

The Effect of Pressed Density on Fired Density of Alumina Compacts

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알루미나의 성형밀도가 소성밀도에 미치는 영향

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초 록

본 보문은 알루미나의 초기 성형밀도가 소결후 소결밀도에 미치는 영향에 대한 것이다. 실험에서는 두가지의 Reactive alumina powder (ALCOA A-16, A-17)를 사용하였으며, 넓은 범위의 성형밀도를 얻기 위하여 낮은 성형압을 가할 수 있는 기계적으로 작동하는 press 와, 높은 성형압을 가할수 있는 유압식 press를 사용하여, 이론밀도의 35%에서 65% 범위의 시편들 (A-16 alumina powder)과 46%에서 66% 범위의 시편들 (A-17 alumina powder)을 준비하였다. 성형밀도를 Log (성형압)에 대해 plot 해 A-16과 A-17 alumina powder 들에 대한 Compaction mechanism 이 추론되었다. 1350, 1450, 1550°C 에서 소결하여 밀도증가량 ($D_F - D_P$)을 상대 성형밀도 (D_P/D_T)에 대해 plot 해 Ford 가 제안한 두 변수 사이의 포물선 관계가 잘 성립 함을 보였다.

SUMMARY

The main objective of this work was to study the effect of pressed density of pellets prepared from an oxide powder on their fired density and to prove experimentally the parabolic relationship proposed by FORD.

A-16 and A-17 alumina powders, which showed quite different powder characteristics but were essentially very reactive powders, were employed, and a series of pellets were pressed in a 11.2mm diameter hardened steel die with a hydraulic press and a mechanically operated press in order to obtain a wide range of pressed density.

Density ranges from 35 to 65% theoretical for A-16, and 46 to 66% theoretical for A-17 were obtained and the highest pressed density achieved using lubricant was 67.8% theoretical.

Compaction mechanisms for A-16 and A-17 were deduced.

Several plots of ($D_F - D_P$) against D_P/D_T showed reasonable parabola and temperature dependence and the maximum density increase occurred when the pressed density was approximately 50% theoretical. The parabolic relationship was confirmed by linear plot of ($D_F - D_P$) against $(1 - \frac{D_P}{D_T})$.

1. Introduction

The effect of pressed density on fired density was in-

vestigated by some workers, but did not attract the scientists' attention because this was generally considered as a technological subject.

However, it is obvious that pressed density affects fired density considerably during sintering process and, furthermore, pressed density and fired density can be variable from batch to batch.

Therefore it is necessary to establish a densification parameter or a relationship between pressed density and fired density which is constant for a ceramic material and this work set out to make an investigation and an experimental proof of the parabolic relationship proposed by Ford.

II. Literature Survey

In 1949 Jordan and Duwez¹⁾ studied the densification of copper powder compacts in hydrogen and in vacuum and proposed a densification parameter by comparing the observed change in density with the maximum possible change, the value of which is initially zero and which becomes unity if the compact attains its maximum possible density.

The densification parameter, K , proposed was

$$K = \frac{D_F - D_P}{D_T - D_P} \times 100 \quad (II-1)$$

where D_P : initial density before sintering

D_F : density after sintering

D_T : maximum possible density, i. e., the theoretical density.

Lenel²⁾ adopted the densification parameter, K , proposed by Jordan and Duwez¹⁾ and plotted the densification parameter, K , against Log sintering time and from this treatment he showed the rate of densification and the relative degree of densification of compacts having different green densities.

Studying the effect of additives on the sintering of magnesium oxide, Layden and McQuarrie³⁾ mentioned the necessity for the establishment of a densification parameter. They proposed the following equation for a densification parameter on the hypothesis that, for any given firing treatment, the ratio of the actual shrinkage to the total shrinkage possible if the compact reached the theoretical density of the material is constant.

$$K = \frac{\left[\frac{1}{\sqrt[3]{D_P}} - \frac{1}{\sqrt[3]{D_T}} \right]}{\left[\frac{1}{\sqrt[3]{D_P}} - \frac{1}{\sqrt[3]{D_F}} \right]} \quad (II-2)$$

However, their experimental evidence for that equation was confined to a few points.

At the same time Viola and McQuarrie⁴⁾ discovered that, below a certain initial density, alumina powder would not sinter and postulated a constant D_0 , which would represent the minimum density below which particle-particle contact would be insufficient to support the structure, and proposed the following equation as a densification parameter:

$$K = \frac{\frac{1}{\sqrt[3]{D_F - D_0}} - \frac{1}{\sqrt[3]{D_T - D_0}}}{\frac{1}{\sqrt[3]{D_P - D_0}} - \frac{1}{\sqrt[3]{D_T - D_0}}} \quad (II-3)$$

In 1962, Coleman⁵⁾ pressed 8 pellets from magnesia powder calcined from A. R. magnesium carbonate at 1,000°C and sintered them at 1,500°C for 10 mins and 300 mins and then plotted D_F/D_T against D_P/D_T . He showed that the curves did not pass through the origin, which agreed with Viola and McQuarrie's report⁴⁾.

He also transformed the equation (II-1) in terms of porosity as follows:

$$\begin{aligned} \frac{D_F - D_P}{D_P - D_T} &= \frac{\left(1 - \frac{D_P}{D_T}\right) - \left(1 - \frac{D_F}{D_T}\right)}{\left(1 - \frac{D_P}{D_T}\right)} \\ &= \frac{P_F - P_P}{P_F} \end{aligned} \quad (II-4)$$

where P_P = porosity before sintering

P_F = porosity after sintering

This porosity term can be used as a densification parameter which is equivalent to the percentage reduction on sintering.

He arranged equation (II-1) to obtain a linear relation,

$$D_F = (1 - K) \cdot D_P + K \cdot D_T \quad (II-5)$$

and plotted D_F against D_P .

Although the plots were not exactly straight lines, the densification parameter increased approximately linearly with increasing initial density over the experimental range.

Then, he employed a different treatment for his experimental data and, instead of plotting D_F vs. D_P or D_F/D_T vs. D_P/D_T , he plotted the density increase,

$D_F - D_P$, against the relative pressed density, D_P/D_T , and showed a more meaningful relationship. It can be clearly seen that no density increase on firing will occur when the initial density is either zero or theoretical and the curve should exhibit a maximum where pressed density is approximately half of the theoretical density. However, the maximum of the curve occurred at a slightly lower D_P value than the exact half of the theoretical density and he concluded that the asymmetry can not be expressed in terms of any simple equation.

He also suggested that the fractional shrinkage can be used as a densification parameter. He calculated $\Delta V/V_0$ values, where ΔV and V_0 are changes in volume and initial volume of pellets respectively, from the following equation:

$$\frac{\Delta V}{V_0} = 1 - \frac{D_P}{D_T} \quad (II-6)$$

and found that $\Delta V/V_0$ increases approximately linearly with the logarithm of sintering time, and that zero shrinkage will occur when $D_P = D_T$ or $D_P = 0$.

Recently (1976), Ford⁶⁾ re-examined Coleman's work and found that $(D_F - D_P)$ vs. D_P/D_T curve could be a parabola, the equation for which would have to be transformed from the form $Y = aX + bX^2$ to $Y = aX \cdot (1-X)$ on the X-Y axis, since Y is zero when X is either zero or unity.

He replaced X by D_P/D_T and Y by $(D_F - D_P)$ and obtained the following equation:

$$D_F - D_P = a \cdot \frac{D_P}{D_T} \cdot \left(1 - \frac{D_P}{D_T}\right) \quad (II-7)$$

Dividing through by D_P and rearranging, he derived the following equation:

$$\frac{D_F}{D_P} = 1 + \left(1 - \frac{D_T}{D_P}\right) \quad (II-8)$$

Applying this equation to Coleman's data for 300 mins sintering, he obtained a very significant correlation:

$$\frac{D_F}{D_P} = 0.89 + 1.69 \left(1 - \frac{D_P}{D_T}\right) \quad (II-9)$$

which suggested a close approach to the parabolic relationship since 0.89 is not far from unity.

He simplified the equation (II-9) to:

$$\frac{D_F}{D_P} = 1 + b(1-r)$$

where $b = a/D_T$, $r = D_P/D_T$

Thus, the fractional shrinkage of a pellet was calculable from the equation (II-6), which is given by:

$$\frac{\Delta V}{V_0} = \frac{b(1-r)}{1+b(1-r)}$$

Therefore a parabolic relationship requires that the shrinkage falls continuously and curvilinearly to zero as the relative pressed density rises to unity.

III. Experimental Studies

The powders used in all experiments were ALCOA A-16 and A-17 reactive aluminas which have quite different powder characteristics.

The particle size distribution measured with X-ray Sedigraphy 5,000 is shown in Fig. 1, where the mean particle size of A-16 is approx. $0.5\mu\text{m}$.

A-17 powder was originally designed for slip casting purposes, but can be dry-pressed because of its good particle packing property due to its wide range of particle size distribution. The mean particle size of A-17 is approx. $3.5\mu\text{m}$. The chemical analysis quoted from ALCOA product data is shown in Table 1.

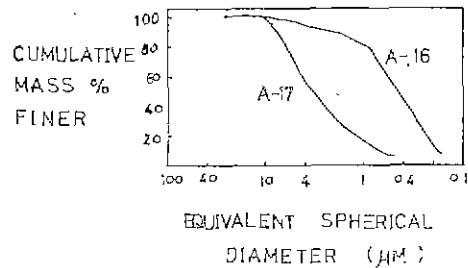


Fig. 1 Particle-Size Distribution of A-16 and A-17 powder measured with X-ray Sedigraphy 5000

TABLE 1 Chemical Analysis of A-16 and A-17 reactive aluminas

	A-16 S.G.	A-17
% Al_2O_3	99.5+	99.5+
% Na_2O	0.06-0.10	0.06-0.10
% SiO_2	0.05-0.12	0.01-0.03
% Fe_2O_3	0.01-0.02	0.01-0.02

Pellets were prepared by two pressing machines.

Firstly, by a mechanically operated press for relatively low pressed density as low load was available by that press, especially below 1,000lbs and secondly, by a hydraulic press for relatively high pressed density.

Pellets were pressed in a 11.2 mm diameter hardened steel die and constant charges of 1.0 and 1.6 gr. of A-16 and A-17 powder were used throughout the study from the determination of optimum weight to give maximum density.

The bulk density of the pellets before and after firing was determined by means of a mercury balance of the type described by Clark and White⁸⁹.

All the sintering was carried out in a carbolite furnace of internal dimensions of 13cm×17.5cm×5cm with ten silicon carbide heating elements in the upper surface (0.5% error in temperature).

Three different temperatures (1,350°C, 1,450°C, and 1,550°C) were employed in order to study the temperature dependence of the parabolic relationship and the soaking time was 3hrs in all cases.

The theoretical density of alumina was assumed to be 3.99gr/cm³.

IV. Results and Discussion

IV-1. Preparation of pellets by die pressing

The density range obtained for A-16 and A-17 by means of both types of presses were from 34.9% to 60.3% and from 46.4% to 65.9% theoretical respectively.

Pellets prepared from A-16 showed a reasonable range of pressed density (about 25% including 50% theoretical), while those from A-17 showed a relatively narrow range largely due to its wide range of particle size distribution, i. e., good particle packing property.

The pressed densities of the various pellets varied with their weights, in other words, with the aspect ratio (the ratio of height to diameter of the pellets) due to the die wall friction, and possibly the dead zone formed near both punches. Therefore there exists an optimum weight which gives the maximum density.

Thus, the optimum weights of A-16 and A-17 were determined as 1.0 and 1.6 gr. respectively by pressing a series of pellets.

This difference can be explained on the basis that A-16 can be trapped in the roughness of the inside die

wall more easily than A-17 because of its smaller mean particle size, thus decreasing the optimum weight needed to produce the maximum density.

In order to investigate the compaction process of A-16 and A-17, the pressed density, D_p , was plotted against log pressure (Fig. 2).

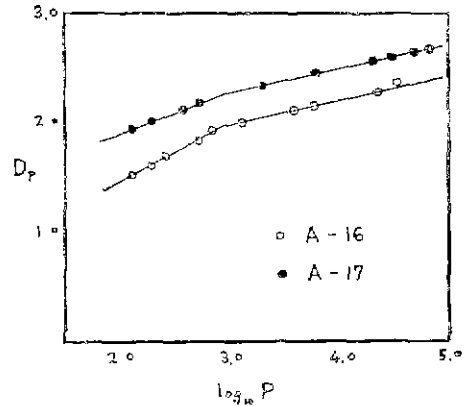


Fig. 2 The variation of Pressed Density with the logarithm of Applied Pressure.

From Fig. 2, it can be said that A-17 can be compacted to higher densities than A-16 at the same applied pressure, largely due to its wide range of particle size distribution. A break was found on each curve and this can be explained on the basis of the fact that particle sliding is predominant up to the breaking point and is followed by the predominance of fragmentation. From the graph, the fragmentation started at approximately 900 and 1,000psi for A-16 and A-17 respectively.

IV-2. Parabolic Relationship

The measured fired densities increased to values between 2.20gr/cm³ (55.1% theoretical) and 3.87 gr/cm³ (97.0%) for A-16 and between 2.15gr/cm³ (53.9%) to 3.16 gr/cm³ (79.2%) for A-17.

The density increase, $D_f - D_p$, were calculated and the results were plotted as $(D_f - D_p)$ against D_p/D_T . (Fig. 3 for A-16 and Fig. 4 for A-17)

In order to verify this relationship, D_f/D_p and $(1 - D_f/D_T)$ values were calculated and plotted to obtain straight lines of slope a/D_T and intercept of 1. (Fig. 5 for A-16 and Fig. 6 for A-17)

The intercept values calculated by the least squares method exhibit approximately 1, which is a very positive result.

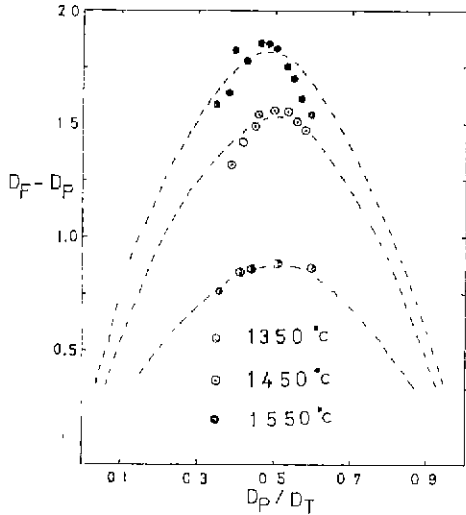


Fig. 3 The variation of the density increase ($D_F - D_P$) with the pressed relative density (D_P / D_T) for A-16

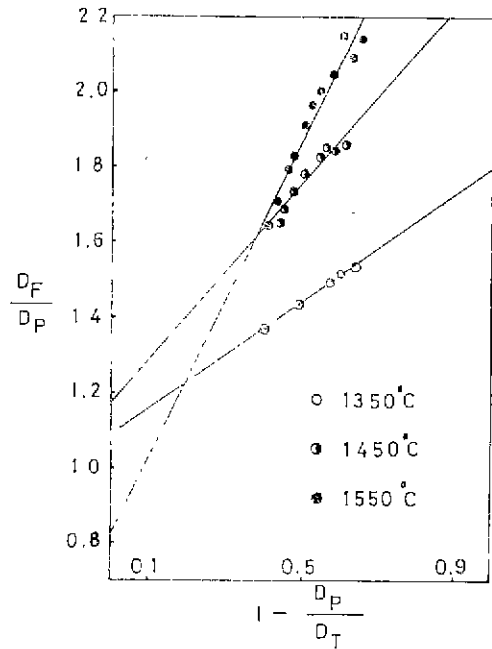


Fig. 5 The linear relationship between D_F / D_P and $1 - D_P / D_T$ values for A-16.

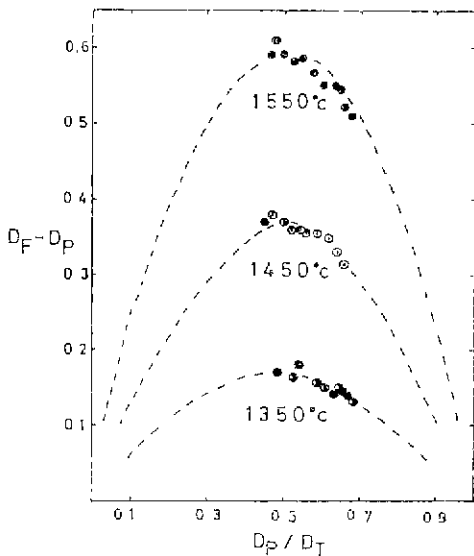


Fig. 4 The variation of the density increase ($D_F - D_P$) with the pressed relative density (D_P / D_T) for A-17

In order to decide accurate 'a' values, equation(II-8) was modified into the following equation to make the lines pass through the origin.

$$\frac{D_F}{D_P} - 1 = m \left(1 - \frac{D_P}{D_T} \right)$$

where $m = a / D_T$

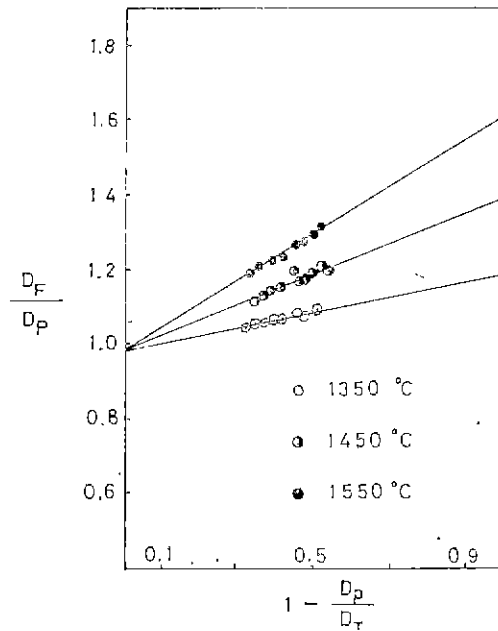


Fig. 6 The linear relationship between D_F / D_P and $1 - D_P / D_T$ values for A-17.

Therefore (D_F/D_P) and $(1-D_F/D_T)$ values were calculated from experimental data and thus 'm' values were also calculated. Then 'a' values were calculated from the average 'm' values for each temperature and the parabolic equations were set. (Table 2)

Table 2 Resulting parabolic equations

Sintering Temperature	A-16	A-17
1350°C	$Y=3.459X(1-X)$	$Y=0.642X(1-X)$
1450°C	$Y=6.023X(1-X)$	$Y=1.500X(1-X)$
1550°C	$Y=7.066X(1-X)$	$Y=2.345X(1-X)$

V. Conclusion

1. The plot of (D_F-D_P) against (D_F/D_T) exhibits a reasonable parabola.
2. The plots of (D_F/D_P) against $(1-D_F/D_T)$ give straight lines of intercept of approx. 1.
3. The different heights of the parabola show temperature dependence.

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