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Plasma spheroidization and high temperature stability of lanthanum phosphate and its compatibility with molten uranium

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ABSTRACT

Lanthanum phosphate has excellent thermal stability and corrosion resistance against many molten metals and other chemically corrosive environments. Lanthanum phosphate ($LaPO_4$) was synthesized from lanthanum oxalate by thermal dissociation of the oxalate to the oxide, followed by conversion to hydrated lanthanum phosphate ($LaPO_4{\cdot}0.5H_2O$). Thermal treatment of $LaPO_4{\cdot}0.5H_2O$ above 773 K resulted in the irreversible transformation of the hydrated phase to the stable monazite phase. Thermal and chemical stability of monazite was studied by plasma spheroidization experiments using a DC thermal plasma reactor set up. Compatibility of monazite with molten uranium was studied by thermal analysis. Results showed that monazite is thermally stable up to its melting point and also is resistant towards attack by molten uranium. Adherent coatings of $LaPO_4$ could be deposited onto various substrates by atmospheric plasma spray technique.

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1. Introduction

There has been a growing interest on rare earth phosphates ($REPO_4$) during the past few years, due to the large application potential of these compounds [1–5]. Lanthanum phosphate and other $REPO_4$ find extensive applications in the form of powders, coatings, composites or dense sintered parts. By virtue of its thermal stability, high thermal expansion coefficient and low thermal conductivity, $LaPO_4$ is considered as a potential material for thermal barrier coating applications. It has also good corrosion resistance in environments containing sulphur and vanadium salts and is reported to be non-reactive with many molten metals [2]. In radioactive waste disposal, RE phosphates are considered as potential matrices for the specific conditioning of separated long-lived radionuclides, such as trivalent actinides [5]

Rare earth phosphates have been synthesized by various methods such as wet chemical precipitation and sol–gel route using phosphoric acid or ammonium phosphate, or by high temperature solid-state reactions [6–9]. Thermal behavior, structural changes and high temperature stability of rare earth phosphate powders $REPO_4{\cdot}nH_2O$ (Re = La, Ce or Y) have been investigated by Lucas et al. [10]. According to these authors, the monazite phase is stable up to 1773 K.

Hikichi and Nomura [11] reported the melting temperature of monazite as 2345 ± 20 K using a heliostat-type solar furnace. Other than the work of these authors, there is hardly any literature related to phase stability of monazite up to its melting temperature. There is also hardly any reported work on the chemical interaction of monazite with molten uranium and water.

The main objectives of the present work are to study the high temperature stability of $LaPO_4$ by plasma spheroidization and interaction of $LaPO_4$ with molten uranium and water. The first part of the paper deals with synthesis of $LaPO_4$ powder by chemical route and its characterization and the second part deals with plasma spheroidization and reaction with molten uranium.

2. Experimental methods

2.1. Synthesis of $LaPO_4$

Lanthanum oxalate powder, 99.99% chemically pure (supplied by M/s Indian Rare Earths Ltd.) was used as the starting material to synthesize lanthanum phosphate. The oxalate powder was calcined at 1273 K for 4 h in a box furnace to completely convert it into lanthanum oxide. The completeness of the reaction was checked by thermo-gravimetric and differential thermal analyses (DTA). The oxide powder was then stored in a desiccater. The oxide was converted to phosphate by reaction with 85% orthophosphoric acid in a glass beaker. Sufficient quantity of the acid was used to ensure total conversion of the oxide to phosphate. The reaction mixture was continuously stirred and cooled in a water bath to control the tem-

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Table 1
Typical experimental parameters for plasma spheroidization and melting experiments

Input power	24 kW
Arc voltage	40 V
Arc current	600 A
Plasma gas (Ar)	25 LPM
Plasma gas (N_2)	05 LPM
Particle size	38–75 μm
Powder feed rate	15 g/min
Temperature (calculated)	12000 K

perature resulting from the highly exothermic nature of the reaction. The beaker was allowed to cool and the precipitate was repeatedly washed with demineralized water until free of acid and then dried at 373 K. The resulting powder was used as feedstock for characterization and further experiments.

2.2. Characterization

X-ray powder diffraction technique was used to identify the phases. X-ray diffraction patterns were recorded using STOE diffractometer (theta–theta geometry) with copper K-α radiation using graphite monochromator. Thermogravimetric analysis (TGA) and differential thermal analysis techniques were used to study the thermal stability of the phosphate. The sample was taken in an alumina cup and thermogram was recorded up to 1473 K in flowing air at a heating rate of 10^0 min^{-1}. Sintered α-aluminium oxide powder was used as the reference material for DTA.

2.3. Plasma spheroidization

Plasma spheroidization and melting experiments were used to ascertain whether the monazite phase melted congruently without decomposition. The as-precipitated ultrafine powder of $LaPO_4$ had poor fluidity and hence it was found unsuitable for plasma spheroidization experiments. Free flowing powder suitable for injection into the plasma jet was synthesized by cold compaction and high temperature sintering followed by crushing and sieving [12]. The powder was compacted by cold isostatic pressing at 30,000 PSI. The compacted mass was sintered at 1673 K for 4 h in air. The sintered mass was broken and crushed into smaller chunks, pounded and milled in a planetary ball mill for 30 min. The milled powder was sieved and graded into different size fractions. The powder fraction having particle size in the range of 38–75 μm was used for plasma melting and spheroidization experiments. The thermal plasma reactor designed and developed in our laboratory for plasma-processing studies [13] was adapted for plasma spheroidization experiments. The reactor consists of water-cooled stainless steel cylindrical vessel, 600 mm long, 240 mm diameter. A 40 kW DC plasma torch is mounted on top of the reactor. Free flowing $LaPO_4$ powder, prepared as described above, was stored in a powder feeder and injected into the plasma jet by means of a carrier gas. The powder was injected through a side port on the torch nozzle.

The powder particles are heated up and melt as they traverse the plasma zone and the molten particles issuing out of the plasma torch nozzle are quenched in water kept at the bottom of the reactor. Plasma melting experiment was also carried out by focussing the plasma jet on to a sintered pellet of $LaPO_4$ until surface melting occurred. Typical process parameters for plasma spheroidization and melting experiments are listed in Table 1.

2.4. Interaction with uranium

Chemical reaction of $LaPO_4$ with uranium was evaluated by differential thermal analysis. Uranium disc of about 3 mm diameter and about 2 mm thick was sandwiched between two buttons of $LaPO_4$ and placed in the crucible of the TG/DTA equipment. The sample was heated in helium gas at a heating rate of 10 K min^{-1} and the thermal energy change was continuously recorded with time. The sample was heated to 1473 K so as to ensure that uranium is in the molten state and held isothermally at that temperature for 2 h. It was then cooled to room temperature.

3. Results and discussion

3.1. Characterization and thermal stability

X-ray diffraction pattern of a sample of as-precipitated $LaPO_4$ powder dried at 373 K is shown in Fig. 1(a). The XRD pattern corresponds to a single homogeneous phase, which could be identified as hydrated lanthanum phosphate ($LaPO_4{\cdot}0.5H_2O$) with Rabdophane structure [14]. Results of TGA and DTA are shown in Fig. 2(A and B),

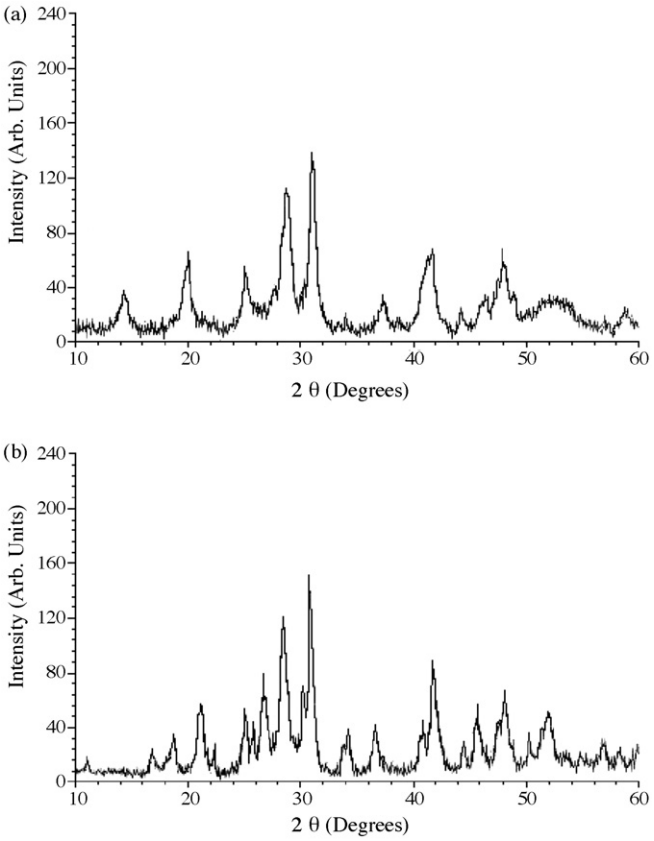

Fig. 1. X-ray diffraction pattern of (a) $LaPO_4{\cdot}0.5H_2O$, (b) residue after TGA/DTA.

respectively. The derivative of the thermogravimetric curve indicating the rate of loss of weight is shown in Fig. 2(C). The TG curve shows continuous loss of weight right from room temperature in a stepwise manner. The low temperature loss of weight, due to escape of residual absorbed water, starts at room temperature and is complete by about 423 K. The corresponding DTA curve shows a broad endothermic peak. A significant weight loss was observed between 473 and 673 K and then the rate of weight loss decreased gradually. The differential thermal analysis curve shows a well-defined

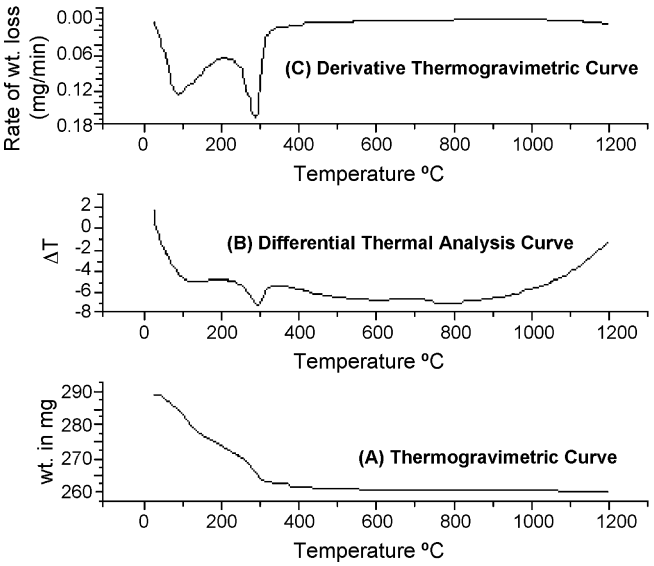

Fig. 2. DTA/TGA of $LaPO_4{\cdot}0.5H_2O$.

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