Thermochimica Acta, 140 (1989) 219–223
Elsevier Science Publishers B.V., Amsterdam – Printed in The Netherlands

KINETICS OF SINTERING OF IRREGULAR PARTICLES OF GLASS. PART I. ISOTHERMAL SINTERING

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(Received 8 April 1988)

ABSTRACT

Earlier research work published by this group described the kinetics of isothermal and non-isothermal sintering of monodisperse glass particles. The present article discusses isothermal sintering of irregularly shaped particles of commercial window glass powder, while Part II describes non-isothermal kinetic data. It is shown that the kinetic behaviour of irregular particles is similar to that of regular monodisperse particles. The activation energy values for densification are also almost identical.

INTRODUCTION

A previous communication from this laboratory discussed the kinetics of isothermal and non-isothermal sintering of monodisperse particles of glass powder (particle size $\simeq 100\ \mu m$) of chemical composition Na_2O 31.2, Al_2O_3 2.64, CaO 8.12, MgO 3.98 K_2O 1.5 and SiO_2 52.6 wt.%. The particles in this powder were all spherical and uniform in size. Therefore, all the uncertainties in the analysis of sintering data that normally arise out of irregularities in the size and shape of particles were automatically eliminated. Kinetic parameters (E, k, n) of sintering were determined using the integral and differential approach [1]. The activation energy for viscous flow was also calculated, using the Arrhenius equation. It was observed that the non-isothermal E value evaluated from observed dilatometric shrinkage data had a value much larger than the isothermal value, i.e. 150 kcal mol^{-1} as compared with 40 kcal mol^{-1}.

In the present work, compacts made from irregularly shaped particles of a commercial window glass were used for dilatometric studies to examine whether the same set of kinetic equations are applicable.

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0040-6031/89/$03.50

EXPERIMENTAL

The glass powder used in the present study was of chemical composition SiO_2 72.2, Al_2O_3 1.0, MgO 3.6, CaO 7.1, Na_2O 14.8, K_2O 0.2 wt.%. The sintering experiments used glass powder of size fraction $-200 + 240$ mesh (average particle size of 37 μm). Cylindrical compacts of the powder were prepared using 1% polyvinyl alcohol as a binder to give an adequate green strength. The dilatometric measurements were carried out using a Stanton Redcroft thermomechanical analyser (TMA 785). Each ampoule was of length 14 mm and diameter 10 mm. The load applied was 2 g in all experiments.

Isothermal kinetic data were obtained at temperatures of 590, 630 and 650 °C, using a heating rate of 30 °C min^{-1}. The TMA plots showed that there was some initial expansion in the heating period, due to vaporization of the binder used for compaction. Plots taken from beyond this initial expansion were used to calculate values of fractional shrinkage α (in this case equal to $\Delta L/L_0$) at various intervals of time t.

RESULTS AND DISCUSSION

Figure 1 shows isothermal kinetic plots of fractional shrinkage ($\Delta L/L_0$) vs. time at different temperatures. These data show that densification is

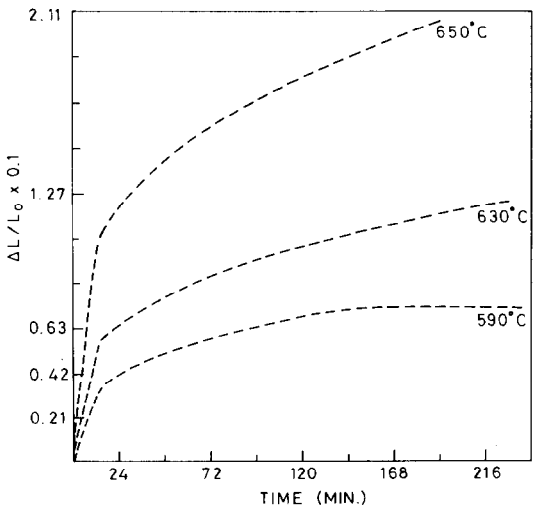

Fig. 1. Isothermal kinetic plots of fractional shrinkage ($\Delta L/L_0$) vs. soaking time.

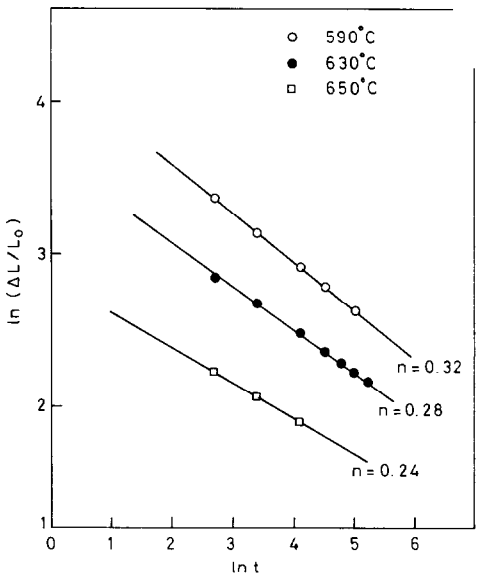

Fig. 2. Plots of $\ln(\Delta L/L_0)$ vs. $\ln t$ for three different temperatures.

more rapid than it is in the monodisperse system [1]. The kinetic plots can be analysed using the well known equation

$$\frac{\Delta L}{L_0} = kt^n \tag{1}$$

or

$$\ln(\Delta L/L_0) = \ln k + n \ln t \tag{2}$$

where k is a rate constant and the exponent n is a constant.

Figure 2 shows plots of $\ln(\Delta L/L_0)$ vs. $\ln t$ for three different temperatures. Values of n and k can be calculated from these plots. Table 1 summarizes the kinetic parameters obtained in this way.

As expected, the values of k are higher at higher temperatures. The values of n are in the range 0.24 to 0.31. This slight variation in the values of n may be due to the overlapping of different sintering mechanisms. It is important to note here that for the system reported earlier, the value of n

TABLE 1

Kinetic parameters calculated from isothermal data

Temperature (°C)	n	k
590	0.31	1.51×10^{-2}
630	0.27	2.71×10^{-2}
650	0.24	5.61×10^{-2}

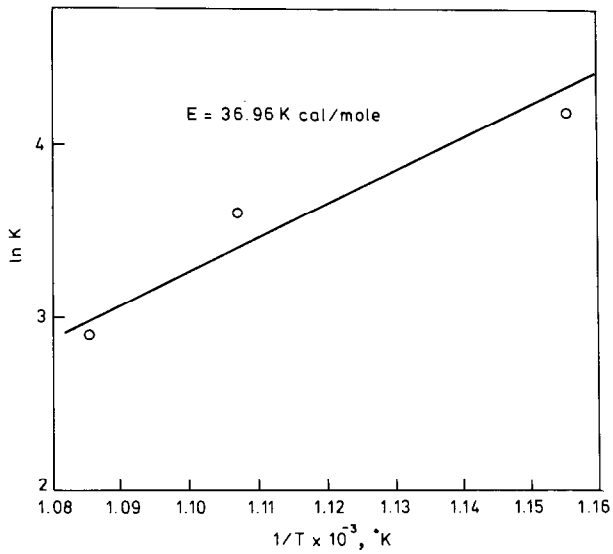

Fig. 3. Arrhenius plot.

was found to be 0.8 [1]. This difference may be attributable to differences in the chemical composition of the glass.

Figure 3 shows the Arrhenius plot of ln k vs. $1/T$, which gives a straight line. The activation energy calculated from the slope was found to be 37 kcal

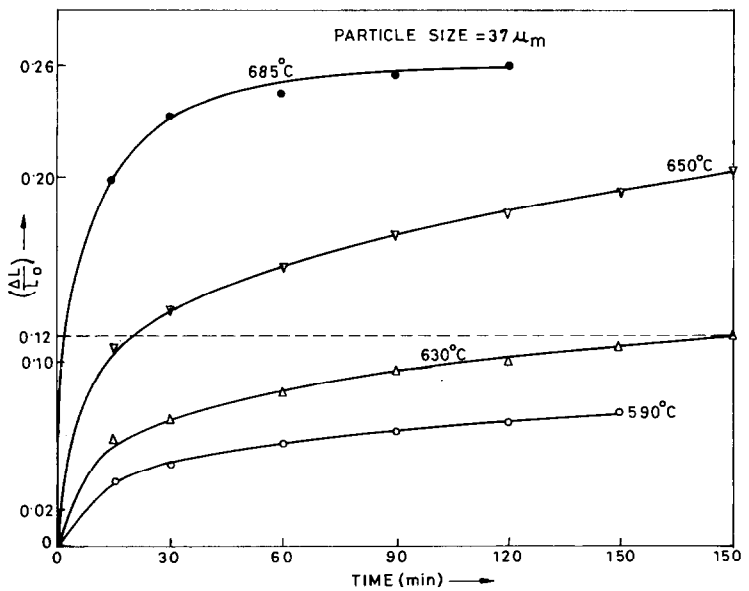

Fig. 4. Differential approach.

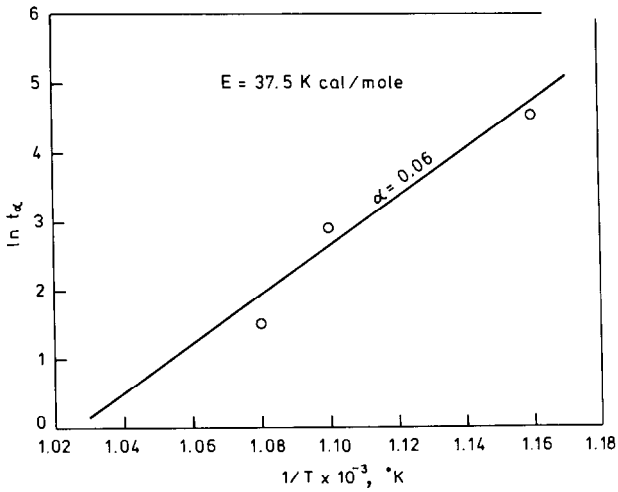

Fig. 5. ln t_α vs. $1/T$ plot.

mol^{-1}. This value compares excellently with the values reported earlier for monodisperse glass particles, i.e. 40 kcal mol^{-1} [1].

The activation energy of isothermal sintering was also calculated using a differential approach. It can be shown that for a fixed kinetic law

$$\ln t_\alpha = \text{constant} + E/RT \tag{3}$$

where t_α is the time required to achieve a given value of α. For $\Delta L/L_0 = 0.06$, values of t_α were calculated for the three temperatures 590, 630 and 650 °C (Fig. 4). Linear variation in ln t_α with $1/T$ is shown in Fig. 5. The slope of this straight line gives an activation energy value of 37.5 kcal mol^{-1}.

CONCLUSIONS

It has been shown that sintering of irregularly shaped particles of a commercial window glass follows a kinetic equation similar to that followed in a monodisperse particle system.

The activation energy of isothermal sintering of irregular particles, as calculated by the Arrhenius equation and as determined by using a differential approach, was found to be around 38 kcal mol^{-1}.

REFERENCE

1 A.N. Virkar, H.S. Ray and A. Paul, Collected papers on XIV Int. Cong. on Glass, New Delhi, India, 1986, Vol. III, pp. 161 (a special publication of the Indian Ceramic Society).