



## Section 12.2. Mechanical properties

**Contribution to irradiation creep arising from gas-driven  
bubble growth**C.H. Woo <sup>a,\*</sup>, F.A. Garner <sup>b</sup><sup>a</sup> Department of Mechanical Engineering, The Hong Kong Polytechnic University, Hong Kong, People's Republic of China<sup>b</sup> Pacific Northwest Laboratory <sup>1</sup>, Richland, WA, USA**Abstract**

Although the majority of the irradiation creep data have been developed in fast reactors where the helium generation rates are rather low, many applications of such data involve neutron or charged particle spectra where the helium generation rates are much larger. It also appears that hydrogen can accumulate in austenitic steels when large amounts of cogenerated helium are also present. In such spectra it has been recently predicted that large gas generation rates can lead to an acceleration of the irradiation creep rate under some conditions often found or expected in some fusion designs, near-core components of light water reactors, and in some accelerator-driven spallation devices. In the present paper, this possibility is theoretically explored and shown to be feasible. As a result, an additional degree of conservatism should be included when fast reactor data are applied to spectral environments where large gas generation rates are expected. © 1999 Elsevier Science B.V. All rights reserved.

**1. Introduction**

Garner et al. [1,2] have shown that, under irradiation, the creep law may be represented in terms of the effective creep rate and the effective stress by the empirical relation

$$\frac{\dot{\epsilon}}{\bar{\sigma}} = D\dot{S} + B_0, \quad (1)$$

where  $\dot{S}$  is the swelling rate,  $D$  is the creep-swelling coupling constant, and  $B_0$  takes into account the non-swelling component of irradiation creep. More recently, Garner and coworkers [3–5] predicted that  $B_0$  would be independent of the dose rate and irradiation temperature over a very wide range of both variables, but would be somewhat sensitive to the helium/dpa ratio. This latter suggestion arises from the consideration that helium bubbles can also contribute a  $D\dot{S}$  component of irradiation creep that would appear to be an enlarge-

ment of  $B_0$ , since “swelling” per se had not yet occurred, and therefore the  $D\dot{S}$  contribution to creep also had not yet started.

It was also predicted that helium was not the only transmutant gas that had to be considered, because hydrogen might be playing a bigger role than previously envisioned in both void swelling and irradiation creep under certain circumstances [4,6,7]. These circumstances involve irradiation at relatively low temperatures and displacement rates, especially when there is large concurrent generation of both helium and hydrogen. Under such conditions, it is proposed that transmutant or environmentally produced hydrogen, which normally migrates out of steels at most reactor-relevant temperatures, can be trapped in helium-nucleated bubbles and form relatively stable hydrogen gas. The combined hydrogen and helium then drive bubble growth and accelerate bubble-to-void conversion. Greenwood and Garner [7] have also shown that the formation of  $\text{Ni}^{59}$ , which produces most of the helium in stainless steels in highly thermalized neutron spectra, also produces a significant new contribution to the hydrogen production. At neutron exposures where helium begins to build up via  $\text{Ni}^{59}$  and trap hydrogen, the hydrogen generation rate is increased.

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Such conditions are not usually found in fast reactors, in which most of the irradiation creep data are obtained and used for the design and maintenance of many other types of irradiation devices. However, high gas generation rates are common in several types of devices, such as some fusion designs, like ITER, as well as in some near-core components of pressurized water reactors, and in some accelerator-driven spallation neutron designs. While the possibility of factors of two to three under-estimation of the  $B_0$  component of creep might not seem at first glance to be very significant, there are some situations where such an enhancement can have important consequences. This is especially true when stress relaxation of long-lived components, such as bolts or tie-rods, can slacken preload stresses that must be maintained for safe operation. In order that the fast reactor data be applied to such cases with greater confidence, a better understanding of the effect of gas generation on irradiation creep is called for.

This paper addresses the possibility that  $B_0$  may be sensitive to the gas generation rate, especially at relatively low temperatures and displacement rates typical of mixed spectrum reactors, some fusion designs and some accelerator-driven spallation environments. For illustration of the concept, calculations are performed specifically for 316 stainless steel tubes, internally pressurized at a hoop stress of 100 MPa and irradiated at 350°C at an NRT dpa rate of  $10^{-7}$  dpa/s. The possible contribution of hydrogen to this process will not be addressed directly in this paper, but a range of He/dpa ratios large enough to cover the possible effect of both gases will be used. Unlike the situation in mixed spectra reactors where the helium via  $\text{Ni}^{59}$  formation is nonlinear, we will assume constant He/dpa ratios to demonstrate the concept. In this sense, such constant rates are closer to the accelerator-driven spallation neutron case.

## 2. Effects of gas accumulation on irradiation creep

Irradiation creep due to Stress Induced Preferred Absorption (SIPA) and SIPA Induced Growth [8] (SIG) and, more recently, the relationships between void swelling and irradiation creep under cascade damage conditions [9–11] have been studied in some detail. The production and annihilation of primary interstitial and vacancy clusters during cascade irradiation has also been accounted for in irradiated pressurized tubes in Ref. [11]. However, the effect of gas generation (helium/hydrogen) has not been considered in this line of work.

The pressure created by the accumulation of gases at voids reduces the rate of emission of the vacancies and increases that of the interstitials. The strain field of a highly pressurized cavity may also induce a bias on the sink strength of the void. To account for the effect of gas generation on the creep behaviour, the existing theory

would have to be modified accordingly. We are interested to find out how such modifications will affect the creep law derived earlier.

In the absence of helium generation, theoretical analysis shows [11] that the creep law can be put in exactly the same form as the empirical relation proposed by Garner et al. [1,2] as in Eq. (1). Expressions for  $D$  and  $B_0$ , in terms of the hoop stress, the dislocation structure and the swelling rate, have been derived.

We now consider the case where transmutation gases are continuously generated by irradiation. Suppose  $G$  is the effective point-defect production rate after intracascade recombination is taken into account [12]. If  $\alpha$  is the fraction of intracascade recombination, then  $G = (1 - \alpha)K$ , with  $K$  being the displacement damage rate (NRT). We follow a general formulation where there are  $N$  classes of dislocations, including the primary clusters, each with Burgers vector  $\mathbf{b}^{(k)}$ , line density  $\rho_k$ , bias factor for vacancies  $Z_k$  (i.e.,  $Z_k \rho_k$  is the dislocation sink strength), and bias  $p_k$ . Under the operation of DAD (*Diffusional Anisotropy Difference*),  $p_k$  can be positive or negative, as determined by the geometric orientation by the line direction relative to the principal crystallographic directions [13]. Let  $\rho$  be the total dislocation density,  $k_d^2$  be the total sink strength of the dislocations for the vacancies, and  $k_c^2$  the void sink strength for vacancies. In addition we assume a gas pressure-induced bias for voids denoted by  $p_c$ . Let us first neglect recombination due to long-range migration. Within the Production Bias Theory [12], the steady-state concentrations of the freely migrating interstitials,  $C_i$ , and vacancies,  $C_v$ , are given by

$$(1 - \epsilon'_i)G - [(1 + p_c)k_c^2 + (1 + \bar{p})k_d^2]D_i C_i = 0, \quad (2)$$

$$(1 - \epsilon'_v)G - (k_c^2 + k_d^2)D_v C_v = 0, \quad (3)$$

In Eqs. (1) and (2),  $D_v$  and  $D_i$  are the vacancy and interstitial diffusion coefficients, respectively,  $\epsilon'_v$  and  $\epsilon'_i$  are given by

$$\epsilon'_v = \epsilon_v - K_v^e/G \quad \text{and} \quad \epsilon'_i = \epsilon_i - K_i^e/G. \quad (4)$$

Here the vacancy emission rate  $K_v^e$  is a sum of contributions from the voids,  $K_c$ , the interstitial loops,  $K_{il}$ , the network,  $K_N$ , the Primary Vacancy Clusters (PVCs),  $K_{vc}$ , and the Primary Interstitial Clusters (PICs),  $K_{ic}$ . The interstitial emission rate  $K_i^e$  is the equivalent of  $K_v^e$ . In practice, however, it is only significant for the voids under a very high gas pressure and at a high enough temperature. We define the mean bias factor  $Z$  and the mean bias for dislocations  $\bar{p}$  by

$$Z = \sum_k f_k Z_k \quad \text{with} \quad f_k = \rho_k / \rho, \quad (5)$$

$$\bar{p} = \sum_k Z_k p_k \rho_k / Z \rho. \quad (6)$$

Following the analytic procedure in Ref. [9], we found that the total strain rate under an external stress  $\sigma_{mn}$  is given by

$$\dot{\epsilon}_{mn} = \frac{\dot{S}}{3} + \sum_k f_k \left( \hat{b}_m^{(k)} \hat{b}_n^{(k)} - \frac{1}{3} \right) \dot{S} + (1 - \epsilon'_i) G \Delta \varphi \frac{\sigma_{mn}}{\mu} \times \sum_k q_k \hat{b}_m^{(k)} \hat{b}_n^{(k)} \left( \hat{\omega}_m^{(k)} \hat{\omega}_n^{(k)} - \sum_l f_l \hat{\omega}_m^{(l)} \hat{\omega}_n^{(l)} \right). \quad (7)$$

Here  $\hat{b}_m^{(k)}$  is the  $m$ th Cartesian component of the unit vector in the direction of  $\mathbf{b}^{(k)}$ ,  $\hat{\omega}_i^{(k)}$  the  $i$ th Cartesian component of the unit Burgers vector for loops of the  $k$ th class, or  $\sqrt{-2}$  times the  $i$ th Cartesian component of the unit line direction vector for line dislocations, and  $q_k$  the ratio of the dislocation sink strength of the  $k$ th class to the total sink strength for interstitials (i.e. including contributions from the primary clusters, see Ref. [14]).

Eq. (7) has exactly the same form as Eq. (34) of Ref. [9]. Gas production affects the nucleation and growth of cavities and therefore the void sink strength and swelling rate, its effects being contained in  $\dot{S}$  and  $q_k$ . Since Eq. (7) is the basis of the creep equations (11) and (12) of Ref. [11], it is clear that they are also applicable to cases where gas generation effects have to be accounted for. However, for a quantitative study, the swelling rate  $\dot{S}$  and the relative sink strength of the dislocations  $q_k$  have to be obtained from a numerical integration of the swelling-rate equation.

Let us consider a specific case in which only helium is generated. For simplicity, we assume all of the helium is accumulated at the voids. The helium gas pressure in the voids and the corresponding vacancy and interstitial emission is calculated following Refs. [15,16].<sup>2</sup> We assume at the start of the integration ( $10^{-3}$  dpa), the bubble nucleation process is completed, and the void nuclei are in the form of helium bubbles with a temperature-dependent number density. The starting bubbles are assumed to be in equilibrium [16], that is, the surface tension is balanced by the gas pressure. Subsequent void growth is caused by the net vacancy influx and the helium accumulation. The growth rate is limited by the emission of the vacancies through surface of the void when the combined force of the gas pressure and the vacancy influx is not enough to support the void from collapsing under the action of the surface tension. The strain rate tensor is calculated from the dislocation climb velocities  $V_k$  according to

$$\dot{\epsilon}_{mn} = \sum_k V_k \rho_k b_k \hat{b}_m^{(k)} \hat{b}_n^{(k)}. \quad (8)$$

The  $V_k$ 's are in turn calculated from rate equations following the formulation of Woo and Semenov [14],

with SIPA taken into account in calculating the dislocation bias of the various classes of dislocations. The dislocation densities are calculated according to Ref. [11]. In addition, recombination due to long-range migration is also taken into account, following Ref. [12].

The hydrostatic component of the strain rate tensor gives the swelling rate used in the integration, and the shear components give the creep rates as a function of dose. At the same time the relative total sink strength of the dislocations  $q$  to be used in Ref. [11] can also be obtained. We note that in this calculation only the line dislocations are taken into account in  $q$ , the primary clusters are neglected. For an irradiated internally pressurized tube, we apply a biaxial stress with a two to one stress ratio. The creep constants  $D$  and  $B_0$  can then be calculated following Ref. [11].

### 3. Results

Calculations are performed for 316 stainless steel tubes, internally pressurized at a hoop stress of 100 MPa and irradiated at 350°C assuming an NRT dpa rate of  $10^{-7}$  dpa/s. We consider four helium production rates: 0.5 appm/dpa, 5 appm/dpa, 20 appm/dpa, and 40 appm/dpa. The input parameters are the same as those in Refs. [12,14] (upper limits of network dislocation density of  $10^{15} \text{ m}^{-2}$ , loop number density of  $5 \times 10^{21} \text{ m}^{-3}$  and void number density of  $5 \times 10^{22} \text{ m}^{-3}$ , respectively are imposed).

The swelling and the creep rates (in units of  $\text{dpa}^{-1}$ ) for different helium generation rates are plotted against the dose in Fig. 1(a) and (b). It can be seen that the helium generation rate indeed has a significant effect on both the swelling and creep rates at low doses ( $< 1$  dpa), in the gas-driven/enhanced swelling regime. It can also be seen from this calculation that swelling at these low temperatures is indeed possible, as observed in low carbon stainless steel in PWRs [3,4]. Due to the higher void number density used, the swelling rate in these figures is higher than the observed values [3,4]. Using a void number density closer to the observed value (i.e.,  $\sim 10^{21} \text{ m}^{-3}$ ), the calculated results agree well with the observed values (i.e.,  $> 0.01\%$  but,  $< 0.1\%$  at a dose of between 0.1 and 1 dpa). In all cases, a large swelling of over 10% can be obtained at doses close to 100 dpa. Indeed, experimental results presented at this conference [17] support the prediction that swelling levels of 10% or greater can be reached at these temperatures and doses.

The creep rates, on the other hand, is less sensitive to the void number density. The calculated diametral creep rate compares well with the experimental values of about  $10^{-4} \text{ dpa}^{-1}$  in Ref. [18]. The effect of helium production is apparent and is confined to the low dose regime.

<sup>2</sup> A minor error in Eq. (15) of Ref. [15] has been corrected.

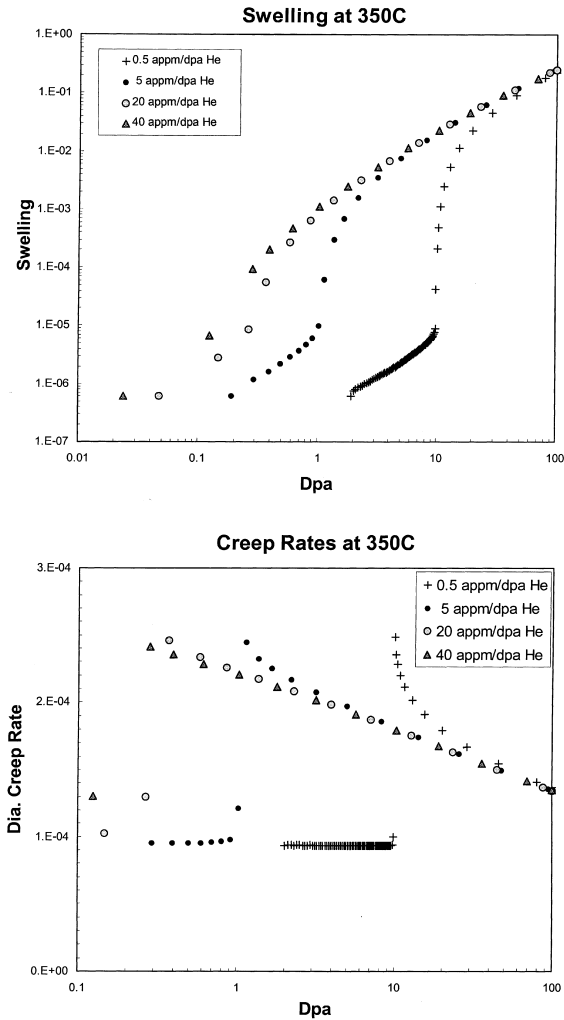


Fig. 1. Calculated (a) swelling and (b) creep rate at 350°C for a dose rate of  $10^{-7}$  dpa $^{-1}$  as a function of dose for austenitic stainless steels for different helium generation rates.

We can understand this behaviour through the relationship among the four variables: swelling rate (Fig. 2), creep rate, helium generation rate and dislocation anisotropy. In the low dose regime, when the cavity radius is smaller than the critical radius, the swelling rate is very small, and depends on the helium generation rate. In this case, the creep rate is dominated by the  $B_0$  term, which has been shown to increase when the dislocation anisotropy decreases from the maximum [11]. Earlier studies have also shown that the dislocation anisotropy decreases with increasing swelling rate [11,18]. Thus, increasing the helium production rate increases the swelling rate, reduces the dislocation anisotropy and finally increases  $B_0$  and the creep rate. Thus, the reason for the creep rate sensitivity to the helium generation rate in this regime lies in the relation between the creep

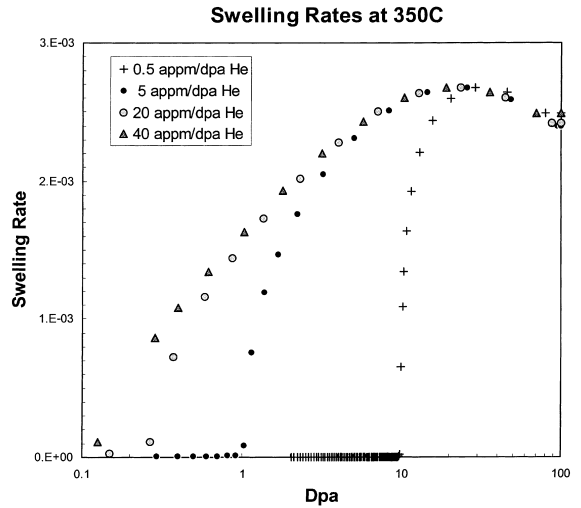


Fig. 2. Calculated swelling rate at 350°C for a dose rate of  $10^{-7}$  dpa $^{-1}$  as a function of dose for austenitic stainless steels for different helium generation rates.

rate and the swelling rate, through the dislocation anisotropy.

At a higher dose, as the bubbles grow through the critical radius, the swelling rate increases sharply and becomes independent of the helium generation rate. The sharply higher swelling rate reduces the dislocation anisotropy and sharply increases  $B_0$ . At the same time, the  $DS$  term starts to contribute significantly to the creep rate, producing a rapid increase in the dose regime between 0.1 and 1 dpa. The subsequent void growth reduces the relative dislocation sink strength, translating into a creep rate that decreases with increasing dose and is insensitive to the helium generation rate. In Fig. 3(a) and (b), we plot the values of  $D$  and  $B_0$  as a function of dose for different rates of helium generation. The dependence of both values on the helium generation rate is obvious. Similar to the creep rate and swelling rate, the dependence only concentrates in the low dose regime when the cavities are below the critical size. The dependence occurs, as discussed in the foregoing, and earlier in the literature [11,18], through the effect of the swelling rate on the dislocation anisotropy. At high dose, the reduction of the relative dislocation sink strength as the voids grow is the mainly responsible for the drop of both  $D$  and  $B_0$ . The values of both parameters agree well with experimental values.

The calculation has been repeated for a dose rate of  $10^{-6}$  dpa s $^{-1}$  with similar results, except for the factor of three reduction in the swelling rate, creep rate, as well as  $B_0$ . This reflects a square-root dose-rate dependence due to the enhanced recombination resulting from a higher cluster density generated from cascade debris from a higher dose rate. This result is to be expected from the present calculation where the primary clusters are as-

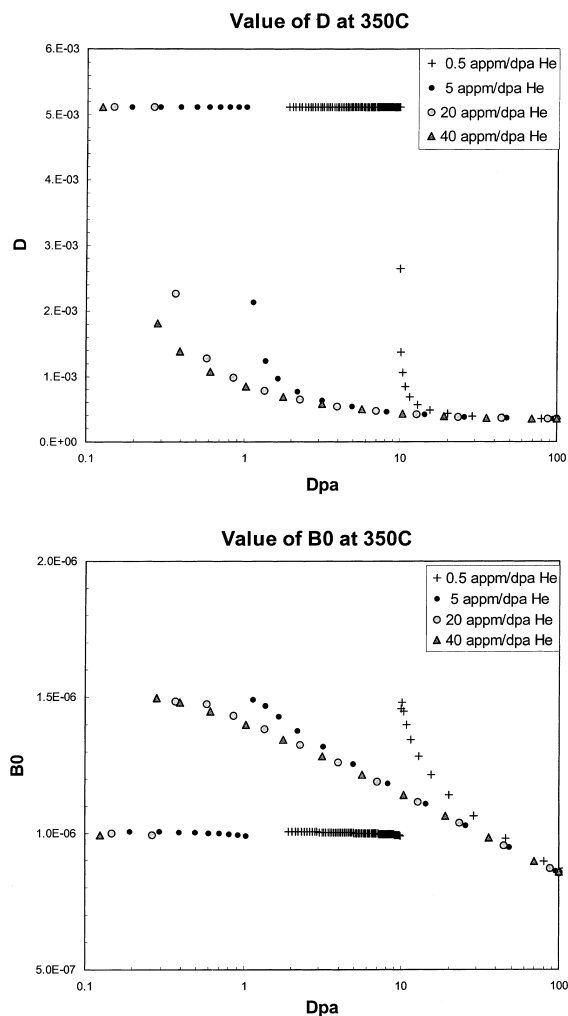


Fig. 3. Calculated creep compliances (a)  $D$  and (b)  $B_0$  at 350°C for a dose rate of  $10^{-7}$  dpa $^{-1}$  as a function of dose for austenitic stainless steels for different helium generation rates.

sumed not to contribute to the SIPA process as normal line dislocations. This may cause the dose-rate dependence of  $q$  to be over-estimated. The value of  $D$ , on the other hand is not sensitive to the dose rate.

The effect of void number density is also investigated. At a maximum void number density of  $10^{21}$  m $^{-3}$  the results show substantially less swelling, and less dependence (Fig. 4(a)) on the helium generation rate, for the three rates studied. At a smaller void number density, the gas accumulation per void is much faster, and the voids can get through the critical size much faster, without showing too much sign of dependence on the helium generation rate. The same effect is also found for the creep rates (Fig. 4(b)). As a consequence, for the three helium generation rates used in the present calculation, the assumption that all helium generated ends up

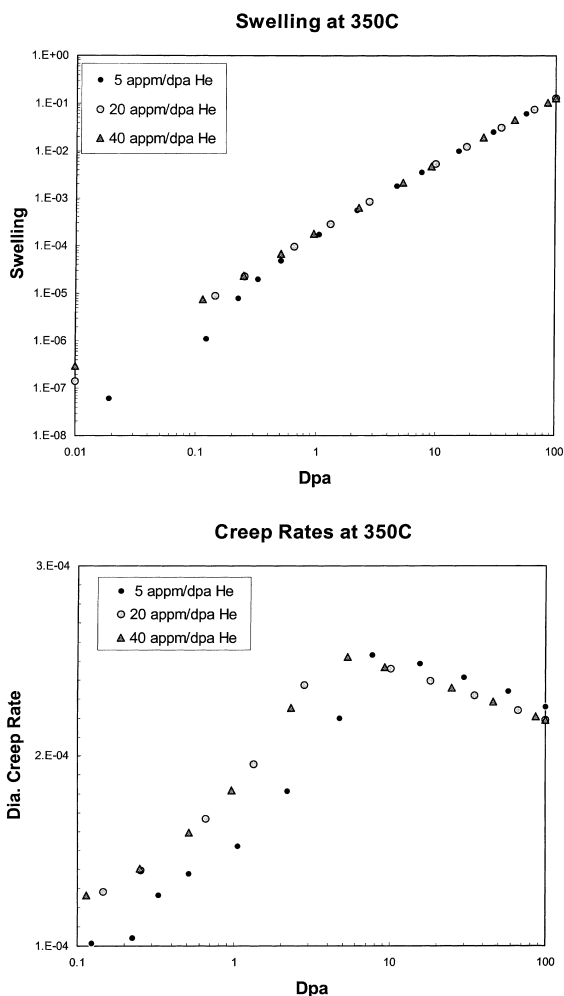


Fig. 4. Calculated (a) swelling and (b) creep rate at 350°C for a dose rate of  $10^{-7}$  dpa $^{-1}$  as a function of dose for austenitic stainless steels with a maximum void number density of  $10^{21}$  m $^{-3}$  for different helium generation rates.

in a void under-estimates the effect of the helium generated rate.

#### 4. Conclusion

It indeed appears possible that enhanced rates of transmutant gases and concurrent helium bubble formation not only can accelerate swelling at low dpa levels, but can also accelerate irradiation creep. These effects are largely transient in nature, however, and exert their largest influence at relatively low (<10 dpa) dpa levels. During this period, the creep rate at a high gas generation environment can be a factor 2–3 larger than that in a low gas production environment. The effects are most pronounced when the bubble density is large and the bubbles

are small, conditions found when the temperature is relatively low and the gas generation rates are large.

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