



Effect of Y_2O_3 on acid resistance of alumina ceramic

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ARTICLE INFO

Keywords:

Alumina ceramic
Yttrium oxide
Acid resistance
HCl-HF solution

ABSTRACT

This work aimed to improve the acid resistance of an alumina ceramic. Acid corrosion of alumina ceramic composed of Al_2O_3 - $CaCO_3$ - SiO_2 - MgO - Y_2O_3 (ACSMY) was investigated in a hydrochloric-hydrofluoric acid solution at 65 °C for 30 min. The effect of Y_2O_3 content on sintering temperature, density, and acid solubility were discussed. The composition and microstructure of this material were analyzed. The acid solubilities of minor crystal phases ($Y_3Al_5O_{12}$, $CaAl_{12}O_{19}$, $Ca_2Al_2SiO_7$, and $CaAl_2Si_2O_8$) and the effect of them on acid resistance of this alumina ceramic were studied. The results showed that Y_2O_3 additive can enhance density and change the type of phases. Phases with good acid resistance and dense structure lead to a crust formed on the surface of ceramic during acid corrosion. The crust can effectively protect the interior structure of the sample from acid solution, and then improve the acid resistance of the material.

1. Introduction

The application areas of ceramic with good corrosion resistance are very wide, such as the industries of bio-pharmaceuticals, food and beverage processing, environmental protection, water treatment, smelting, chemical fiber, gas or oil exploration, etc. [1–8]. Corrosion resistance of ceramic plays an important role in the security and lifetime of products. Chemical corrosion resistance of alumina ceramic is powerful, due mainly to the strong binding force of aluminum ion with oxygen ion. Another reason is that alumina is amphoteric oxide. So alumina ceramics can be used in many industrial fields. In particular, an increasing number of applications in very harsh environments need alumina ceramics with high corrosion resistance, such as ceramic proppants.

Proppants, used to “prop open” rock cracks to increase oil well production, work underground over distances of several kilometers. So they must endure all kinds of harsh environments, such as high temperatures and pressures from both the ground water found in deep wells and the HF-HCl mixtures sometimes pumped into wells to clear blockages, and corrosion from various corrosive media in the crust and fracturing fluid. And, more and more low-permeability and deep oil wells are being mined using acidification and fracturing composite technology, which necessitates a higher standard of acid resistance in proppants. Petroleum and Gas Industrial Standard (SY/T5108-2006) requires the acid solubility is less than 5%. However, acid resistance of ceramic proppant products on the market is poor. The acid solubility usually reaches about 6–8%. Individual product is up to the standard

and the acid solubility is 2.1%, but it is still on the high side. So it is very important to improve the acid resistance of alumina ceramic.

Numerous papers have been published on the corrosion of alumina ceramics under various acidic conditions. Schacht et al. [9] reported the corrosion resistance of alumina ceramics (99.99% alumina) in aqueous acidic solution (HCl, H_2SO_4 , and H_3PO_4) under hydrothermal conditions. The authors found that most grains were attacked by the formation of etching channels due to the dissolution of alumina in HCl and H_2SO_4 . However, alumina ceramics were attacked by a formation of brilliant ($AlPO_4$) in phosphoric acid. The corrosive effect of the acids on alumina ceramics decreases in the order $H_3PO_4 > HCl > H_2SO_4$. Lidija et al. [10] researched the corrosion behavior of high purity alumina ceramics in aqueous HCl and H_2SO_4 solutions at room temperature (25 °C). The authors concluded that the stability of high purity alumina ceramics is low at low concentrations of both acids and the corrosion is mainly attributed to the dissolution of MgO, SiO_2 , CaO, Na₂O and Fe_2O_3 grain-boundary impurities. The dissolution of Al^{3+} in bulk material (Al_2O_3) is negligible. They [11] also pointed that the degree of dissolution in a descending order is as follows in HCl: Ca > Si > Na > Fe > Mg > Al. Mikeska et al. [12] investigated the corrosion resistance of ceramics in HF acid. They indicated that corrosion in the case of polycrystalline alumina ceramics with 99.9% Al_2O_3 occurred primarily at the grain boundaries. But, single-crystal sapphire (Al_2O_3) did not corrode and showed good resistance to the HF acid.

Most literatures deal with the corrosion of high purity alumina ceramics (> 99%) and reveal the dominant corrosion mechanisms that are intergranular attack and dissolution of Al_2O_3 . In general, proces-

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<http://dx.doi.org/10.1016/j.ceramint.2017.01.023>

Received 23 July 2016; Received in revised form 6 January 2017; Accepted 6 January 2017

Available online 07 January 2017

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Table 1
Chemical composition of samples (wt%).

| No. | Al ₂ O ₃ | Y ₂ O ₃ | CaCO ₃ | MgO | Kaolin |
|-----------------|--------------------------------|-------------------------------|-------------------|-----|--------|
| Y ₀ | 90 | 0 | 3 | 3 | 4 |
| Y ₁ | 89 | 1 | 3 | 3 | 4 |
| Y ₃ | 87 | 3 | 3 | 3 | 4 |
| Y ₅ | 85 | 5 | 3 | 3 | 4 |
| Y ₇ | 83 | 7 | 3 | 3 | 4 |
| Y ₉ | 81 | 9 | 3 | 3 | 4 |
| Y ₁₁ | 79 | 11 | 3 | 3 | 4 |

sintering and manufacturing of pure alumina products is a difficult and expensive task. Therefore, additives are added to alumina to minimize manufacturing costs. Besides, the acid resistance of alumina ceramics can be greatly altered by the presence of minor constituents [13]. However, less research about the effect of minor crystal phase on acid resistance has been done. In the present study, we focused on the effect of Y₂O₃ on the acid resistance of high alumina ceramic in an ACSMY system and choose proppant as an entry point. Through the design of the formulation and control of the preparation process, an alumina ceramic with good acid resistance is gained. The sintering temperature, acid solubility, phase, and microstructure were investigated to provide strong evidences for the acid resistance mechanism.

2. Experimental

The samples were prepared using a commercial alumina powder (purity > 99 wt% and a mean particle size of 0.65 μm). The alumina powder was mixed with CaCO₃, MgO, kaolin and Y₂O₃ in specific proportions (Table 1). A homogeneous mixture, obtained by ball milling for 48 h, was dried in an oven and then formed into spherical green bodies with 16–20 mesh using a sugar-film coating machine (Temperature: 25 °C, Air pressure: 5 kg/cm³). The green bodies were sintered at different sintering temperatures in air for 1 h at a heating rate of 5 °C/min.

Water absorption is used to judge sintering temperatures of the samples. A sample was weighed (W_1). Then put it into water. After 30 min, wipe the water from the surface of the sample for weighing (W_2). Water absorption is determined by the formula: $W = (W_2 - W_1) / W_1 \times 100\%$. The W is water absorption, the W_1 is the weight of dry sample (g), and the W_2 is the weight of sample absorbed water (g). The tests of density and acid solubility are determined by the Chinese Petroleum and Gas Industrial Standard (SY/T 5108-2006). Density was measured by the Archimedes method using deionized water as the immersion medium. Density is calculated by the following formula: $\rho_a = M_s / V_s \times 100\%$. The M_s is the weight of dry sample (g) and the V_s is the volume of sample (cm³). The acid solubility test is performed in 12-3 hydrochloric-hydrofluoric acid solution (i.e., 12 wt% HCl + 3 wt% HF) at 65 °C for 30 min. The testing process is as follows for each sample: about 5 g (W_s) samples were put into 100 ml acid solution and heated in a water bath at 65 °C for 30 min. A filter was weighed (W_r). Then put the samples after acid treatment into the filter. They were washed with deionized water and then dried in an oven at 105 °C. Finally, the dry filter and samples were weighed (W_{fs}). The acid solubility is calculated by the following formula: $S = (W_s + W_r - W_{fs}) / W_s \times 100\%$. The S is acid solubility, the W_s is the weight of samples before hydrothermal treatment in acid solution (g), the W_r is the weight of filter (g), and the W_{fs} is the weight of filter containing samples after hydrothermal treatment in acid solution (g).

Phases of the samples before and after acid treatment were tested by XRD. The test was carried out in an “X’Pert PRO” multi-purpose X-ray diffractometer (PANalytical B.V., Almelo, Netherlands) with 40 kV and 40 mA. X-ray patterns were taken by measuring 2θ from 10° to 90°, at a step size of 0.02° and a dwell time of 5 s per step. The results of the powder diffraction patterns were analyzed with X’Pert High

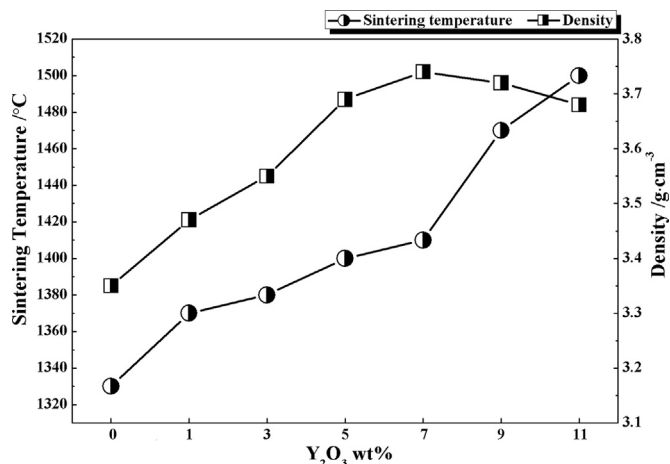


Fig. 1. Curves of relationship among sintering temperature, density, and Lu₂O₃ contents.

Score Plus software. Cross-sectional specimens were sputter coated with gold and observed by a Hitachi S4800 (Hitachi, Japan) field emission scanning electron microscope (FESEM) equipped with an energy dispersive X-ray spectroscopy (EDS) analyzer.

3. Experimental Results

3.1. Effect of Y₂O₃ on sintering temperature and density

The water absorption of the sample can be used to judge sintering temperature. The sintering temperature was determined by the water absorption that is 0% and the acid solubility that is the lowest. Fig. 1 shows the relationship among Y₂O₃ content, sintering temperature and density. The sintering temperatures of the samples increase with Y₂O₃ content. However, the density curve varies parabolically with the increase of Y₂O₃ content. The sintering temperature of Sample Y₀ without Y₂O₃ is 1330 °C. When Y₂O₃ content is added from 1 wt% to 11 wt%, the sintering temperatures increase from 1370 °C to 1500 °C. The result shows that Y₂O₃ additive can increase the sintering temperature of this material. The densities of the samples increase firstly and then decrease. When Y₂O₃ content is 7 wt%, the density reaches the maximum (3.74 g/cm³). It is indicated that the density of alumina ceramic can be improved by a proper addition of Y₂O₃.

3.2. Effect of Y₂O₃ on acid resistance

The acid solubility decreases firstly and increases subsequently with the increase of Y₂O₃ content (Fig. 2). The acid solubility test runs on

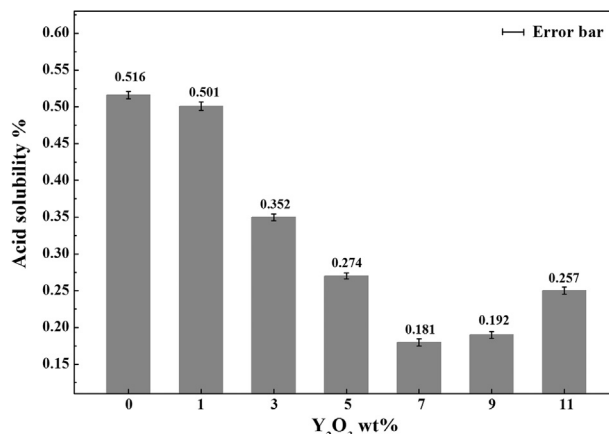


Fig. 2. Acid solubility of samples with different Y₂O₃ contents.

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