and calcination method was successfully used to prepare Mullite powders at 600 °C. mullite preparation conditions were 0.5 mol of 2, 4 pentanedione. 100 mol of H_2O and an Al_2O_3/SiO_2 weight ratio of 3.
D. Amutha Rani *et al.*[62] (2000)	$Al(NO_3)_3$ nano-hydrate with ethyl silicate	Sol-Gel method	1600 °C	Mullite precursor was successfully derived from the sol-gel route. At room temperature, flexural strength was obtained 172 kg/cm^2 and compressive strength at 785 kg/cm^2, respectively.
C.Y. Chen *et al.*[63] (2000)	Kaolin and alumina	Sintering process	1000 °C to 1600 °C	The formation of mullite crystals starts at 1300 °C. The strength of the specimen sintered at 1500 °C, in the range between 170 to 195 MPa.
R. Baranwal *et al.*[64] (2001)	Aluminum hydroxide hydrate	The flame spray pyrolysis method	800 °C to 1000 °C	Ultrafine mullite particles were prepared by the flame spray pyrolysis method, and the size of the primary particles was 20 to 30 nm.
J. S. Jung *et al.*[65] (2001)	Coal fly ash and alumina	Sintering process	1400 °C to 1600 °C	At 1500°C mullite formation starts. The samples are sintered at 1400 °C, 1500 °C, and 1600 °C, which shows bending strength of 80, 169, and 115 MPa.
S. V. Sotirchos *et al.*[66] (2002)	$AlCl_3$, $SiCl_4$, H_2, and CO_2	Chemical vapor deposition (CVD)	1050 °C	An experiment was successfully conducted in a tubular CVD reactor to form mullite coatings.
G. Chen *et al.*[67] (2008)	Kaolin, $Al(OH)_3$ and AlF_3	Sintering process	1300 °C to 1550 °C	Prepared porous mullite obtained porosity of 27% to 55.6%, an average pore size of 0.73 to 1.50 µm, and mechanical strength of 15.5 MPa to 66.7 MPa.
Y. Dong *et al.*[68] (2011)	Bauxite, MgO and fly ash	Sintering process	1300 °C to 1500°C	The addition of magnesium oxide to the raw material admixture enhances the densification (2.8 g/cm^3). Height strength 47 MPa was obtained for the sample that contained 4 wt.% magnesia at 1400 °C.

Continued

Researcher	Raw materials	Synthesis Route	Temperature	Observations
R.M. Khattab *et al.*[69] (2012)	Silica fume, Alumina, Ball Clay, magnesia, and magnesia	Sintering process	1300 °C, 1350 °C & 1450 °C	The author successfully fabricated mullite at 1450 °C. The sample's bulk density, apparent porosity, and cold crushing strength were 2.10 g/cm3, 11.49%, and 489 MPa, respectively.
F. Yang *et al.*[70] (2012)	Mullite Powder	Sintering process	1300 °C to 1500 °C	The prepared porous mullites, apparent porosity, compressive strength, and thermal conductivity were 75.4~81.6%, 3.0~16.02 MPa, and 0.14~0.47 W/(m·K), respectively.
M. Srivastava *et al.*[71] (2014)	Sillimanite and alumina	Fusion method	200 °C to 800 °C	Mullite particles were synthesis in the thermal plasma reactor. The synthesized particles were reinforced in nickel. matrix to obtain Ni–mullite composite. Coatings. The Ni–mullite coating exhibited microhardness of 405 Hk at 800 °C.
Y. Li *et al.*[72] (2015)	Fly ash, clay, and alumina	Sintering process	1300 °C to 1500 °C	Mullite becomes the main phase with little corundum and quartz produced for the sample 30 % mass ratio of Al_2O_3 at 1300 °C.
X. Deng *et al.*[73] (2015)	Mullite powder	Sintering process	1600 °C	Porous mullite was obtained within 17% to 76% porosity, with an average pore size of 57–313 μm. Porosity with 76%, their compressive strength, and flexural strength were 15.3±0.3 MPa and 3.7±0.2 MPa, respectively.
J. Ma et al.[74] (2018)	Kaolin clay, alumina and boehmite	Sintering process	700 °C to 1200 °C	Mullite whiskers were formed at 900 °C. Height porosity and compressive strength obtained 80.10% and 11.12±0.14 MPa, respectively.
C. Thye Foo *et al.*[75] (2019)	Aluminum dross and fly ash	Sintering process	1200 °C to 1500 °C	Mullite was prepared at 1500 °C. The resultant ceramics exhibited good thermal expansion properties with a coefficient of thermal expansion values ranging from 4.0 to 5.9×10^{-6} °C^{-1}.
N. Ecebas *et al.*[76] (2019)	Mullite powder and titanium oxide	Sintering process	1450 °C to 1540 °C	Nano TiO_2 was added to form a liquid phase to densify the ceramic component. At 1540 °C, mullite was formed, and the dielectric constant was found as nearly 7.5 up to 10 GHz.
Mao Fu *et al.*[77] (2019)	MoO_3, fly ash, and $Al(OH)_3$	Sintering process	1100 °C to 1500 °C	High porous mullite ceramic membranes were fabricated at 1550 °C with a flexural strength of 100 MPa.

Continued

Researcher	Raw materials	Synthesis Route	Temperature	Observations
Y. Liu *et al.*[78] (2019)	Alumina refractory solid wastes and gangue	Sintering process	1500 °C	Fabricated mullite obtain 2.515 g/cm^3 density with fracture toughness, and bending strength values were 1.82 MPa·m$^{1/2}$ and 71.76 MPa, respectively.
P. Suhasinee Behera *et al.*[79] (2020)	kaolin, NH_3, glycine, $Al(NO_3)_3$, titanium dioxide, and MgO	Sintering Process	1500 °C to 1600 °C	1 wt.% TiO_2 and MgO show complete mullite phase formation at 1500 °C with a density of 2.91 g/cm^3 and flexural strength 126 MPa. At 1600°C, the achieved bulk density and flexural strength were 2.97 g/cm^3 and 156.33 MPa.
M. Rashad *et al.*[80] (2020)	China clay, aluminum fluoride trihydrate, and alumina	Sintering process	1400 °C	A monolithic needle-like shaped mullite crystal was synthesis at 1400 °C. Narrow pore size distribution with pure water permeance of 1031 L/m^2 h bar was obtained. The surface roughness value of the membrane is 157 µm.
S.K.S. Hossain et al.[81] (2020)	Alumina, rice husk, NaOH plates, HCl acid, and sodium silicate solution	Sintering process	1100 °C to 1400 °C	Mullite formation starts at 1250 °C. Mullite foamed at 1300 °C shows 11.07 MPa of compressive strength and 75.99% porosity.
F. R. Barrientos-Herna´ndez *et al.*[82] (2021)	Kyanite and aluminum	Sintering process	1100 °C to 1600 °C	The reaction between the kyanite and aluminum forms mullite at 1400 °C.

material, mullite films, coating, turbine engine components, infrared transparent windows, crucibles, thermocouple tubes, dental ceramic parts, multilayer packaging, radome manufacturing, *etc*.[3,6-16,75]

Presently, the commercially used other ceramic materials replaced by mullite-based ceramics are, for example; heat protection tubes, tube suppliers, Spyderco ceramic rods, mullite tubes, zirconia corundum mullite honeycomb, thermocouple protection tubes, *etc*.[83] Before, these things would have been made by using pure alumina, silicon nitride, silica, *etc*. As a result of using mullite, it gives a working temperature range between 1500 °C to 1600 °C in an oxidizing and reducing atmosphere, provides better thermal conductivity, better temperature uniformity, robust thermal shock resistance, low-cost raw materials required for mullite fabrication, and sustains for long life.

5.2 Mullite synthesis challenges

Mullite has a stoichiometry of 3Al2O3·2SiO2, stable in a narrow region in alumina and silica system under high temperature (called mullitisation temperature). This temperature depends on parameters such as raw material source, the particle size of raw materials, different synthesis methods, heating or cooling rate, *etc*. We have seen in Table 2 where other researchers have synthesis mullite, using an additional source of alumina and silica, followed by various synthesis routes, studying different temperatures. Every system has its individuality, especially the temperature, to form a mullite crystal. These things make mullite synthesis challenges to the researchers.

5.3 Cost-benefit of mullite ceramics

Commonly, mullite is fabricated from technical grade pure alumina,[62,57] and pure silica,[57] or commercial mullite powder[58,69,75] as raw material, making the overall manufacturing process expensive. An alternative way to lower the product cost by the use of waste materials which is a source of alumina and Silica, where some researchers used fly ash,[64,67,71,74] silica fume,[68] bauxite,[67] mining waste[84] as raw materials. This waste material poses several environmental problems.[85-89] In addition to the economic benefits, the use of waste materials brings out environmental benefits as it prevents the over-exploitation of natural resources and reduces the volume of waste that needs to be managed. Besides, the waste material utilization industry should consider the megatrends (social, economic, political, environmental, or technological changes) as driving technology demands for future economic growth.

6. Conclusion and future scope of research

In the modern ceramic world, mullite is becoming one of the most important phases. During the last two decades, mullites applications have changed remarkably from its classical applications like pottery, whiteware, porcelain, refractories, building blocks, tiles, *etc*., to modern applications, such as infrared transparent window, composite, turbine engine components, radome, *etc*. This review paper describes mullite crystal structure, and essential properties and describes all possible synthesis methods, mullite synthesis challenges, *etc*. The primary motivation for researchers has to utilize waste materials to fabricate mullite-based ceramics. It can be an excellent effort to minimize ecological problems, reduce the volume of waste materials and most importantly, cut down the overall cost of the product. In addition, increasing the area of mullite-based ceramics applications, finding more synthesis routes, forming complex components, and utilization of raw materials will play an increasingly important role shortly.

Conflict of interest

There are no conflicts to declare.

Supporting information

Not applicable.

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