

## Two dimensional model for intergranular void elimination by hot isostatic pressure

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**Abstract.** A two dimensional model for void sintering under hot isostatic pressure controlled by grain boundary self diffusion is presented. Equations for mass transport and traction distributions in the grain boundary are developed. A differential equation for void elimination is established, and numerically solved, where the driving forces are the isostatic pressure and the surface free energy. The qualitative differences of the present approach for densification with previous models based on "neck growth" calculations, by Kuczynski and others, are discussed. Results are curves for material densification, void size and void elimination rate as a function of time. Also the dependence of sintering time with isostatic pressure and with capillarity angle are shown

**Resumen.** Se presenta un modelo bidimensional para la sinterización de cavidades bajo compresión isostática a alta temperatura controlada por autodifusión en fronteras de grano. Se desarrollan ecuaciones para el transporte de masa y la distribución de tracciones en las fronteras de grano. Se establece, y resuelve numéricamente, una ecuación diferencial para la eliminación de cavidades impulsada por la compresión isostática y la energía libre de superficie. Se discuten las diferencias cualitativas de este enfoque para la densificación con los modelos de "crecimiento de cuello" de Kuczynski y otros. Los resultados son curvas de densificación del material, tamaño de cavidad y razón de eliminación como funciones del tiempo. También se obtiene la dependencia del tiempo de sinterizado con la presión isostática y con el ángulo de capilaridad.

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## 1. Introduction

Sintering by hot isostatic pressing has been studied extensively during the last decades and theories have been developed in different directions. Technological applications of sintering of uranium pellets for nuclear reactors motivated the early work on the area on the 50's and presently there is interest in the powder metallurgy of superalloys. The early work of Kuczynski and others [1, 2, 3, 4] approached sintering as a coalescence of two material particles and since then the concept of "neck growth" has concentrated attention. Later works [5, 6] have considered sintering as more collective process where attention is focused on interparticle void elimination. Six or more mechanisms of mass transport occur when a powder aggregate is sintered [6]. These mechanisms are connected mainly with self diffusion along different paths in the material and only a few of them contribute effectively to densification. Grain boundary self diffusion has been considered an effective mechanism of mass transport during sintering [6].

The present work is an approach to sintering controlled by grain boundary self diffusion where the process is studied as interparticle void elimination and includes the effects of hot isostatic pressure and the capillarity angle formed at the points where voids are in contact with the grain boundaries. Results show that the void elimination approach has a differential equation with qualitative differences with the two particle coalescence method.

## 2. Geometry

Consider a compact array of monocrystalline long wires of radius  $r$ . In sintering conditions there is some definite time, which is not calculated here, after which the voids in the compact assume the shape shown in Fig. 1. In this configuration the curvature is constant and surface tensions are in equilibrium at the points where contact is made between void surfaces and grain boundaries. The angle formed by the tangent of the void surface and the plane of the grain

boundary, at the point of contact, is given by (see Fig. 2)

$$\alpha = \cos^{-1} \left( \frac{\gamma_b}{2\gamma_s} \right), \quad (1)$$

where  $\gamma_b$  and  $\gamma_s$  are the grain boundary and void surface free energies.

Let  $a$  be the distance from the center to the tip of the void. Then the curvature of the void surface is

$$\kappa = \frac{1}{u(\alpha)a}, \quad (2)$$

where

$$u(\alpha) = \frac{\sqrt{3}}{\sqrt{3} \sin \alpha - \cos \alpha}. \quad (3)$$

The area of a void cross section is

$$A = p(\alpha)a^2, \quad (4)$$

where

$$p(\alpha) = \frac{9(\alpha - \pi/6)}{(\sqrt{3} \sin \alpha - \cos \alpha)^2} - \frac{3\sqrt{3} \cos \alpha}{\sqrt{3} \sin \alpha - \cos \alpha}. \quad (5)$$

If the amount of densification prior to the initial formation of the configuration of Fig. 1 is assumed to be negligible, simple trigonometry can be used to obtain the initial value of  $a$ ,  $a_0$ , given by

$$a_0 = r \sqrt{\frac{(\sqrt{3} - \pi/2)}{p(\alpha)}}. \quad (6)$$

The distance from center to center of neighboring voids is  $2b$ , where

$$b = r \sqrt{\frac{\sqrt{3} \pi}{18} + \frac{\sqrt{3} p(\alpha) a^2}{9r^2}}. \quad (7)$$

Notice that during densification the void distance is going to be reduced.

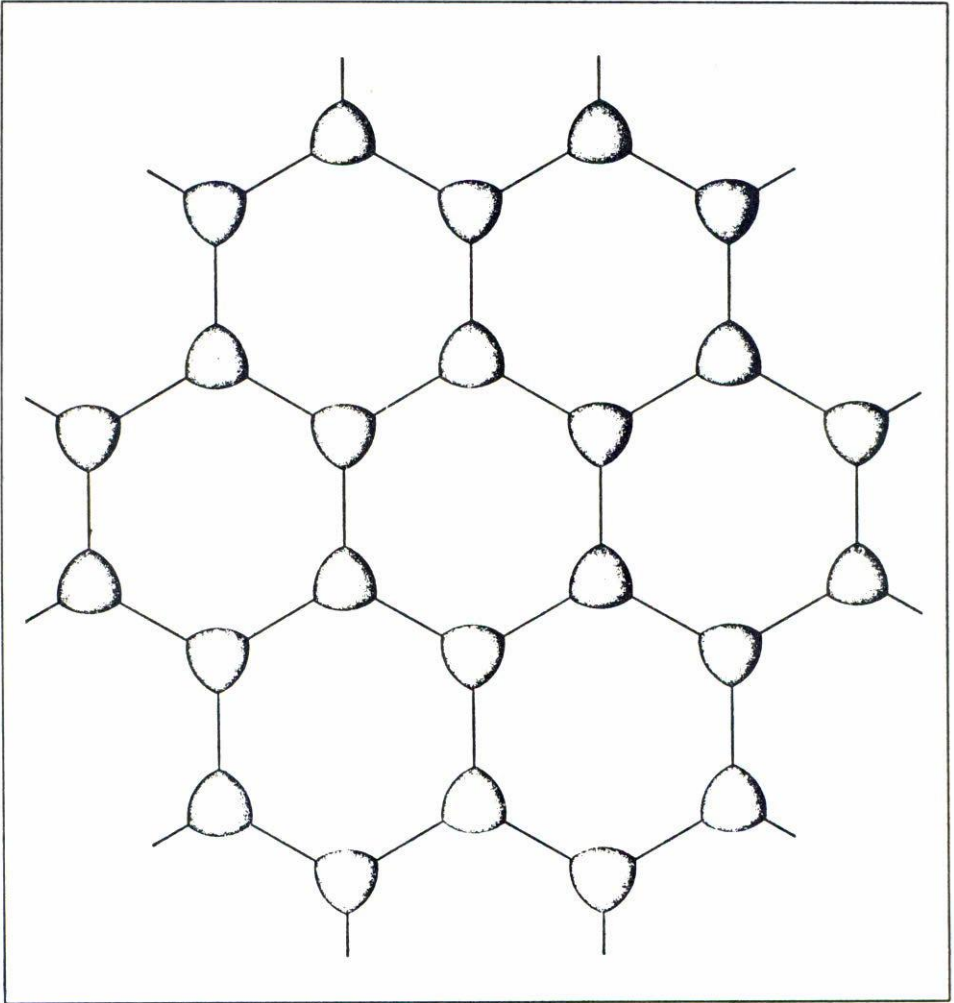


FIGURE 1. Array of voids after the first stage of densification. The amount of densification during this stage is considered negligible.

### 3. Self diffusion equations

If surface self diffusion is fast enough to keep the void curvature,  $\kappa$ , constant, then the sintering process is controlled by grain bound-

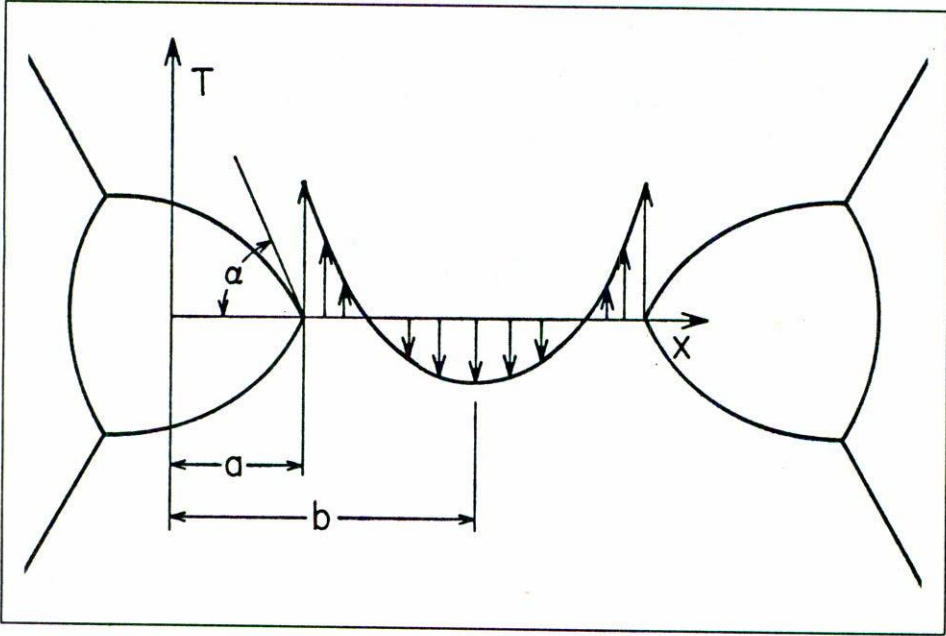


FIGURE 2. Traction distribution in a grain boundary. At point  $a$  there is chemical potential continuity and at the point  $b$  there is a zero flux condition by symmetry. Between points  $a$  and  $b$  the traction distribution averages the hydrostatic pressure.

ary self diffusion [7]. The chemical potential at any point on the void surface is [8]

$$\Delta\mu = -\Omega\gamma_s\kappa, \quad (8)$$

where  $\Omega$  is the atomic volume. In the grain boundary the diffusion equation is [9, 10]

$$\frac{D\delta\Omega}{kT} \frac{\partial^2 T_n}{\partial x^2} + \dot{W} = 0, \quad (9)$$

where  $D$  is the self diffusion coefficient in the grain boundary,  $\delta$  the effective grain boundary thickness,  $kT$  is the Boltzmann factor and  $\dot{W}$  is the rate of volume depletion per unit of grain boundary area.

Assuming that grain boundary self diffusion removes material from the neighboring grains uniformly [9], the traction distribution

will be parabolic as shown in Fig. 2. At the point  $a$  there is chemical potential continuity and at the point  $b$  there is, by symmetry, a zero flux condition. Also the traction distribution between points  $a$  and  $b$  averages the isostatic or hydrostatic pressure,  $\sigma$ , on the material and is given by

$$T_n(x) = \gamma_s \kappa + 3 \left( \frac{\sigma_b}{b-a} + \gamma_s \kappa \right) \left[ \frac{1}{2} \left( \frac{x-a}{b-a} \right)^2 - \left( \frac{x-a}{b-a} \right) \right], \quad (10)$$

where  $\sigma$  is assumed to be positive in compression conditions.

#### 4. Sintering

The rate of change of a void cross section is given by

$$\frac{dA}{dt} = 3\Omega\delta J(a), \quad (11)$$

where  $J(a)$ , the atomic flux coming into the void from each of the three adjacent grain boundaries can be obtained from Eq. (10) [7]:

$$J(a) = -\frac{3D}{kT(b-a)} \left[ \frac{\sigma_b}{b-a} + \frac{\gamma_s}{u(\alpha)a} \right]. \quad (12)$$

The differential equation for the variable  $a$  can be obtained from (4), (11) and (12):

$$\frac{da}{dt} + \frac{9D\delta\Omega}{2p(\alpha)kT a(b-a)} \left[ \frac{\sigma_b}{b-a} + \frac{\gamma_s}{u(\alpha)a} \right] = 0. \quad (13)$$

The differential equation can easily be integrated numerically. Solutions  $a(t)$  for different values of  $\Sigma = \sigma_r/\gamma_s$ , a proper adimensional variable associated to the hydrostatic pressure, are shown in Fig. 3. The time is given in units of

$$t_D = \frac{kT r^4}{D\delta\gamma_s\Omega}, \quad (14)$$

a variable which includes most of the intrinsic physical quantities of the material.

Figure 4 shows that the rate of void elimination increases to infinity at the final stage and this is consistent with the fact that no gaseous species in the voids are considered in this model. This behaviour of the velocity shows a qualitative difference with the results that may be expected in a model of sintering by coalescence of two particles. The later necessarily ends with a neck growth velocity approaching to zero [3].

Curves for the densification,

$$\rho = \frac{\rho_{\text{mat}}}{\left[1 + \frac{2p(\alpha)a^2}{\pi r^2}\right]}, \quad (15)$$

where  $\rho_{\text{mat}}$  is the density of the material without voids, as shown in Fig. 5. The dependence of the time of sintering with the hydrostatic pressure is described in Fig. 6. At low hydrostatic pressures the process is driven by capillarity forces. When the hydrostatic pressure is higher the sintering time becomes inversely proportional. However, since this model does not include plasticity, the results are limited to values of the hydrostatic pressure not big enough to induce power law creep.

The effect of the capillarity angle, that is, of the ratio of grain boundary to twice the void surface free energies, on the sintering time is shown in Fig. 7. The interesting interval for capillarity angles is from 60 to 80 degrees where many metals are included [11].

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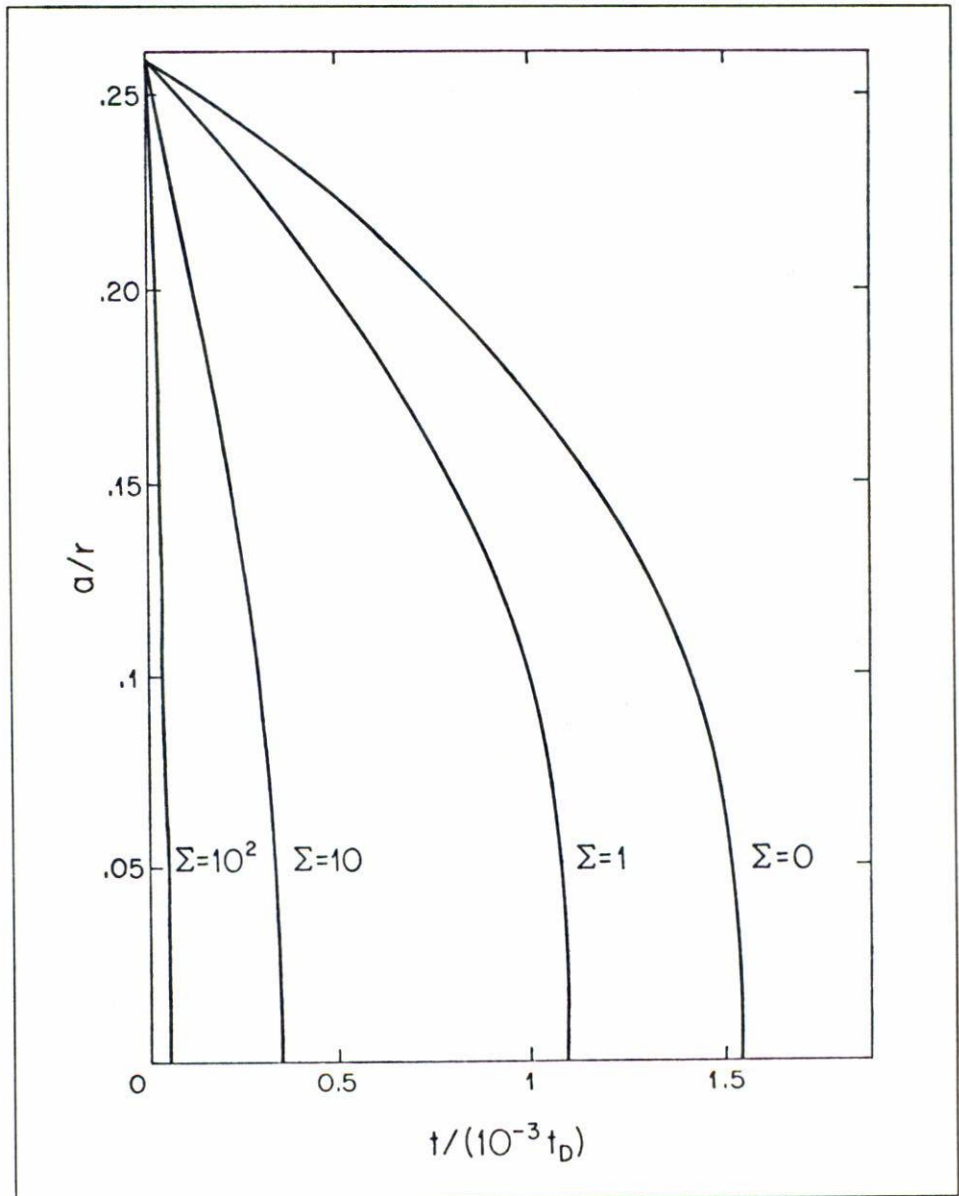


FIGURE 3. Solutions of the differential equation governing the sintering process for different values of the hydrostatic pressure in adimensional units.



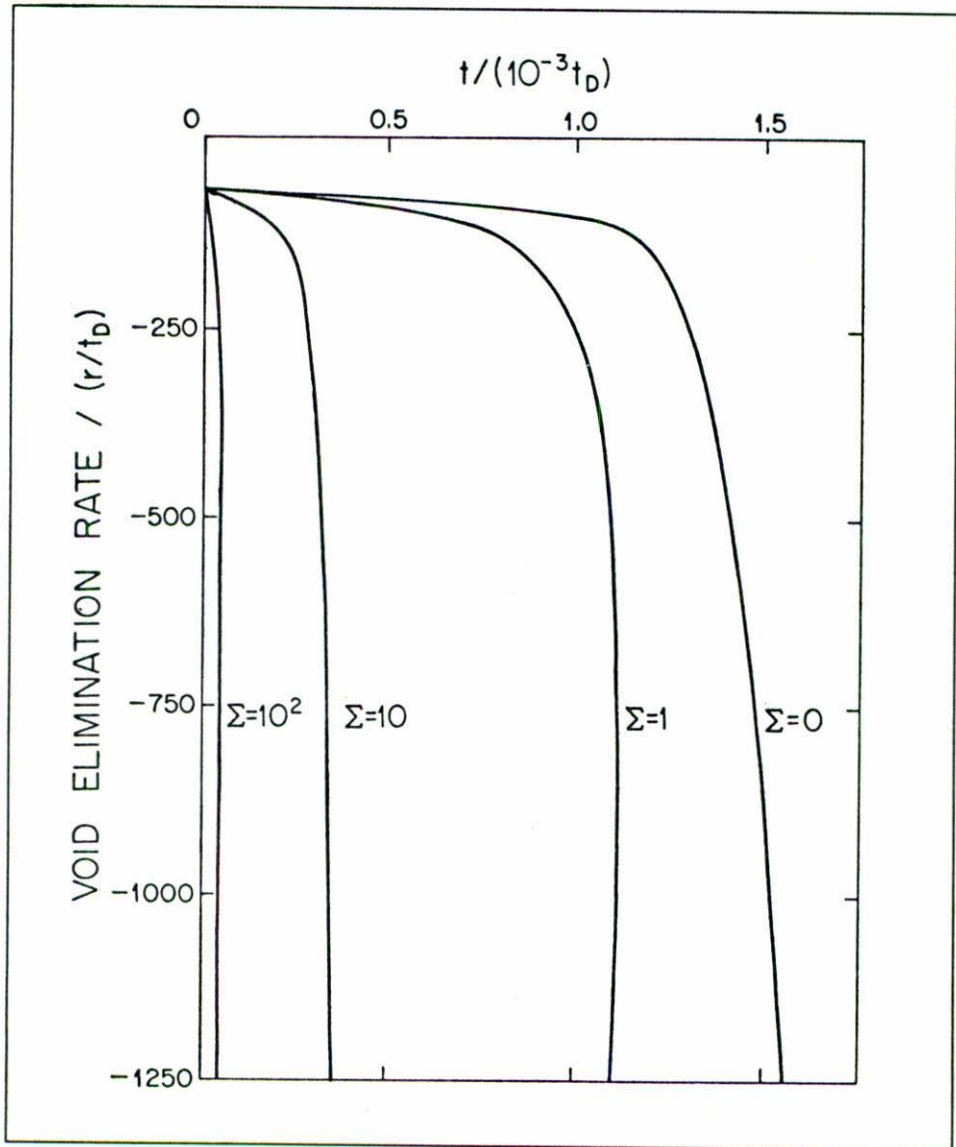
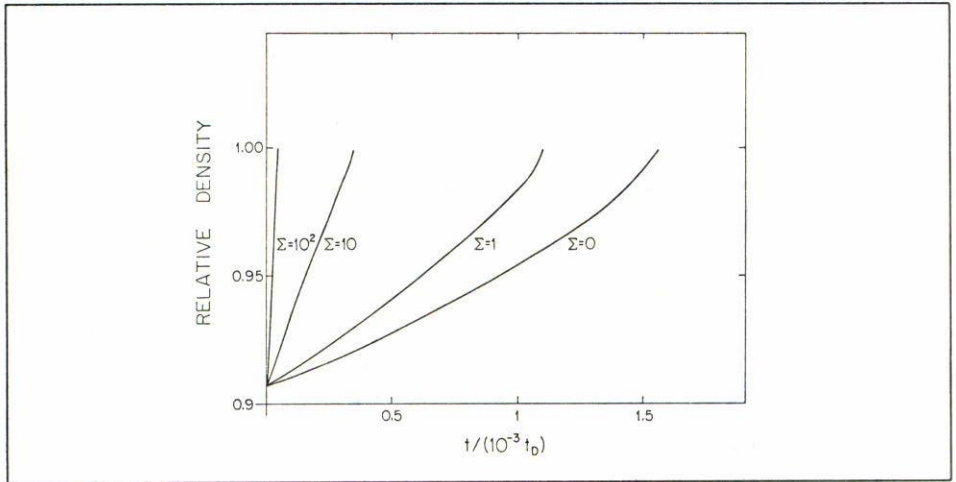
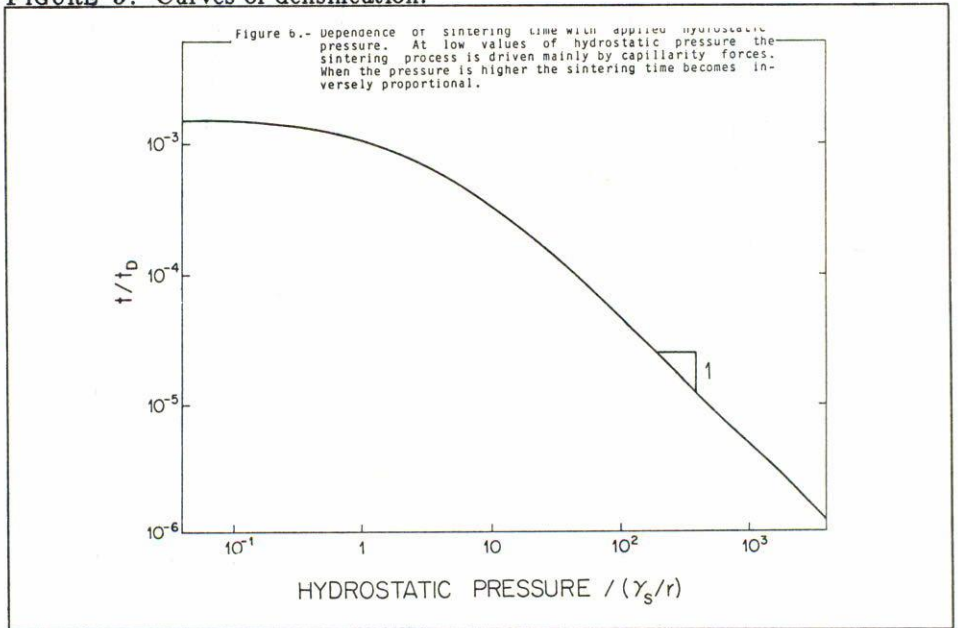


FIGURE 4. Velocity of void elimination. The fact of increasing to infinite at the final stage of sintering shows a qualitative difference with the two particle coalescence method where the driving force for sintering approaches at the same stage to zero.



**FIGURE 5.** Curves of densification.



**FIGURE 6.** Dependence of sintering time with applied hydrostatic pressure. At low values of hydrostatic pressure the sintering process is driven mainly by capillarity forces. When the pressure is higher the sintering time becomes inversely proportional.

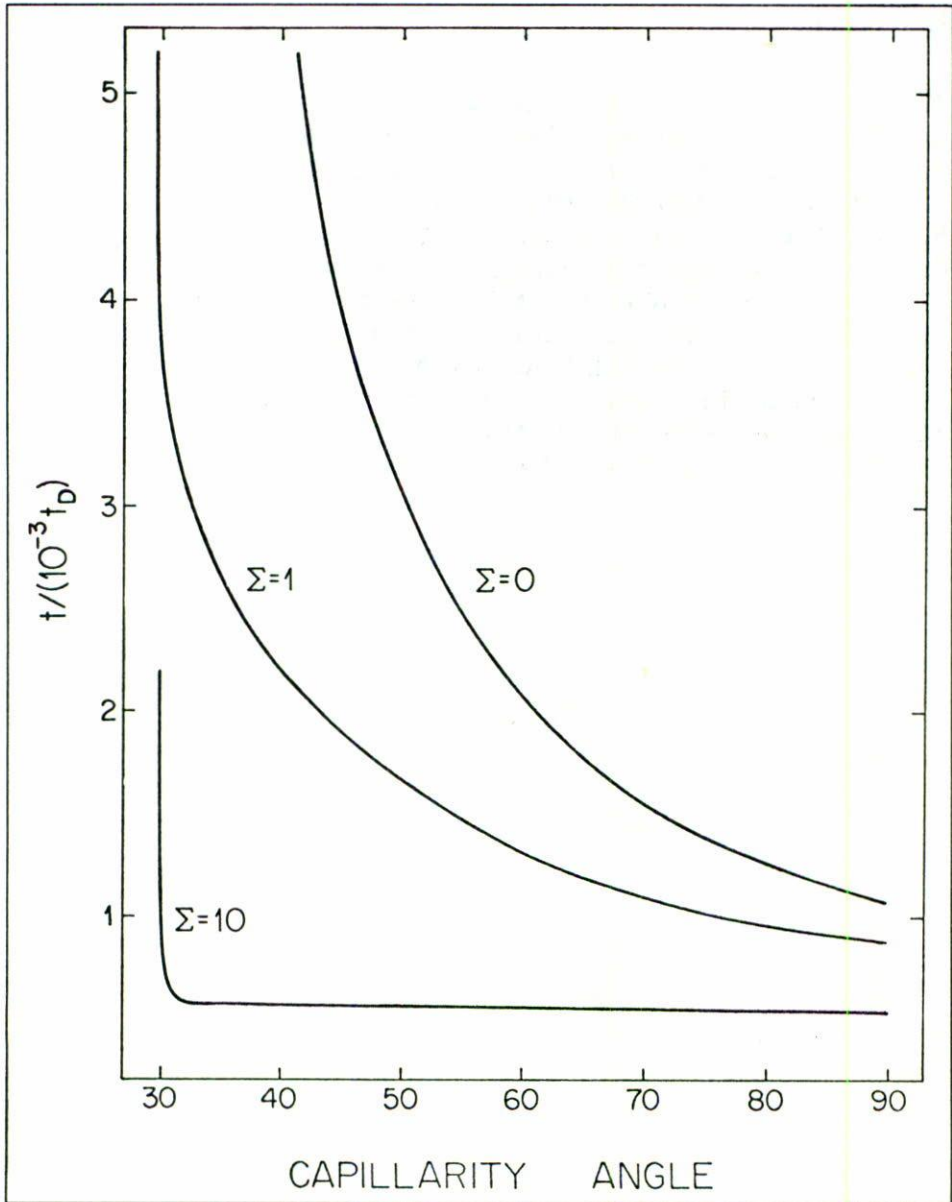


FIGURE 7. Effect of the capillarity angle on sintering time. The interval of interest is between 60 to 80 degrees where many metals are included.

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