

## A phenomenological description of the rate of the aluminum/oxygen reaction in the reaction-bonding of alumina

Jill M. Aaron<sup>a</sup>, Helen M. Chan<sup>b</sup>, Martin P. Harmer<sup>b</sup>, Maytee Abpamano<sup>a</sup>, Hugo S. Caram<sup>a,\*</sup>

<sup>a</sup> Department of Chemical Engineering, Lehigh University, Bethlehem, PA 18015, USA

<sup>b</sup> Department of Materials Science and Engineering, Lehigh University, Bethlehem, PA 18015, USA

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### Abstract

The isothermal reaction kinetics of aluminum oxidation in loose Al/Al<sub>2</sub>O<sub>3</sub> powder was studied in the temperature range 455–555 °C using thermogravimetry. The powder compositions ranged from 30 to 65 vol.% aluminium, with average Al particle sizes in the range 1.4–2.7 μm. The oxygen mole fraction was varied between 20 and 100%. It was found that only 70–80% of the aluminum can be oxidized below the melting point of 660 °C. The weight gain curves were well fitted by assuming that the rate of oxidation was first order with respect to the amount of aluminum, which can be oxidized below 660 °C, left in the sample. The reaction rate was observed to be independent of both the partial pressure of oxygen and the aluminum particle size. It was shown that the same kinetics relationship could also be applied to a broad range of experimental oxidation data in the literature, yielding a common value of the Arrhenius constant and activation energy. The kinetic model was successfully applied to the controlled processing of RBAO powder compacts for constant temperature hold and constant reaction rate cases.

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### 1. Introduction

The reaction-bonded aluminum oxide (RBAO) process was developed by the Advanced Ceramics Group of the Technical University of Hamburg-Harburg in Hamburg, Germany.<sup>1</sup> The RBAO process involves heat treating attrition-milled Al/Al<sub>2</sub>O<sub>3</sub> or Al/Al<sub>2</sub>O<sub>3</sub>/ZrO<sub>2</sub> powder compacts in an oxidizing atmosphere to produce Al<sub>2</sub>O<sub>3</sub>-based ceramics.<sup>1–10</sup> RBAO precursor powders are made using 30–80 vol.% aluminum powder, 20–70 vol.% alumina powder, and 0–20 vol.% zirconia powder. The addition of zirconia improves the microstructure and mechanical properties of the final ceramic.<sup>2,3</sup>

The oxidation of aluminum occurring during the RBAO process is given by the reaction



This reaction is extremely exothermic and results in volume expansion and weight gain. Due to the weight gain, the reaction rate can be determined using thermogravimetry (TG).

Reaction behavior of a typical RBAO powder heated in air is shown in Fig. 1. The furnace temperature was increased at 1 °C/min. The weight gain is plotted against the furnace temperature, which may be different from the actual sample temperature. After the initial weight loss, associated with the evaporation of fugitive species, there is a rapid acceleration of the rate of reaction at a furnace temperature of about 450 °C. The reaction rate reaches a maximum in the furnace temperature range of 500–550 °C. Under these conditions, the sample ignites and there are temperature excursions of up to 1000 °C. As the furnace temperature increases above 550 °C

\* Corresponding author. Tel.: +1 610 758 4259; fax: +1 610 758 5057.  
E-mail address: [hsc0@lehigh.edu](mailto:hsc0@lehigh.edu) (H.S. Caram).

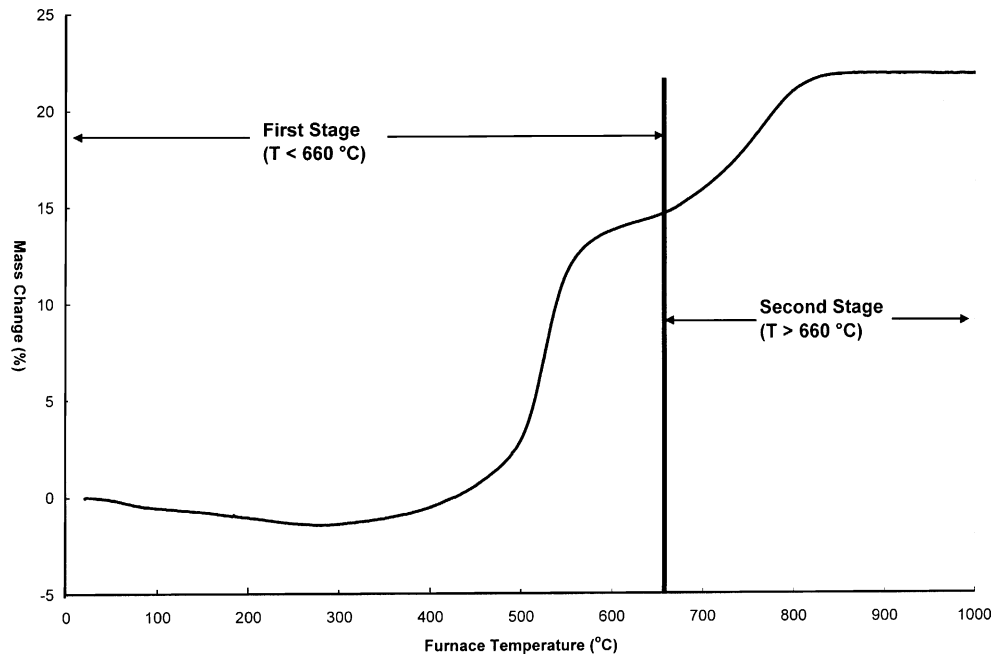


Fig. 1. Reaction behavior of a typical, RBAO powder heated 1 °C/min in air.

the reaction slows down to the point that no further conversion will occur if the furnace temperature does not exceed the melting point of aluminum at 660 °C, when total conversion will occur. It is the uneven volume gains and sharp thermal gradients associated with the ignition phenomena that cause stresses which result in sample cracking.<sup>2–4,6,7</sup> Cracking can be avoided by using low heating rates and isothermal temperature holds that avoid reaction runaway.<sup>2–4,6,7</sup> Knowledge of the reaction kinetics of the oxidation of aluminum can be used to avoid uncontrolled reactions.

The purpose of the present study is to determine the oxidation kinetics of RBAO powders comprising a mixture of Al and Al<sub>2</sub>O<sub>3</sub>. The oxidation of the Al was studied using thermogravimetry, and kinetic constants were determined as a function of aluminum concentration, oxygen partial pressure, and aluminum particle size. Note that in previous modeling studies, the reaction rate had been assumed to be second order with respect to aluminum, and first order with respect to oxygen, but this had not been confirmed experimentally.<sup>3,6</sup>

As will be seen, the data from the present study yields a kinetic model which, in addition to being directly pertinent to RBAO conversion, has general applicability to studies of Al oxidation in the literature.

## 2. Background

### 2.1. Oxidation of aluminum

The oxidation of aluminum has been the subject of many studies.<sup>11–21</sup> Most of them have used thin, flat samples of

aluminum, but some work has also been reported in powder samples. The rate of oxidation of aluminum has been observed to be extremely temperature sensitive, and various kinetic models with different mechanisms have been used to describe the oxidation at various temperatures.

Cabrera and Mott<sup>22</sup> developed theories for the mechanisms which control the oxidation of metals for thick, thin, and very thin oxide layers. For thick oxide films and sufficiently high temperatures, either the metal or oxygen ions are soluble in the oxide and diffuse through the oxide layer. The driving force is a concentration gradient that is inversely proportional to the film thickness leading to the classic parabolic rate law of the form

$$X^2 = 2At \quad (2)$$

where  $t$  is the time;  $X$ , proportional to the amount of aluminum converted and  $A$ , a constant proportional to the species diffusivity across the oxide. Since oxygen ions have poor solubility in aluminum oxide, the rate of diffusion of aluminum ions is commonly assumed to control the reaction rate. Thus, the rate of oxidation can be considered to be relatively independent of the oxygen pressure.

Isothermal weight gain experiments below 450 °C resulted in weight gain curves that fit a parabolic rate law.<sup>5,11–13,22</sup> As discussed above this corresponds to the growth of an amorphous oxide layer which is controlled by aluminum ion diffusion through the layer.<sup>14,22</sup> This is also valid for the initial weight gain region above 450 °C, but as will be discussed below the fit does not extend to higher conversions.

An Arrhenius plot of the rate coefficients from data fitting parabolic kinetics below 450 °C and in the initial region

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