ROLE OF ENERGY AND GAS PRESSURE ON PHASE EVOLUTION OF NANO-ALUMINA IN WIRE EXPLOSION PROCESS

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Abstract

Phase control of alumina nanoparticles (NPs) is successfully shown by wire explosion process under gas ambient. Aluminium wire was exploded at different levels of energy in oxygen ambient under pressures of 20 and 180 kPa. X-ray diffraction patterns indicate that the 20 kPa tests yield δ-alumina while 180 kPa tests yield γ-alumina NPs with an appropriate value of energy. Spherical NPs were obtained as visualised from SEM and TEM micrographs. Size of NPs decreased with an increase in energy and/or a decrease in pressure of oxygen. The formation mechanism of different phases and sizes of NPs are detailed in this paper.

1 Introduction

Ceramics are widely used in applications ranging from home utensils, parts of airplanes to insulation engineering. Alumina is one of the key ceramic materials used by industry. Alumina produced in nanostructures provides the advantages of high surface area and more reactive sites. Alumina nanoparticles (NPs) find applications in numerous fields like a support in catalyst science [1] or as a filler in insulation engineering [2-3]. Properties of nanomaterials are highly dependent on its size and phase composition. Dependency of catalytic property of alumina NPs on its phase composition is shown in [2, 4]. Hence, it is necessary to produce different phases of alumina NPs with controlled size to get the required characteristics.

Several methods have been developed and devised to produce Alumina NPs, e.g. high temperature oxidation of boehmite powder [5], or precipitation method [6]. Recently, a single method wire explosion process (WEP) was employed to produce alumina NPs in oxygen gas ambient [7] and in deionized water [8]. WEP (also known as pulsed wire discharge, PWD) is a top-down approach where a wire (aluminium, Al) and the cooling/reacting medium (oxygen) are the starting materials. This process is based on Joule heating of the wire when a high current passes through it. As the energy deposited on the wire exceeds the required vaporisation energy, the wire transforms into vapour/plasma state. The ambient (oxygen gas) reacts with the Al vapour and simultaneously cool down. Fast quenching of the vapour leads to formation of alumina NPs. Many types of NPs like metallic, oxides and alloys could be produced by changing the wire and using an appropriate ambient [7-11]. NPs produced by WEP have been tested for many applications. In short, WEP is a reliable method to produce NPs and with a high production rate [11].

Tokoi et al. [8] discussed phase change of alumina NPs produced by WEP with change in ambient from oxygen gas to deionized water. However, there was no change in NPs phase for explosion in gas at various energy/pressure conditions. NPs generation in gas ambient is of increasing interest as it provides an economical solution for relevant industries.

In the present work, phase evolution of alumina NPs was investigated through WEP with change in energy and oxygen ambient pressure. The phase and crystalline structure of NPs were verified using X-ray diffraction (XRD) technique and the morphology was checked through electron microscopy imaging. Particle size distribution study was carried out to validate the effect of energy and ambient pressure on the size of the formed NPs. In the end, temperature and saturation ratio effect were discussed to elucidate the formation mechanism of NPs.

2 Methodology

Enter methodology section here. Alumina NPs were prepared using the setup shown in Fig. 1. Aluminium wire (0.5 mm diameter) was tied between two electrodes inside the explosion chamber. The stored energy ($E_C$) in the capacitor bank was used to overcome the vaporisation or sublimation energy ($E_S$), which is the minimum energy required to transform the solid to vapour state of the wire. Supply voltage of 230 V, 50 Hz AC was fed to auto-transformer (0-
230 V) and then the high voltage transformer (230 V / 25 kV, 25 kVA) to step up the voltage to the required level. DC voltage, Vc was obtained after rectification by diode. Ec (0.5CVc^2) was discharged through a trigatron gap switch after triggering.

Table 1 shows the experimental parameters used in the present work. First Es of Al was calculated using the relation,

\[ E_s = C_s \times (T_m - T_r) + C_f \times (T_b - T_m) + h_m + h_v; \]

Where Cs and Cf are specific heat in the solid and liquid states of Al respectively, Tr, Tm, and Tb are room, melting and boiling temperature of Al respectively, and hm and hv are the latent heat of fusion and vaporization of Al respectively. We assumed equal specific heat in the solid and liquid states of Al. The data for the calculation were taken from the CRC databook [15]. Energy ratio, Er is defined as the ratio of Ec and Es. Er and oxygen pressure, P were the two control parameters used in the present work.

Table 1: WEP experimental parameters

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Value</th>
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<tbody>
<tr>
<td>Capacitance, C (µF)</td>
<td>3</td>
</tr>
<tr>
<td>Wire diameter (mm)</td>
<td>0.5</td>
</tr>
<tr>
<td>Length of Al wire (mm)</td>
<td>10-50</td>
</tr>
<tr>
<td>Charging voltage (kV)</td>
<td>27</td>
</tr>
<tr>
<td>Oxygen absolute pressure (kPa)</td>
<td>20, 180</td>
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</tbody>
</table>

Prepared NPs were characterised using XRD (Bruker D8 discover, Cu-ka radiation, 0.15418 nm) for phase identification. X’pert high score software was used for the refinement of XRD patterns. Morphology was observed with scanning electron microscope (SEM; FEI Quanta FEG 200) and transmission electron microscope (TEM; Philips CM 12). Sample for TEM was prepared using carbon coated copper grid. First, a small quantity of NPs was dispersed in acetone using ultrasonic bath. Then, two drops of the colloid was dispensed on grid and left to dry. The images of NPs obtained for equivalent circle of 2-dimensional projection of the

3. Results and Discussion

Fig. 2 shows the XRD patterns of NPs synthesised by WEP for different Er (3, 5, 7 and 15) and P (20, 180 kPa) and standard patterns (top and bottom vertical lines) for two phases of alumina. Patterns matched with γ-alumina (JCPDS 010-0425) and δ-alumina (JCPDS 016-0394). For Er of 15, some peaks correspond to Al (JCPDS 004-0787) also.

Fig. 2 XRD patterns of alumina NPs generated by WEP tested for different Er under pressures of 20 and 180 kPa.

Complete oxidation of Al happens for all cases except Er of 15 as indicated in Fig. 2. If, we consider the cases for P at 20 kPa for all Er, most of the peaks correspond to δ-alumina (double peaks about 45° and 67°) while for P at180 kPa, peaks correspond to γ-alumina (single peak about 45° and 67°). For Er of 5, 7; and P at 180 kPa, pure δ-alumina while for P at 20 kPa, γ-alumina was obtained. For high Er of 15, presence of Al is due to non-oxidation of vapour. This could be attributed to the smaller volume of chamber used in the present work.

Fig. 3 shows the SEM and TEM images of NPs obtained by WEP. NPs are spherical in shape for all the cases. Particle size varies between few nm to 200 nm. Log-normal distribution of size data was plotted in Fig. 4. Table 2 shows the mean size and standard deviation of WEP synthesised
NPs. Size of the nanoparticles reduces with increase in ER and/or decrease in P. This is due to fast quenching of vapour/plasma of Al discussed in detail henceforth.

Table 2 Mean size and standard deviation of alumina NPs generated by WEP tested for ER of 3 and 5 under pressures of 20 and 180 kPa.

<table>
<thead>
<tr>
<th>ER and P (kPa)</th>
<th>Mean particle size (nm)</th>
<th>Standard deviation (nm)</th>
</tr>
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<tbody>
<tr>
<td>3, 180</td>
<td>55.2</td>
<td>1.5</td>
</tr>
<tr>
<td>5, 180</td>
<td>45.2</td>
<td>1.8</td>
</tr>
<tr>
<td>5, 20</td>
<td>33.8</td>
<td>1.7</td>
</tr>
</tbody>
</table>

Fig. 3 SEM and TEM images of alumina NPs generated by WEP tested for ER of 3 and 5 under pressures of 20 and 180 kPa.

Fig. 4 Particle size distribution of alumina NPs generated by WEP tested for ER of 3 and 5 under pressures of 20 and 180 kPa.

Fig. 5 shows the possible formation mechanism of alumina NPs with δ and γ phases of different sizes. The symbols T and S indicate temperature and saturation ratio respectively. Embryo is the size of the stable nuclei which grows to form NPs. Increase in energy leads to high T and decrease in ambient pressure leads to high S. With increase in T and/or S, the size of the embryo gets smaller (T₁ and S₁). For smaller embryo, NPs will be smaller (path A) if the cooling rate is high. For low cooling rate, NPs will grow slowly into big size (path B). Interfacial nucleation of smaller embryos (path C) will lead to formation (path D) of bigger size NPs. For bigger embryos, formed NPs will be larger (path E). Different cooling rates at different positions of vapour cloud (it is impossible to have one cooling rate practically in WEP) is the reason for mixed size particles as shown in Fig. 3. As we increase ER, we are increasing the specific energy (energy per unit of mass of the wire), which leads to more expanded vapour meaning less dense plasma subsequently leading to faster cooling resulting in low dimensional NPs as tabulated in Table 2. For decrease in P also, vapour expansion is more and it leads to small sized NPs. Decrease in activation barrier and size of embryo supported with increase in nucleation rate of the NPs formation lead to small size NPs [10]. This condition was obtained with increasing ER and decreasing P in the present work.

More amount of oxygen (P at 180 kPa) leads to formation of γ-alumina (Fig. 2). For high pressure ambient, the vapour/plasma will be denser than that at low pressure leading to slow cooling. Slow cooling (path B of Fig. 5) leads to low temperature phase of γ-alumina. For low P (20 kPa) tests, faster cooling (path A of Fig. 5) leads to δ-alumina (Fig. 2). Similar results
were obtained by Tokoi et al. [11] when they compared the formation of different phases of alumina in deionized water.

4 Conclusion

Alumina nanoparticles (NPs) of different phases were synthesised using WEP starting with Al wire. For low and high pressure oxygen ambient, δ-alumina and γ-alumina NPs were obtained respectively. However, for high ER, some traces of non-oxidised Al was also obtained. Size of NPs follows log-normal distribution and it decreases with increase in energy and/or decrease in pressure. Smallest mean size NPs of 33 nm was obtained for ER of 5 and P at 20 kPa. One has to be careful while selecting ER values as high ER yields in non-oxidation of vapour resulting in unwanted traces of metallic NPs.

6 References