

Emmons and Leigh tables for  $A$  as small as 0.53, 0.23, and 0.39, respectively.

The reactor dynamics problem can be slightly generalized to include the effect of a linear reactivity insertion or removal, so that  $d\rho/dt = b - aQ$ . Instead of the substitution (1) we now suppose that

$$F = -\alpha\rho/\rho_0, \quad \eta = \eta_1 + (\rho_0 t/\alpha),$$

$$A = a^2\rho_0^{-3}P_0, \quad B = b\rho_0^{-2}$$

in which  $\alpha$  and  $\eta_1$  are constants. Then

$$d^2F/d\eta^2 + Fd^2F/d\eta^2 = 0,$$

$$F(\eta_1) = -\alpha, \quad F'(\eta_1) = -B\alpha^2, \quad F''(\eta_1) = A\alpha^3,$$

and the quantities  $\alpha$  and  $\eta_1$  are selected so that  $F'(0) = 0$ ,  $F'(\infty) = 2$ . Numerical results may be obtained from the tables in (4) as follows. For each negative value of  $F(0)$  and each  $\eta_1$  used by Emmons and Leigh, values of  $F(\eta_1)$ ,  $F'(\eta_1)$ , and  $F''(\eta_1)$  can be read from the tables. Then  $\alpha = -F(\eta_1)$ ,  $B = -F'(\eta_1)/\{F(\eta_1)\}^2$ ,  $A = F''(\eta_1)/\{F(\eta_1)\}^3$ , and the total energy release  $Q(\infty)$  is such that

$$a\rho_0^{-2}Q(\infty) = B + 2\alpha^{-2}.$$

Of course  $\eta_1$  must be restricted to insure that  $F(\eta_1) < 0$ , in order that  $\alpha > 0$ . If the value  $\eta_0$  where  $F(\eta_0) = 0$  is found by interpolation in the tables, as well as the values  $F'(\eta_0)$  and  $F''(\eta_0)$ , then the state of the reactor at the time  $\tau$  when it has become critical again can be found as follows:

$$\rho_0 t^{-1}\tau = \alpha(\eta_0 - \eta_1), \quad a\rho_0^{-2}Q(\tau) = B + \alpha^{-2}F'(\eta_0),$$

$$P(\tau)/P_0 = F''(\eta_0)/F''(\eta_1).$$

Unfortunately, all the values  $F'(\eta_1)$  in the tables are positive, so that only negative values of  $B$  can be handled in this manner. Hence, linear reactivity removal problems can be solved, but linear reactivity insertion problems cannot until the tables in (4) are extended to negative values of  $\eta$ .

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Received July 30, 1962

Revised January 16, 1963

## Sintering of $\text{UO}_2$ in Carbon Dioxide Atmosphere

As compared with conventional hydrogen sintering, the use of slightly oxidizing atmospheres in the high temperature stage of the  $\text{UO}_2$  sintering cycle offers the advantage of reducing both temperature and time necessary to attain a certain fired density.

Among the possible atmospheres of this kind, steam has been widely and thoroughly investigated for many years, and several setbacks have shown up together with the merits, mainly as a consequence of the vicious nature of steam as a furnace atmosphere (1, 2).

Carbon dioxide, on the contrary, which is free of almost all of the disadvantages of steam, has been suggested as a sintering atmosphere (3), but, as far as we know, never used. We tried it,<sup>1</sup> in comparison with conventional hydrogen sintering, on a certain brand of depleted  $\text{UO}_2$  powder, supplied by the Mallinckrodt Chemical Works, St. Louis, Mo., U.S.A., the properties of which are summarized in Table I. Sintering cycles, both in  $\text{H}_2$  and  $\text{CO}_2$ , are shown in Fig. 1.

Pellets were pressed to about 6 gm/cc from a powder with 2 wt.% polyethylene glycol, 1 wt.% polyvinyl/alcohol

TABLE I

PROPERTIES OF MCW CERAMIC GRADE DEPLETED POWDER

Real density ( $\text{CCl}_4$ ), gm/cc	10.34
Tapped density, gm/cc	2.18
Average particle diameter (Fisher), $\mu$	1.28
Total surface area (B.E.T.), $\text{m}^2 \text{gm}^{-1}$	3.45
External surface (Blaine), $\text{m}^2 \text{gm}^{-1}$	0.42
Roughness factor	8.47
O/U ratio	2.03

and 2 wt.% vegetal stearine. Comparative results in  $\text{CO}_2$  and  $\text{H}_2$  are shown in Fig. 2, while the influence of time and temperature in  $\text{CO}_2$  is shown in Fig. 3. The sintering activation energy, calculated according to Jordan and Duwez (4), was  $40,300 \pm 1300$  cal/mol (Fig. 4), to be compared with  $32,000 \pm 3200$  cal/mol for the same powder in steam (1) and  $76,000$  cal/mol in hydrogen (5). Whether this discrepancy is due to the inaccuracy of Jordan and Duwez's method or to a basically different mechanism for steam and  $\text{CO}_2$  sintering, is not known.

The organic additions to the powder were in general deleterious to the final density, as shown in Fig. 5. The properties of the  $\text{CO}_2$  sintered pellets (porosity, grain size, etc.) compared very well with those of  $\text{H}_2$  and steam sintered pellets of corresponding densities. In particular, the stoichiometry of the final product was insured by the final hydrogen cooling and no carbon pickup was detected.

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<sup>1</sup> FIAT patent n° F-1534.

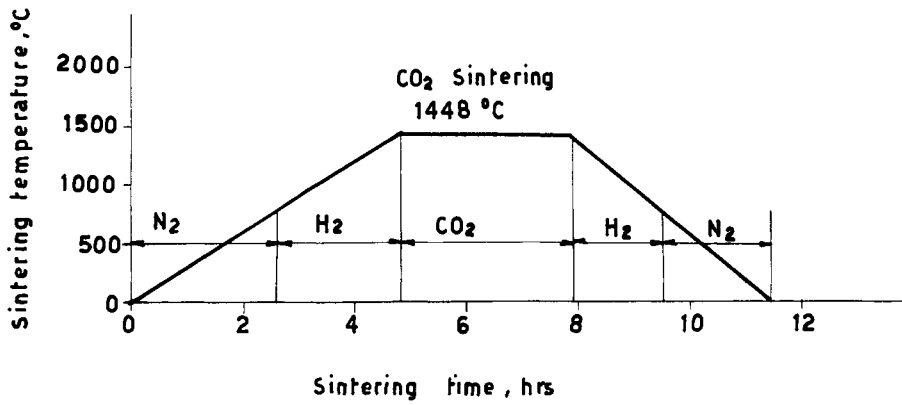
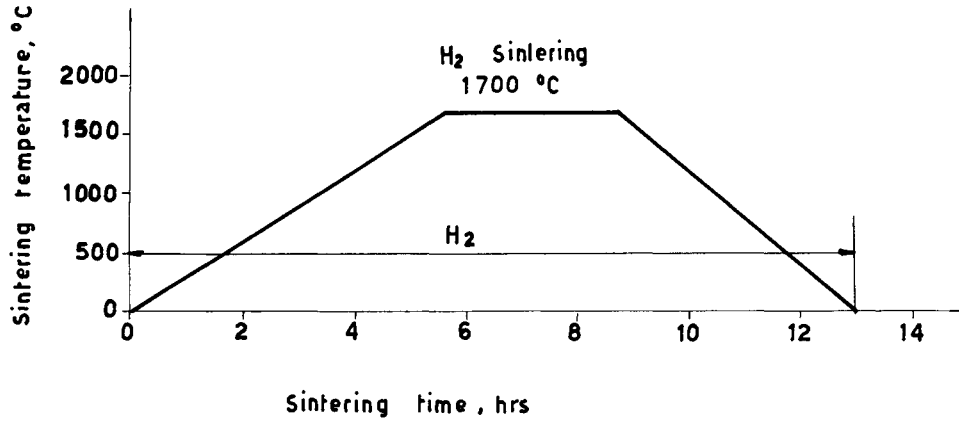


FIG. 1. H<sub>2</sub> and CO<sub>2</sub> sintering cycles

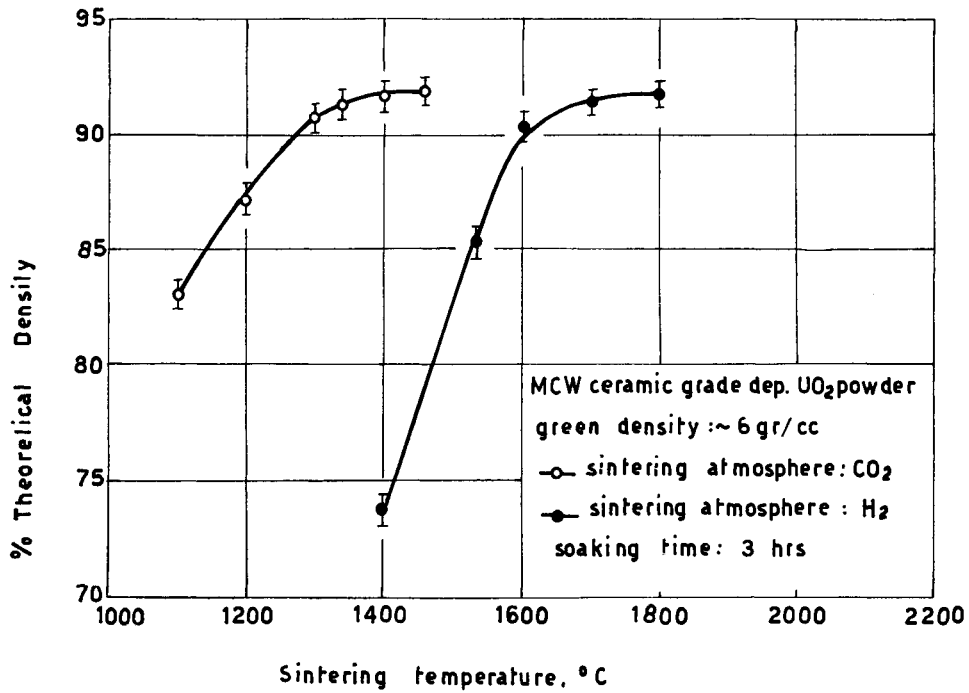


FIG. 2. Comparison between CO<sub>2</sub> and H<sub>2</sub> sintering

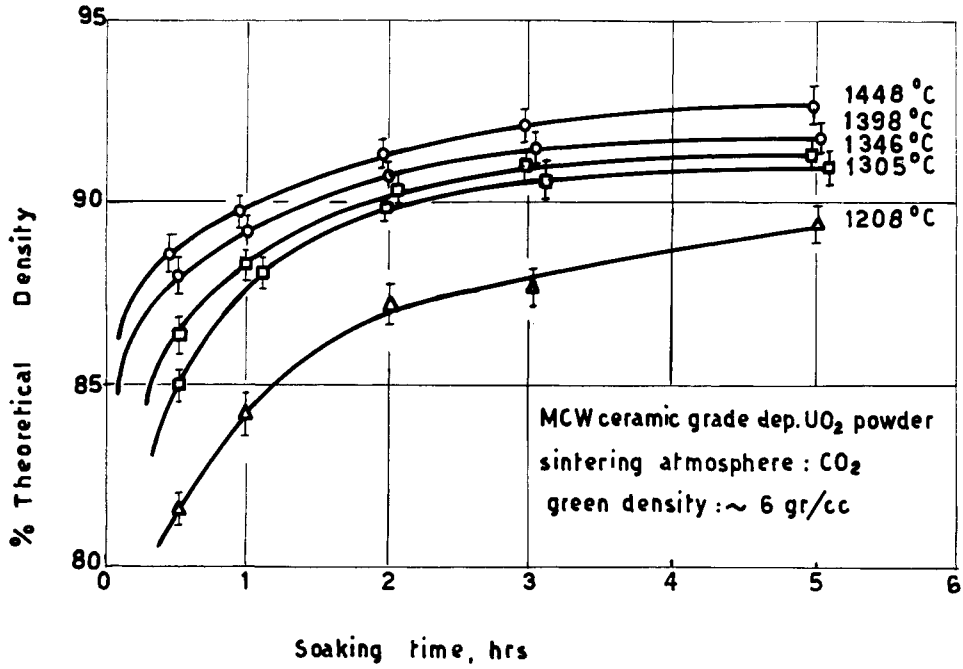


FIG. 3. CO<sub>2</sub> sintering at different temperatures

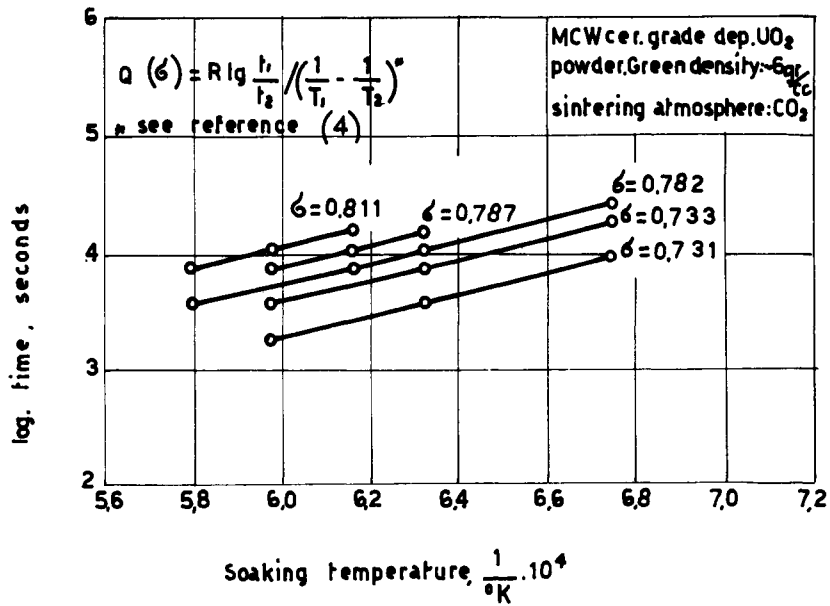


FIG. 4. Variation of densification rate as a function of temperature.

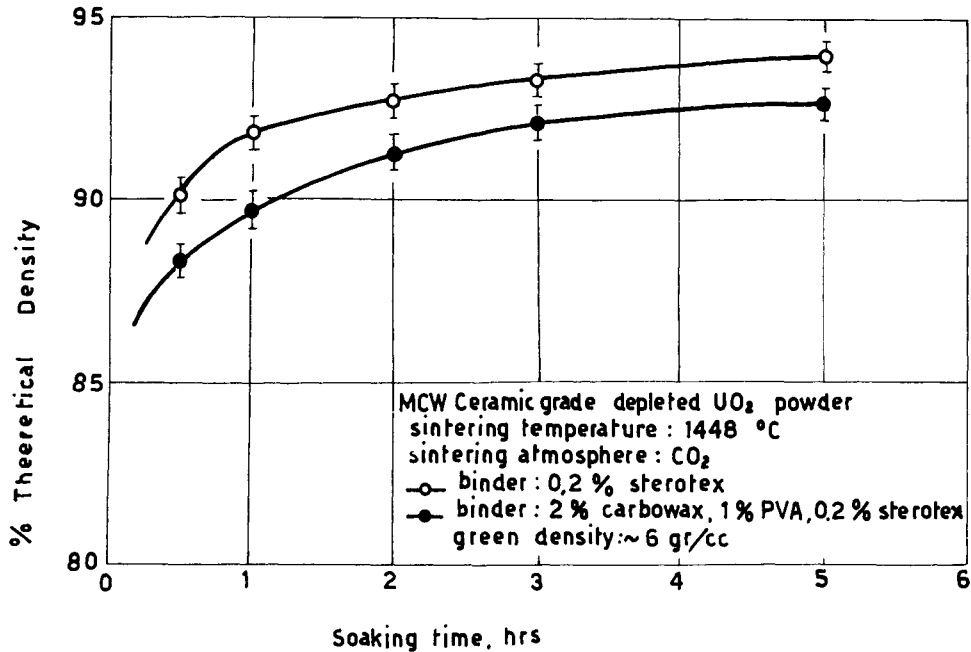


FIG. 5. Effect of organic additives on  $CO_2$  sintering of  $UO_2$  powder.

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Received November 1, 1962

Revised January 28, 1963

### A Remark on the Measurement of the Diffusion Coefficient for Thermal Neutrons\*

In a recent article in this Journal, Baumann (1) has reported a measurement of the diffusion coefficient ( $D$ ) of thermal neutrons in heavy water as a function of temperature in the range 20°C to 220°C. The technique used was to measure the diffusion length ( $L$ ) as a function of copper absorption cross section ( $\Sigma_{Cu}$ ). The copper was added in the form of thin tubes. The reciprocal diffusion coefficient is the slope of curve of  $1/L^2$  vs.  $\bar{\Sigma}_a$  at  $\bar{\Sigma}_a = 0$ . Here  $\bar{\Sigma}_a$  is the Maxwellian (at the moderator temperature) averaged total absorption (copper plus heavy water) cross section. Several corrections must be made to the raw experimental data and these have been made with great care. The correction for neutron spectrum, however, was made on an intuitive basis and overestimates the correction. In this note we show how the correction should be made, estimate the size of the

correction from a theoretical scattering model for heavy water, and recompute  $D$  from Baumann's data.

Baumann recognizes that the neutron spectrum is distorted from the equilibrium Maxwellian spectrum at the temperature of the medium,  $T_0$ , by absorption and diffusion. He then characterizes this distorted spectrum by a Maxwellian at a "neutron temperature"  $T_n$  and uses a formula given by Coveyou, Bate, and Osborn (2) to relate  $T_n$  to  $T_0$  and  $\bar{\Sigma}_a$ . Using an empirical expression for the Maxwellian averaged  $D$  as a function of temperature, and the  $\sqrt{T_0/T_n}$  variation for the absorption cross section, the diffusion length is corrected back to a neutron temperature equal to the moderator temperature. Our objections to this procedure are:

1. The formula of Coveyou was derived for slowing down spectra where most of the distortion is caused by the  $1/E$  tail of the spectrum. In the present experiment there is no slowing-down source, the spectra have no  $1/E$  tail, and the distortion for a given absorption is much smaller (3-6).

2. The actual spectrum distortion can only be approximated by a shifted Maxwellian. Thus, while it may be possible to describe the shift of one parameter (say  $\bar{\Sigma}_a$ ) by an effective Maxwellian temperature shift, the shift of another parameter ( $D$ ) may require a different Maxwellian temperature shift.

The first of these objections is the most important since it determines the size of the correction. The second objection merely states that the form of the correction used was intuitive and lacked a firm theoretical foundation. An appropriate treatment of this correction has been discussed by Honeck (7). Koppel and Starr (8) have treated a similar measurement in light water in the manner described below.

The reciprocal diffusion length in a poisoned moderator can be expressed by

$$\frac{1}{L^2} = \alpha_1 \Sigma_{a0} - \alpha_2 \Sigma_{a0}^2 + \dots \quad (1)$$

\* Research performed under the auspices of the U.S.-A.E.C.