A combined experimental and theoretical study of enthalpy of phase transition and fusion of yttria above 2000 °C using “drop-n-catch” calorimetry and first-principles calculation

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Abstract

“Drop-n-catch” calorimetry was performed on Y₂O₃ spheroids of 2–3 mm in diameter, prepared by laser melting of powders. Samples were aerodynamically levitated in a splittable nozzle levitator in air or argon flow, laser heated from ~2200 to 3000 °C and dropped into a calorimeter at 25 °C, thus measuring their enthalpy as a function of temperature. The fusion enthalpy of cubic Y₂O₃ was derived from the step in the temperature–enthalpy curve as 119 ± 10 kJ/mol. Calculations performed using density functional theory and molecular dynamics techniques produced the value 127 ± 3 kJ/mol Y₂O₃. This combined methodology enables accurate determination of the enthalpies of fusion and phase transition of refractory oxides, including those containing lanthanides and actinides.

Keywords: aerodynamic levitation, enthalpy, first-principles calculation, thermodynamics, yttria

1. Introduction and Motivation

Design and processing of ultra-high-temperature materials for aerospace and nuclear applications demand detailed understanding of thermodynamic properties of their constituents. Nevertheless there is a lack of experimental thermodynamic data above 2000 °C. Even for refractory binary oxides like La_2O_3 , Y_2O_3 , HfO_2 , and ZrO_2 , high temperature thermochemical data are often absent and must be estimated using a number of assumptions [1]. For the rare earth oxides, fusion enthalpy data are largely missing [1, 2]. This lack of data hampers evaluation of phase diagrams (even at lower temperatures) and evaluation of materials stability and compatibility.

The scarcity of data can be attributed to limited calorimetric capabilities above 1500–1700 °C, to lack of reliable phase transition standards for the calibration of thermal analysis equipment at high temperature, and to intrinsic problems of sample–container interactions under extreme conditions. Though up to the 1980s, a number of custom built “drop calorimeters”, utilizing large samples (tens to hundreds of grams) encased in metallic containers and dropped from a furnace into a calorimeter at room temperature, produced ultra-high-temperature heat content data for oxides such as UO_2 [3], most of these systems are no longer operational.

A recently modified commercial Setaram Setsys differential thermal analyzer (DTA) working up to 2500 °C allows observation of phase transitions but the lack of reliable enthalpy standards above the melting temperature of Al_2O_3 (2054 °C) limits accuracy of measurements of heat effects at higher temperature [1].

Y_2O_3 was considered by IUPAC as a secondary standard for melting temperature for data evaluation [4]. There was a single report on the pre-melting phase transition and fusion enthalpy

measurements for Y_2O_3 by drop calorimetry from a hot furnace into a boiling water calorimeter [5] on which the latest assessment of thermodynamic functions for rare earth oxides at high temperature is based [2].

There was an attempt to use the value of 84 kJ/mol of fusion enthalpy of Y_2O_3 [5] to extend sensitivity calibration to 2439 °C [6], but measurements of enthalpy of fusion on two Y_2O_3 samples in sealed crucibles gave values of 130–150 kJ/mol, using the Al_2O_3 fusion enthalpy for calibration. It was concluded that “... melting of Y_2O_3 cannot be used for sensitivity calibration in DTA until its fusion enthalpy is measured accurately by drop calorimetry or some other independent method” [6]. After initial experiments with ultra-high-temperature thermal analysis [6] (as well as measurements of fusion and phase transition enthalpies in La_2O_3 [7]), a new experimental approach, called “drop-n-catch calorimetry”, was developed in our laboratory for measurements of enthalpies of phase transitions and fusion in rare earth oxides and other refractory compounds [8]. In this method laser heated aerodynamically levitated sample spheres weighing 10–50 mg are dropped into a specially designed room temperature calorimeter.

Our first goal, now achieved [8], was to measure the fusion enthalpy of Al_2O_3 in order to validate system performance and calibration of the calorimeter. In the current work, we apply the “drop-n-catch” technique to determining the heat of fusion of Y_2O_3 ($T_m = 2439 \pm 12$ °C). At the same time, we apply first principles calculations (based on density functional theory and molecular dynamics) to the same transition, obtaining excellent agreement between theory and experiment.

2. Experimental Procedures

2.1. Preparation of samples for calorimetry

Powders of Y_2O_3 (REacton, 99.999 %) were melted into polycrystalline spheroids 2–3 mm in diameter in a copper hearth in air with a 400 W sealed CO_2 laser with 6 mm beam diameter (10.6 μm , Synrad FSi401SB). The copper hearth was placed on the motorized X–Y stage under the beam and the laser power was adjusted by a LabVIEW controlled laser controller (Synrad UC-2000) which allows incrementing the power by 0.5 %. The high surface tension of rare earth oxide melts produces spheroidal shapes for beads of this diameter.

2.2. Cooling profile recording

Cooling profiles on crystallization of Y_2O_3 in air and argon were recorded and analysed to elicit any possible effect of atmosphere on melting and phase transition temperatures. Selected beads of Y_2O_3 were levitated and rotated in an aerodynamic levitator (see [8] for details) in air or argon flow and then heated with the laser. Temperature on laser melting and cooling traces during quenching from the liquid state were measured by a fast single band pyrometer (IR-CAS8CS CHINO, 900 nm, 10 ms response time, 700–3500 $^{\circ}\text{C}$) providing 0.64–1 mm spot size and were recorded with voltmeter and LabVIEW software.

2.3. “Drop-n-catch” calorimetric experiment

The experimental prototype “drop-n-catch” (DnC) system, designed and built in the Peter A. Rock Thermochemistry Laboratory at UC Davis, is shown schematically in Fig. 1. Detailed description of the laser heating system for operation at temperatures up to 4000 $^{\circ}\text{C}$ is in [8]. The DnC calorimeter system consists of the aerodynamic levitator with splittable nozzle and copper catch plates with two semiconductor thermopiles. The thermopiles are positioned

between each catch plate and a copper jacket thermostated at 25 °C by water cooling from a recirculating chiller.

Weighed Y_2O_3 beads of 2–3 mm diameter were aerodynamically levitated in air (or argon) flow in the aluminum nozzle with a 90 ° cone and a 1 mm orifice and subsequently heated with the laser beam to desired temperature (Fig. 2a). Operating, power of the laser was ~0–35 %. The latter power was enough to reach the highest temperature of 2911 ± 7 °C (see Table 1). The levitated solid sample rotates in the nozzle due to surface imperfections and the cavity in the bead formed on quenching from the melt due to volume reduction. The beads continue to rotate by inertia in the molten state for a few seconds. Under such rotation the registered sample temperature is a more accurate representation of the average temperature of the sample.

When the desired temperature was reached, the nozzle was split, and the sample dropped and was caught by the copper catch plates which fully enclose the sample (Fig. 2b). Temperature was controlled by manual laser power adjustment through LabVIEW software. Transition between solid and liquid states was observed visually on display from a color CCD camera (Hitachi KP-D20A). The surface temperature (T_s) was measured before each drop with a multiwavelength spectropyrometer (500–1000 nm, 800–4000 °C, 1 s response time, on-line tolerance; FAR Associates) providing 1.3 mm spot size with the laser aiming illuminator directed at the top of the bead close to where the heating laser beam was incident on the sample. The spectropyrometer was chosen, instead of single band pyrometer, as it provides temperature values not dependent on emissivity. Its response time (though less than that of the high speed pyrometer) is adequate for monitoring sample temperature prior to drop.

Samples were dropped by splitting the nozzle with two solenoids and caught with copper plates operated with another set of solenoids. At the time of the drop, the laser was turned off

through LabVIEW software. The delays for splitting the nozzle and catching the sample were controlled and adjusted with LabVIEW software. The total distance of the drop was ~30 mm and it took 88–97 msec. The thermopile signal returned to its initial baseline level in about 20 min. After the experiment the beads were reweighed on a microbalance. Mass changes were not more than 0.02 mg. The heat effects on cooling to 25 °C were obtained from integral value of the peak in thermopiles voltage vs. time. Integration of heat flow during cooling was made in Origin 8.5 software using linear baseline (see Fig. 4 for two samples caught in liquid (a) and solid (b) states).

2.4. Computational techniques

First-principles density functional theory [9] was employed to model Y_2O_3 . All electronic structures were calculated by the Vienna *Ab-initio* Simulation Package (VASP) [10], with the projector-augmented-wave (PAW) [11] implementation and the generalized gradient approximation (GGA) for exchange-correlation energy, in the form known as Perdew-Burke-Ernzerhof (PBE) [12]. The valence configuration for yttrium was $([\text{Ar}]3d^{10})4s^24p^64d^15s^2$ with cutoff radius of 1.815 Å; for oxygen, the 2s and 2p electrons were relaxed with cutoff radius of 0.820 Å. This required a plane-wave basis set with cutoff energy of 400 eV.

Electronic temperature was accounted for by imposing a Fermi distribution of electrons on the energy level density of states. The electronic temperature was consistent with the ionic temperature. We used automated *k*-meshes generation with a *k*-point density of $(20)^3/\text{\AA}^{-3}$ in the Brillouin zone.

SLUSCHI code [13] was employed to automate calculations. First-principles molecular dynamics (MD) techniques were utilized to simulate atomic movements and trajectories. The

MD simulations were carried out under constant number of atoms, pressure and temperature condition (*NPT*, isothermal-isobaric ensemble). The thermostat was conducted under the Nosé-Hoover chain formalism [14, 15]. The barostat was realized by adjusting volume every 80 steps according to average pressure. Although this did not formally generate an isobaric ensemble, this approach had been shown to provide an effective way to change volume smoothly and to avoid the unphysical large oscillation caused by commonly used barostats [16].

We apply a correction to the PBE fusion enthalpy based on the hybrid Heyd–Scuseria–Ernzerhof (HSE) functional [17], which is generally considered more accurate than PBE. For both the solid and liquid phases, we compute energy differences between PBE and HSE on randomly chosen snapshots from MD trajectories, and then combine them as a correction on the fusion enthalpy.

3. Results and Discussion

3.1. Cooling traces analysis

Cooling traces for a 61.80 mg Y_2O_3 bead levitated in air and argon are shown in Fig. 3. Two temperature rises (recalcescence peaks) were observed due to the latent heat released during the crystallization of the solid phase from the undercooled liquid and the hexagonal-to-cubic phase transformation. It is worth noting that the measured recalcescence temperatures (T_{rls}) cannot be higher than the melting and phase transformation temperatures. Otherwise arrest(s) on heating would be observed. The emissivity values for Y_2O_3 above 2000 °C are not known. The temperature values in Fig. 3 refer to apparent temperatures from the single band pyrometer with emissivity set to 0.9, giving maximum recalcescence temperatures close the known melting

temperature of Y_2O_3 (2439 ± 12 °C) [18]. The differences in recalescence temperatures between experiments in argon and in air are less than instrumental uncertainties and indicate that melting temperature, H-C phase transformation temperature, and emissivity of Y_2O_3 is not strongly affected by the atmosphere, which is consistent with previous estimations of melting temperature.

3.2. Fusion enthalpy of Y_2O_3 from “drop-n-catch” calorimetry

28 successful drops were performed on different Y_2O_3 beads 30–60 mg in weight (see Table 1). The step on the enthalpy curve, 119 ± 10 kJ/mol (Fig. 5), corresponds to fusion enthalpy of cubic Y_2O_3 and is the sum of fusion enthalpy of high temperature hexagonal Y_2O_3 phase and enthalpy of C to H phase transition. Due to the thermal gradient in the levitating bead we cannot separate these heat effects from DnC calorimetry data.

These heat effects were separated on heating in earlier reported Y_2O_3 melting in DTA (see Fig. 3 in [6]). Assuming constant sensitivity of DTA for 2330–2430 °C range, DTA results give the ratio of heat of fusion to the sum of heat of fusion and phase transformation enthalpies as 0.64. Using this value we can separate heat effect from DnC data as $\Delta H_{\text{fus,H}} = 76 \pm 6$ kJ/mol and $\Delta H_{\text{trans C-H}} = 43 \pm 4$ kJ/mol.

In drop calorimetry measurements by Shpilrain et al. [5] samples tens of grams in weight were vacuum sealed in tungsten or molybdenum containers and heated in a resistance furnace with a tungsten heater. The steps corresponding to C-H transition and fusion were distinct, giving values 25 ± 8 kJ/mol at 2327 ± 30 °C and 84 ± 8 kJ/mol at 2430 ± 12 °C, respectively. This corresponds to 109 ± 8 kJ/mol value for fusion enthalpy of cubic Y_2O_3 . Thus the values from conventional drop calorimetry and DnC calorimetry are in agreement within experimental error.

3.3. Fusion enthalpy of Y_2O_3 from calculations

We ran molecular dynamics simulations on the cubic solid and the liquid separately, and then computed fusion enthalpy as enthalpy difference between the two phases. As shown in Table 3, fusion enthalpy was calculated to be 127 ± 3 kJ/mol, which agrees with experimental measurement. Simulations were performed on two different supercells, one smaller with 80 atoms and the other one larger with 240 atoms. This allows us to scrutinize the finite-size effect, which is expected to be large in the Y_2O_3 system due to the long-range Coulomb interaction among charged ions. In our experience, in typical systems, a simulation cell size of about 100 atoms is sufficiently large [13, 16]. Hence we increased system size up to 240 atoms to guarantee convergence in this system.

3.4. Heat capacity of Y_2O_3 near melting temperature

Linear fitting of enthalpy values versus surface temperature before the drop below and above the step gives a slope of 116 ± 43 J/mol·K and 168 ± 42 J/mol·K, respectively. These values do not correspond to true heat capacity and until a reliable procedure for the temperature corrections is established and evaluation of average temperature of the sample caught in the calorimeter can be made accurately, application of DnC is presently limited to the measurements of enthalpies of fusion and phase transitions. Results from DFT and MD calculations of enthalpies for C-phase and liquid at 2500 and 2650 K give heat capacity values as 218 ± 28 J/mol·K and 208 ± 15 J/mol·K, respectively. The high heat capacity for the C-phase in this range might result from onset of the C-H phase transition and its associated disordering. A

similar trend in heat capacity before and after melting observed in UO_2 [19, 20], with the heat capacity of liquid UO_2 decreasing to the Dulong-Petit 9R limit only at 4000 °C.

4. Conclusion

Heat of fusion of C-phase Y_2O_3 was measured by “drop-n-catch” calorimetry as 119 ± 10 kJ/mol is consistent with the value of 127 ± 3 kJ/mol from first principles calculations and with the previously reported value 109 ± 8 kJ/mol from conventional drop calorimetry. Agreement between computational and experimental results on fusion enthalpy render higher confidence in calculated heat capacity values for Y_2O_3 around the melting temperature, not yet confirmed from experiment. This agreement also gives confidence to calculations for other lanthanide and possibly actinide heats of fusion.

The first results open the door to thermochemical measurements previously inaccessible in terms of temperature range, controllable atmosphere, and lack of sample–container interactions. They establish the melting of yttria with as a reference material for ultra-high-temperature thermal analysis. The latter allows the use of thermal analysis complementary to drop calorimetry for further measurements on refractory oxides and other systems of interest, for example $\text{RE}_2\text{O}_3\text{--Al}_2\text{O}_3\text{--HfO}_2\text{--ZrO}_2$ (RE = rare earth) compositions.

Table 1. Drop-and-Catch experiments on laser heated levitated Y_2O_3 in air and argon

#	Mass [mg]	Catch Delay [ms]	T_s^a [°C]	Heat flow [mV·s]	$\Delta H (T_s^a - 25^\circ\text{C})$ [kJ/mol]	Condition
1	30.37	90	2287 ± 4	3847	249.2 ± 4.3	air
2	61.48	91	2382 ± 6	7198	255.5 ± 4.8	air
3	41.85	90	2407 ± 9	5544	260.7 ± 4.5	air
4	39.61	90	2448 ± 6	5169	256.8 ± 4.4	air
5	41.85	90	2467 ± 7	5584	262.5 ± 4.5	air
6	31.55	90	2476 ± 7	4394	274.0 ± 4.7	air
7	31.55	90	2538 ± 6	4508	281.1 ± 4.8	air
8	40.69	88	2589 ± 8	5867	314.6 ± 5.9	air
9	31.55	90	2594 ± 7	5103	318.3 ± 5.3	air
10	31.53	90	2617 ± 7	5822	363.3 ± 6.3	air
11	40.68	88	2629 ± 10	7170	384.6 ± 7.2	air
12	46.28	92	2649 ± 9	8319	392.3 ± 7.3	air
13	40.68	88	2677 ± 10	7959	426.9 ± 8.0	air
14	35.70	90	2698 ± 8	7182	395.8 ± 6.8	air
15	35.70	90	2741 ± 8	7564	416.9 ± 7.2	air
16	39.24	90	2778 ± 8	8547	428.6 ± 7.4	air
17	35.12	88	2797 ± 7	7164	445.1 ± 8.3	air
18	32.92	88	2824 ± 7	6444	427.1 ± 8.0	air
19	30.38	90	2853 ± 8	6907	447.3 ± 7.7	air
20	36.03	88	2869 ± 7	7052	427.1 ± 8.0	air
21	31.25	88	2871 ± 8	6406	447.3 ± 8.3	air
22	30.41	90	2887 ± 7	6712	434.3 ± 7.5	air
23	45.16	88	2911 ± 7	9412	454.8 ± 8.5	air
24	39.79	97	2304 ± 18	4838	239.2 ± 4.1	Ar
25	35.86	95	2371 ± 10	4889	268.3 ± 4.6	Ar
27	39.79	97	2440 ± 7	4934	244.0 ± 4.2	Ar
26	32.61	95	2713 ± 7	6794	409.9 ± 7.1	Ar
28	32.60	95	2757 ± 8	7038	424.8 ± 7.3	Ar

^a Surface temperature measured by FAR spectropyrrometer

Table 2. Lattice constant and density of Y_2O_3

Structure	Lattice constant [$\text{\AA}$]	Experiment [$\text{\AA}$]	Density [g/cm^3]
cubic, 80 atoms, 2500 K	11.012	10.82 [1]	4.491
cubic, 240 atoms, 2500 K	11.019		4.483
cubic, 240 atoms, 2650 K	11.054	10.83 [1]	4.440
liquid, 80 atoms, 2500 K			4.300
liquid, 240 atoms, 2500 K			4.246
liquid, 240 atoms, 2650 K			4.150

Table 3. Enthalpy of Y_2O_3

Structure	Enthalpy	Heat of fusion		w/ HSE correction
	[eV/atom]	[eV/atom]	[kJ/mol]	[kJ/mol]
cubic, 80 atoms, 2500 K	-8.422 ± 0.007	0.200 ± 0.010	97 ± 5	109 ± 5
liquid, 80 atoms, 2500 K	-8.221 ± 0.008			
cubic, 240 atoms, 2500 K	-8.403 ± 0.004	0.237 ± 0.005	115 ± 3	127 ± 3
liquid, 240 atoms, 2500 K	-8.165 ± 0.004			
cubic, 240 atoms, 2650 K	-8.335 ± 0.008	0.234 ± 0.009	113 ± 5	125 ± 5
liquid, 240 atoms, 2650 K	-8.100 ± 0.003			

Table 4: HSE correction on Y₂O₃ fusion enthalpy

Structure		H (HSE-PBE) [eV/atom]	H (HSE-PBE) [eV/atom]	ΔH (HSE-PBE) [eV/atom]	ΔH (HSE-PBE) [kJ/mol]
solid	#1	-2.616	-2.610 ± 0.002	0.024 ± 0.003	12 ± 2
	#2	-2.606			
	#3	-2.607			
	#4	-2.612			
liquid	#1	-2.589	-2.586 ± 0.002		
	#2	-2.581			
	#3	-2.589			
	#4	-2.585			

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Figure Captions

Fig. 1. Schematic “drop-n-catch” system.

Fig. 2. Photographs of “drop-n-catch” operation before (a) and after (b) drop.

Fig. 3. Cooling traces of Y_2O_3 in (a) air and (b) argon, measured by high speed (10 ms response time) Chino pyrometer. L, H and C – liquid, H- and C-phases, respectively.

Fig. 4. Heat flow curves recorded during the “drop-n-catch” experiments of Y_2O_3 caught in (a) liquid, and (b) solid states.

Fig. 5. Enthalpy change measured in “drop-n-catch” experiments on laser heated levitated Y_2O_3 in air and argon. T_s – surface temperature measured by FAR spectropyrometer.

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