International Journal of Applied Nanotechnology Research
Volume 5 • Issue 1 • January-June 2020

Synthesis and Characterization of Mullites From Silicoaluminous Fly Ash Waste

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ABSTRACT

Fly ash is considered one of the major hazardous pollutants around the globe. Every year a million tonnes of fly ash is disposed of into the fly ash ponds which are major sites of pollution. The major fractions of fly ash are silicates, aluminates, and ferrous substances followed by minor traces element oxides. The aluminates and silicates comprise of 70% of the fly ash. The aluminates and silicates are present in fly ash in the form of crystalline mullites and sillimanites. Mullites being inert and crystalline are retractile to mineral acids. So, here the authors have reported a novel and simple step for the recovery of all the major elements of fly ash along with recovery of mullites by using hydrofluoric acid at room temperature. The method comprises of treatment of fly ash with diluted hydrofluoric acid for 12 hours under agitation. The recovered white color mullite powder, rod shaped of size 90-300 nm, was analyzed by the sophisticated instruments for the confirmation of the mullite particles.

KEYWORDS

Aluminosilicate, Fly Ash, Mullites, Refractile, Sillimanites

1. INTRODUCTION

Mullite is an exceptionally advanced solid solution of aluminum silicate ($3Al_2O$. $2SiO_2$), which is developed by the sintering of rare raw minerals that consists of alumino-silicate under extreme temperature and low pressure (Wang & Sacks, 2005). Mullites are crystalline compositions prominently, comprising of elements like Al, Si, and O (H. Schneider, R. Fischer, & J. Schreuer, 2015a). They are non-stoichiometric compounds structurally similar to impure magnetite which belong to the compositional series of orthorhombic alumino-silicates with the general composition $Al_2(Al_{2+2x}Si_{2-2x})O_{10-x}$ (Li & Thomson, 1991)(Fischer, Gaede-Köhler, Birkenstock, & Schneider, 2012). They usually

DOI: 10.4018/IJANR.20200101.oa2

exhibit two stoichiometric forms, i.e. $3Al_2O_32SiO_2$ or $2Al_2O_3SiO_2$ and can be characterized by using high-end techniques like X-ray diffraction (XRD) and Scanning electron microscope (SEM) in the fly ash (Y. Gong, Sun, Sun, Lu, & Zhang, 2019). Mullites are found in high temperature metamorphosed rocks of the sanidinite and clay (Laita, Bauluz, & Yuste, 2019). They are also known as "porcelainite" as they are moulded under extremely high temperature using clay (Tripathi, Ghosh, Halder, Mukherjee, & Maiti, 2012). Among the minerals, mullites are also found in hornfels rock (porcellanite) (Searle, 1962), e.g., at the point of contact of bauxites with olivine dolerite intrusions. Special and rare occurrences of mullite are in alumino silicate lechatelerite glasses produced by lightening impact in sandstones (Pasek, Block, & Pasek, 2012). In addition to this, mullites presence can also be found in small druses of volcanic rocks (e.g., in the Eifel mountain, Western Germany), where it probably grew under moderate hydrothermal conditions (H. Schneider, Schreuer, & Hildmann, 2008).

They own magnificent and unique properties in the form of needles in porcelain (Martin-Marquez & Romero, 2010) like low thermal expansion (Oikonomou, Dedeloudis, Stournaras, & Ftikos, 2007), low thermal conductivity (L. Gong, Wang, Cheng, Zhang, & Zhang, 2014), high thermal and corrosion stability (Baspinar & Kara, 2009), high strength (Liu, 2011), high fracture toughness (Santos & Rodrigues, 2003), excellent creep resistance (Torrecillas et al., 1999), good thermal shock and stress resistance (Uribe, Moreno, & Baudín, 2001), good strength, wear-resistant and useable to high temperatures (H. Schneider, Schmüker, & MacKenzie, 2005). In addition to this, mullite is the only stable binary phase existing system of the $A1_2O_3$-SiO_2 under ambient conditions (Martin-Marquez & Romero, 2010). Its chemical configurations empirically include 71.8 wt.% $A1_2O_3$ and 28.2 wt.% SiO_2, which is designated as 3/2- mullite ($3A1_2Oy_2SiO_2$) (Ohtake et al., 1991). Moreover, mullite has no charge balancing cations present in them (O'Connor, Mackenzie, Smith, & Hanna, 2010). As a result, there are three different aluminium sites: two distorted tetrahedral and one octahedral (H. Schneider, R. X. Fischer, & J. Schreuer, 2015b). They have two common morphologies: platelet shaped and needle shaped. Platelet shape particles have low aspect ratio while needle shape particles has high aspect ratio. If it forms during the process of sintering, then it provides increased mechanical strength and thermal shock resistance (Chen, Lan, & Tuan, 2000).

Mullite is undoubtedly one of the most imperative oxide material for the both, the conventional and the advanced ceramics (Igo, 2019). Mullite is formed in fly ash during various melting and firing processes, and is used as a refractory material due to its high melting point of 1840 °C (Kamara, Wei, & Ai, 2020). Besides this, due to their distinctive properties, it treasures applications in ceramic whiteware (Sadik, Amrani, & Albizane, 2014; Hartmut Schneider et al., 2015b), porcelains (Anggono, 2005), high-temperature insulating refractory materials (Aksel, 2003), furnace liners (Sadik, El Amrani, & Albizane, 2014), electrical insulators, protection tubes, kiln furniture, rollers, heat exchanger components, heat insulation parts, pressed parts (Eom, Kim, & Raju, 2013) and isostatically pressed parts. It also is utilized as a synthetic analog of mullite that can be an effective standby for platinum in diesel engines (Cui, Zhang, Fu, Wang, & Zhang, 2020; Eom et al., 2013).

The composition of mullite is commonly denoted as $3Al_2O_3.2SiO_2$ (71.83 wt. % Al_2O_3) (T. F. Choo, Mohd Salleh, Kok, Matori, & Abdul Rashid, 2020). However, commercially available mullite which is a solid solution generally consists of 71-76 wt.% Al_2O_3, 23-24 wt. % SiO_2, and small quantities of TiO_2, Fe_2O_3, CaO, and MgO (T. F. Choo et al., 2020). Stoichiometric ($3Al_2O_3.2SiO_2$) mullite can be produced without a glassy grain boundary phase that results in an extraordinary strength being maintained at high temperatures (Treadwell, Dabbs, & Aksay, 1996).

The importance and the demand for mullites are progressively increasing day by day in the various industries due to their exceptional properties. The current commercial production of mullites in industries takes place from the rocks (Toya, Tamura, Kameshima, & Okada, 2004), which is laborious, energy intensive and also an expensive process. Besides this, it is also produced in the laboratory by executing any of these methods i.e. sintering of raw materials of Al, and Si carbide, organic precursors, a sol-gel technique (Jurado, Arévalo Hernández, & Rocha-Rangel, 2013; Won & Siffert, 1998), and chemical vapor deposition (Mulpuri & Sarin, 2011). But, as all the approaches