Yttrium disilicate stability in aqueous medium

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ABSTRACT

Recently Brazil has been considered as ascending potency from South America as 6th biggest economy in the world, whose international political is seen as "Soft power", which means diplomacy, economic development and common interests. To enhance the progress of Brazilian nation is extremely important to establish a sustainable energy security program, wherein the processing of materials to save energy plays an important role. Yttrium disilicate ($Y_2Si_2O_7$) presents considerable luminescent proprieties making it a potential substitute of rare earth oxides in gas burners technology. In this work $Y_2Si_2O_7$ was synthesized by a simpler hydrothermal method and its stability in aqueous media was evaluated by measuring electrophoretic velocity of particles, zeta potential determination and flow curves in controlled shear rate mode. As a result yttrium disilicate was more stable at alkaline pH range, showing a minimum viscosity at pH 11.

Key words: disilicate, ceramic processing, rheology.

Introduction

Among renewable sources, biogas derived from anaerobia decomposition of organic wastes (urban, rural and sewage) is a promise and sustainable energy alternative, besides its use can become a wise solution for garbage management, which is widely subject of many studies[1-6]. Since biogas is in essence a set of gases as CH_4 (50%), CO_2 (40%), Ni_2 (7%), O_2 (3%)[3], it may replace natural gas, or other fossil fuels in engines and lighting technology. Promoting the use of modern biomass as energy source is essential in order to decrease the dependency in fossil fuels as well an important improvement in energy security program.

Lighting emission by gas combustion process in porous components is more efficient rather than a free flame, seeing that porous structures work as a heat circulator improving flame speed and stability[7]. Moreover, this technology fulfills requirements on lower pollution emissions (NOx, CO), according to United Nations' Low Carbon plans[8]. In this way, many efforts have been done in order to improve combustion efficiency of porous burners[1, 4, 9-13], where materials selection is a fundamental step.

Rare earth disilicates (RE:Y_2O_3:SiO_2) have been very studied due to its magnetic, electric and optical proprieties. Yttrium disilicate presents important structural proprieties as refractoriness (1777°C) and stability in oxidant environmental. In addition, it is very applied in microwave technology, ferromagnetic, lasers and phosphors. Among its five polymorphs ($Y, \alpha, \beta, \delta, \gamma$) the last one is the most stable phase and presents significant mechanical proprieties like yttrium oxide and silica. These proprieties associated with its luminescence[14, 15] make it a potential material to be applied in gas burners technology for lighting. However, many contributions have been reported that is not easy to synthesis this material in a single phase[16-19]. Besides, there is no publication on colloidal stability of yttrium disilicate. Thus, this work is aimed to evaluate some processing parameters of yttrium disilicate, which are important to prepare porous ceramics based on stable suspensions to be applied in replica processing.

Experimental

Gama phase yttrium disilicate powders ($Y_2Si_2O_7$-γ) synthesized from hydrothermal method adapted by our research group[15]. Main powder characteristics: mean particle size (d_{50}=364.36nm), density (ρ=4.04g.cm^{-3}) and specific surface area (SSA=10.92m^2.g^{-1}).

The stability of $Y_2Si_2O_7$ nanoparticles (Zeta Potential, ζ) in aqueous medium was evaluated by measuring the electrophoretic mobility of the particles and determining Zeta potential (ζ) using a zeta-meter based on light phase scatter analyzing (ZetaPALS, Brookhaven Instruments Corporation, USA). Stock solutions with 0.5 g.L^{-1} of solids concentration were prepared, having NaCl 10^{-3}M as indifferent electrolyte (Casa Americana, Brazil). HCl and

KOH solutions were used to set pH of stock solution from acid to alkaline (pH=5.6-12). In order to compare the effectiveness of dispersant dosage on promotion of stability, concentrations from 0.5 to 2wt.% (weight percent, wt.%) of polyacrylic ammonium acid (PAA, Duramax D3005, Rohm and Haas Co., USA) were added in stock solution. Before measurements, all suspensions were homogenized in ultrasound cleaner for 2min (Dr. Hielscher 400US, Germany).

Suspensions elaboration: $Y_2Si_2O_7$ aqueous suspensions were prepared varying solids load from 5 to 25vol.% (volume percent, vol.%). In order to improve the stabilization of the suspensions was used as dispersant polyacrylic ammonium acid (PAA, Duramax D3005, Rohm and Haas Co., USA). To set pH of suspensions to alkaline medium Tetramethylammonium Hydroxide (TMAH, Sigma-Aldrich, USA) was used. Carboxymethyl-Cellulose (CMC, Sigma-Aldrich, USA) as a binder from 0.3 to 1wt.% (based on suspension weight). All suspensions were homogenized in ball milling for 24hs, using alumina balls ($\emptyset_{balls}$=10mm).

Rheological characterization: the flow behavior of suspensions was performed by a rheometer (Haake RS600, Thermo Scientific, Germany). The sensor system consisted on a double cone rotor and a stationary plate (DC60/1°). Suspension characterizations were carried out by flow curves in a control rate mode (CR). Measurements were performed at 25°C by increasing the shear rate from 0 to 1000 s^{-1} in 5 minutes, holding at 1000 s^{-1} for 2 minutes and returning to 0s^{-1} in 5 minutes. For each CR run 200 points were measured.

Results and discussion

Zeta potential curves of $Y_2Si_2O_7$-γ nanoparticles as a function of dispersant concentration (PAA - weight percent, wt.%) and pH are presented in Figure 1. Suspensions with no dispersant (0wt.%PAA) presented an isoelectric point (IEP) at pH=7.3. At this condition Zeta potential is zero (ζ=0mV), which means no repulsion between particles by electrostatic mechanism[20]. To prepare stable suspensions is desired to set up pH value far from IEP. For all pH range evaluated the stability of $Y_2Si_2O_7$-γ takes place at pH<6.5 (|ζ=38mV|)

and pH≥8.0 (|ζ=50mV|). Using polyacrylic ammonium (PAA) as dispersant promotes stability of particles by electro steric mechanism[20]. The prior interface liquid/particle surface changes to liquid/dispersant/particle surface. As a consequence Zeta potential and IEP are resulted from this interaction. Suspensions prepared with 0.5wt.% of PAA presented stable conditions at 6.5>pH≥7.5. In addition, for 1wt.% of PAA the stability is achieved at 6>pH≥7.0, whereas for 2wt.% of PAA this condition is established at 5.5>pH≥6.5, resulting in highest zeta potential values (|ζ=70mV|). However, using 3wt.% of PAA promoted an increase on ionic strength of medium and consequently decreased the repulsive forces between particles. Based on these results, using 2wt.% of PAA and setting up pH=9.0 were the best parameters to prepare highly stable suspensions.

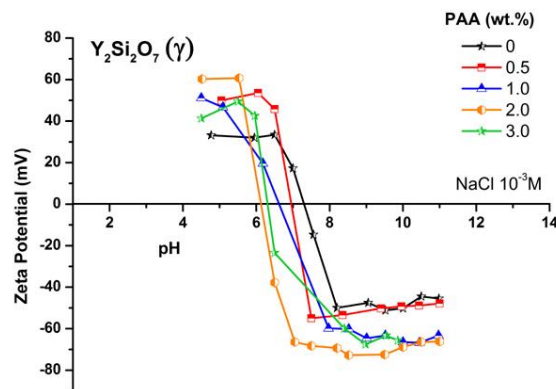Figure 1. Zeta potential curves of $Y_2Si_2O_7$-γ particles as a function of pH and dispersant (PAA, wt.%) concentration.

The flow behavior of $Y_2Si_2O_7$-γ suspensions was evaluated by control rate curves (CR) as shown in Figure 5a. Suspensions with up to 15vol.% of solids showed linear flow for both up and down curves, which was characteristic of diluted suspensions, fitting to Carreau Yassuda Model[20]. As a result, these suspensions presented very low viscosity (Figure 5b, η<20mPa.s), which is not desired for replica method. However, increasing solids load to 20 and 25vol.% the flow behavior changed from linear to dilatant tendency, where the viscosity (η) increases according to shear stress (τ) applied (Figure 5a). In this case the flow behavior fitted to Oswald de Walle model[20]. For replica method dilatant suspensions are not desired seeing that they show higher viscosity when

subject of shearing as immersion of template into suspension. Despite of dilatant tendency for 20-25vol.% suspensions, the viscosity (η=50.15mPa.s, Figure 5b) was suitable for replica processing, seeing that others agents as binders will be added in suspension in order to change its structure (flocculation). As a result, the flow behavior changes and viscosity increases. Dilatant tendency was reported for yttrium oxide suspensions with 30vol.%. However, as adding 0.5wt.% of binder, the dilatant flow behavior changed to pseudoplastic. The desired suspension for replica has to be fluid enough to enter, fill, coat uniformly the template surface and under static condition, its viscosity has to be high enough to remain on the template surface.
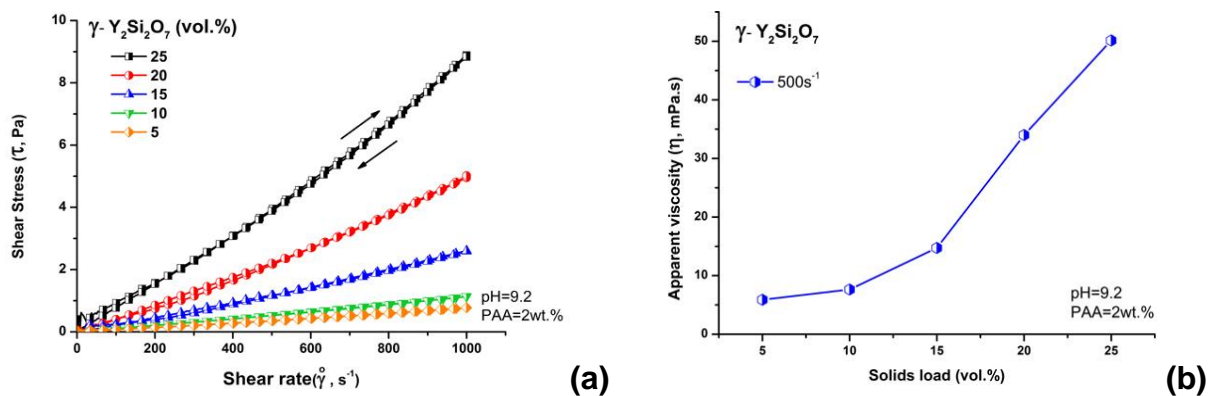

Figure 2. Rheological evaluation of $Y_2Si_2O_7$-γ suspensions with solids load from 5-25vol% and prepared with PAA=2wt.% and pH=9.2. In (A) flow curves and (B) apparent viscosity.

Conclusion

Stability of gamma yttrium disilicate (γ-$Y_2Si_2O_7$) in aqueous medium was evaluated by zeta potential determination and flow curves in controlled rate mode. Nano-particles of γ-$Y_2Si_2O_7$ (d_{50}= 364.36nm) were more stable in the condition of pH$\geq$6.5 and using 2wt.% of dispersant (ammonium polyacrylic, PAA). Suspensions prepared with up to 15vol.%, at pH=9.2 and PAA=2wt.% presented low viscosity (η<20mPa.s) and linear flow, whereas for 20-25vol.% the rheological behavior became more viscous and with dilatant tendency, that was not suitable for replica method. However, complementary rheological analyses and the use of other agents as binders have to be done in order to

complete the evaluation on the flow behavior of concentrated suspensions for replica method.

Acknowledgements

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