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THE SWELLING BEHAVIOR OF Ti-STABILIZED AUSTENITIC STEELS USED AS STRUCTURAL MATERIALS OF FISSILE SUBASSEMBLIES IN PHENIX

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The swelling behavior of Ti-stabilized austenitic steels used as structural materials of fissile subassemblies in Phénix

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Abstract.

In this paper we analyse the main results obtained on pressurized tubes, fissile pins and hexagonal cans, allowing us to characterize the swelling and irradiation creep resistance of Ti-Mod. austenitic steels, used as reference materials for the fast breeder subassembly. After having compared the global behavior of 316Ti and 15-15Ti steels irradiated as fissile pins we examine in more detail the leading variables acting on swelling and irradiation creep resistance of CW 316Ti clads and wrappers.

The irradiation creep associated to the principal mechanical stresses (sodium pressure for the wrapper, fission gas pressure for the clad) explain the plastic deformation observed on the wrappers not on the clads.

Fissile pins swell more and the scatter of the results is larger than for wrappers or samples. It does not seem possible to invoque flux or primary stress differences to explain this fact. On the opposite the thermal gradient in the thickness of the components appears to be a significant parameter. In fissile pins it gives rise to a swelling gradient observed by electron microscopy that must be taken into account when comparing to the wrapper.

As compared to CW 316Ti, CW 15-15Ti is an important improvement since its incubation dose for swelling is far beyond 100 dpa. Further more since it swelling temperature dependence does not seem to be as important as for 316Ti, it should be less sensitive to the effect of thermal gradients.

KEYWORDS : neutron irradiation, Ti-Mod. Austenitic Steel, swelling, irradiation creep, fuel pin, hexagonal can, thermal gradient, swelling gradient.

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1. INTRODUCTION.

Titanium stabilized austenitic steels are actually the reference structural materials of the fuel subassemblies of the French Fast Breeder reactors Phenix and Superphenix. Two variants are in use : 316Ti and 15-15Ti for clads and 316Ti for wrappers. These materials are only used in the cold worked condition in order to minimise their dimensional changes during irradiation. For clads these deformations are essentially due to swelling whereas in the case of wrappers, both swelling and irradiation creep associated with the sodium pressure in the subassemblies must be taken into account. As described in another paper (1) the swelling of these structures also governs their mechanical behavior.

A good knowledge of this property is thus essential in this class of materials ; that is why a great number of investigations are performed not only on the subassemblies, but also on specimens irradiated in capsules so as to have a better control of the different variables affecting swelling and irradiation creep.

In this paper we will analyse the main results in this field for doses up to 100 dpa*. After having compared the global behavior of 316Ti and 15-15Ti steels irradiated in fuel elements we will examine in more detail the leading variables acting on swelling and irradiation creep resistance of CW 316Ti clads and wrappers.

2. GLOBAL BEHAVIOR OF CW 316Ti AND 15-15Ti.

2.1. The different structural materials of the fast breeder fuel element.

Fig. 1 compares the performance of the different cladding materials in use in Phenix in the cold worked condition. This figure emphasizes the gain achieved when moving from unstabilized 316 to 316Ti and 15-15Ti. Let us recall that from a 316 basis with 17% Cr, 14% Ni, 2.5% Mo and about 0.4% Si and 0.05% C, the two stabilized versions are obtained by adding 0.4% Ti either on the initial 316 (316Ti) or on a slightly different base with 15% Cr, 15% Ni, 1.2% Mo and about 0.4% Si, 0.1% C (15-15Ti).

In this family of CW austenitic steels the stabilization improves substantially the irradiation behavior specially at high temperature where titanium clearly stabilizes cold work microstructure. This extends to the high temperature range the beneficial effect of CW on the different properties before or under irradiation. As we will see on Fig. 2 the effect is quite drastic on the high temperature swelling which was a limiting factor for unstabilized 316 (2, 3).

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* The dpa are here NRT dpa.

The fig. 1 shows that with CW 316Ti cladding and specially CW 15-15Ti we can hope to go above the actual burn up which in Phenix represents about 100 dpa. Some standard fuel subassemblies have actually reached much higher doses without any problem. The record in dose for a Phenix subassembly (135 dpa) has been reached with a 316Ti cladding (**Boitix 3** subassembly). As for a CW 15-15Ti it has been irradiated as standard in Phenix at a dose close to 130 dpa (**Nestor 3** subassembly) without excessive deformation.

This good behavior of 15-15Ti achieved in Phenix is an excellent confirmation of the results already obtained in Rapsodie in a large irradiation program including reirradiation of fissile standard pins started in 1975 with the **Viking** program and ended in 1982 with the shut down of the reactor. A that time the dose achieved on **Drakkar 1bis** subassembly with CW 15-15 Ti clad pins was already slightly above 130 dpa. 15-15Ti is also the reference cladding material for Superphenix 2nd re-loading.

Fig. 1 gives rise to other remarks. The 3 alloys presented here exhibit differentiated behaviors due to large differences in their average incubation dose for swelling (major phenomena in pin deformation). These are around 45 dpa for CW 316, 95 dpa for CW 316Ti and well beyond 100 dpa for 15-15Ti. But the dispersion band for the behavior of each of these materials is large, since, due to the important value of the swelling rate beyond incubation,small fluctuations in the swelling take off can induce significant differences in deformation. Thus for a given dose close to incubation, at 100 dpa in 316Ti for instance deformations range from 1.5 to 6% approximatively. This dispersion is related both to metallurgical variability and differences in irradiation condition.

The metallurgical origine not being completely understood, we will focus our talk on the effect of irradiation variables analysing the dispersion within a single fabrication lot.

2.2. General caracteristics of pin deformation.

Fig. 2 illustrates the difference in behavior between 316Ti and 15-15Ti irradiated as fuel pin clad in identical conditions to 90 dpa approximatively. One can see that there is a factor of two difference in the maximum deformation of these two clads. At this dose the maximum value lies around 500°C and 450°C for 316Ti and 15-15Ti respectively. It also exhibits a quasi absence of deformation in the upper part of the fissile colum above 550°C. In Phenix this high temperature stability remains at higher doses on both stabilized alloys presented here.

The next figure explicits the effects of dose and temperature on CW 316Ti. Both clads have been irradiated in the same outer core subassembly but with different irradiation conditions associated to the flux gradient in this type of subassembly. Let us note that the temperature regime is different for the two pins 610°C max for one of them and 580°C for the colder one. The maximum deformation lies between 480°C and 500°C since it is both dose and temperature dependent. A clear temperature dependence is observed since beyond 480°C for similar of even increasing doses deformation falls from its maximum to its minimum within fourty degrees approximatively. The comparison of the two pins at the level where the temperature is 500°C emphasizes the importance of dose since a slight flux difference between the two pins ($\sim$ 2.5% increase) leads to a factor of two on measured deformations.

As all the phenomena exhibiting a threshold (see fig. 1), swelling of austenitic steels can differ widely if dose is or not above incubation. The importance of dose and temperature effect (fig. 3) can easily be explained by a strong dependence of incubation dose with temperature.

2.3. Behavior differences between 316Ti clads and wrappers.

Swelling of wrappers induces flat to flat and length changes as well as bowing susceptible to cause handling problems. It occurs that for 316 steels the maximum allowable doses on wrapper and clads of the same kind are generally compatible.

Further more as shown on Fig. 4a were we have plotted the swelling of the wrapper and clad of a 316Ti subassembly the maximal swelling deformations observed on both components are comparable and lie at neighbouring core levels. This situation is favourable since it limits the interactions between the pins beam and the wrapper.

In fact, despite of evidence this does not mean that clads and wrappers have the same swelling behavior since these two components do not see the same temperature dose distribution and there is no reason for them to have the same axial swelling distribution.

In Fig. 4b we have plotted the same results as in fig. 4a but this time as a fonction of temperature and not core level. We can clearly see that in the 500°C region where clads and wrappers receive the same flux (thus the same dose) the clad swells much more than the wrapper.

Later on in this paper we will develop more extensively this point.

3. PARAMETRIE STUDY OF SWELLING OF SAMPLES, WRAPPERS, AND CLADS IN 316Ti.

We will study in this part the influence of the different irradiation variables on the swelling and irradiation creep of CW 316Ti. Before going in the analysis of clads and wrappers of fissile subassemblies, let us analyse the results obtained on pressurized tubes to appreciate better the part of irradiation creep in the observed deformations and evidence the role of some variable as dose rate or stress.

3.1. Experimental procedure.

The alloys studied are stated in Table 1.

type			chemical composition weight %									cold work
			Cr	Ni	Mo	Mn	Si	Ti	C	P	B	%
316 Ti	S1	sample	16.9	14	2.3	1.7	0.58	0.44	0.057	0.023	0.002	20
	W1	wrapper	17	13.7	2.7	1.7	0.50	0.37	0.047	0.027	0.003	26
	C1 C2	clad	16.7 17.1	13.6 14.1	2.4 2.75	1.45 1.5	0.46 0.49	0.45 0.34	0.066 0.049	0.019 0.015	0.002 0.002	15 à 25

Table 1 : Metallurgical description of the different nuances of 316Ti under consideration.

S1 steel is irradiated as samples, W1 corresponds to a heat present both in Phenix and Superphenix, finaly C1 and C2 represent two different Phenix clad lots.

The samples are irradiated as pressurized tubes of Phenix geometry welded end to end so as from a pin irradiated in an experimental capsule placed in a special subassembly named DIMEP. Working temperatures vary between 400 and 500°C but at both ends the dose rates available are limited. Profilometries and density measurements allow us to separate the contribution of creep and swelling.

On some wrappers full non destructive tests have been performed (longitudinal and transverse planimetries) as well as density measurements at different levels to determine independently swelling deformation (uniform expansion of the hexagon) and irradiation creep under the effect of sodium pressure (deflection of each face). The temperatures are computed ones and correspond at each level to an average during life time of the thermal history of the mean fiber of each wrapper face and each clad section.

The tests performed on the fuel pins are in general profilometry. However density measurements are made on some of them to determine at each dose digression, the part of plastic deformation. In this study we have always used pin profilometry where the plastic deformation has been calculated using different density measurements.

3.2. Stress and dose rate effects on the swelling of the CW 316Ti.

Fig. 5 groups the density measurements performed on pressurized tubes (S1 heat) irradiated up to 100 dpa at temperatures between 450 and 430°C and stresses ranging from 0 to 170 MPa ; one can observe a stress induced swelling (4) which appear to be rather correlated to a reduction of the incubation dose (5) than to a clear increase of steady state swelling rate. However since the used stresses where high, one can see that the effects are limited.

For the same stress of 90 MPa Fig. 5b gives the swelling kinetics obtained for different dose rates by similar temperatures of about 410°C. One can observe as for unstabilized steels (2, 3) a slight increase of incubation dose with dose rate.

3.3. Irradiation creep of 316Ti.

In this paragraph we would like to develop the importance of irradiation creep on the dimensional changes occuring in 316Ti irradiated under stress. To do so we will first analyse the plastic strains observed on pressurized tubes.

It is now well known that the irradiation creep strain ($\varepsilon = \varepsilon_1 + \varepsilon_2$) measured under a constant stress has a contribution which varies linearly with stress $\varepsilon = (\frac{3}{4} K\phi t + \frac{3}{4} S)\sigma$ where K and α are constants, ϕt is the dose, σ the stress and S the swelling, and a contribution with a quadratic variation ($\varepsilon_2 \propto \sigma^2$). In our experiments the irradiation creep strains measured on pressurized tubes for stresses up to 280 MPa exhibit a negligible contribution of the σ^2 term at least up to 150 MPa, stress which is far above the usual stresses in clads and even wrappers in nominal working conditions.

So, we will only be concerned here with the two terms having a linear stress dependence.

Fig. 6a represents the results obtained when selecting the sole low swelling points ($S < 0.2\%$) for which $\varepsilon \backsimeq \frac{3}{4} K\phi t\ \sigma$. One can see that the irradiation creep modulus thus determined has only a significant value for temperatures below 500°C.

When keeping only the results of the high swelling specimens ($S > 2\%$) we can determine the α factor from the slope of the line obtained by plotting $A = \varepsilon/0.75\quad t$ as a function of $S/\ \phi t$. We can see that α is almost temperature independant.

The irradiation creep modulus thus determined allows a good prediction of the plastic deformations due to the Na pressure measured on wrappers but not those measured on fissile pin. Fig. 7b shows that the experimental values are larger that those expected from the stress due to fission gas pressure. The origin of this discrepancy is not get clearly understood, we have thus been conducted to use empirical relations to deduce the clad swelling from profilometries. The relation has been obtained by the analysis of a set of clads where both profilometries and density measurements where available.

3.4. Effect of dose and temperature on the swelling of clads and wrappers.

On Fig. 8 we have plotted for 3 temperatures around the swelling peak of CW 316Ti the swelling of different C2 clad subassemblies as a function of dose (swelling derived from profilometries). At each temperature we observe first an incubation period where the swelling rate increases from a value close to zero up to a steady state one $\dot{S}_0$. Following swelling is linear with $\dot{S}_0$ rate. The transition between those two regimes always occurs around 2% swelling.

The steady state swelling rate does not vary significantly for the different temperatures but the incubation dose increases sharply with temperature as indicated by the evolution of the average dose to get 2% swelling (fig. 9). Thus the average at 2% represents to some extent the average incubation dose at the temperature considered. Around this average, dose dispersion is important and increases with temperature. At 540°C for instance one can note that for C2 clads the 2% swelling value is achieved for doses lying between 86 and 100 dpa approximatively. These incubation dose fluctuations can induce a large swelling differences for a given dose and temperature.

Fig. 8b groups in the same way the wrapper swelling results (measured by immersion density) for lot W1 whose behavior is the closest to C2 clads. The same trends are observed but with a clear shift in incubation dose as shown on fig. 9.

We here confirm the fact that wrappers swell less than clads since their incubation dose is larger. This difference between clads and wrappers increases with temperature. On fig. 8b we have also reported the results obtained on samples S1 for zero pressure, they behave as the wrapper.

We can thus conclude that fissile clads swell more than wrappers or samples who bear no fuel adjacency.

This difference cannot be due to a heat to heat effect since we have checked that a given lot of Phenix tubes behaves differently if it is irradiated as a fissile pin clad or as stress free tubes in a capsule. The differences described on fig. 9 are still found.

4. DISCUSSION.

As we have just noted, the knowledge of the couple dose-temperature only is not enough to describe the swelling behavior of the fuel subassembly.

In particular for a given dose, temperature and fabrication lot, the dispersion of swelling observed on fissile clads and the difference between their behavior and the species irradiated without fuel (wrappers and samples) is not understood. For this last point an eventual adjacency effect could be anticipated, but its role on swelling must still be explicited. Before examining that uncertain hypothesis one must first have a look on the role of the other irradiation parameters not taken into account. Particularly, the dose rate, the stress and the thermal gradient existing in the thickness, differ within the different species and can lead to a new variable influencing the properties of irradiated materials.

In fissile pins the average hoop stress due to fission gases is low, less than 20 MPa at a maximum dose of 100 dpa. In view of the extent of the effects anticipated (see fig.5a) one can rule out the primary stresses as an explanation. By all means, this effect, if even it exists, cannot explain the swelling dispersion of pins situated in a given fuel assembly since they see similar stresses.

This type of dispersion is shown on fig. 10a where we have reported the results obtained at 520°C on C1 clads irradiated in core 2 in the same subassembly. This subassembly is an interesting one since the pins are irradiated in variable neutronic and thermal conditions. In particular the flux gradient involves differences in pin swelling, but we can see that the swelling rate of these pins is much higher than the average rate evolution measured on clads of the same lot irradiated at increasing doses in different subassemblies. Moreover, in this peripheric subassembly, we see that we can find at exactely the same dose swelling values ranging between 1 and 6%.

As displayed on fig. 10b the dispersion observed at identical temperature, dose (here dose rate) and fabrication lot can be correlated to the calculated temperature gradient in the thickness of the clad (ΔT) at each level. As sketched on this figure at constant dose and temperature, we can find (on pins at different maximum temperature) different thermal gradients since the axial linear power distribution (hence ΔT) and dose rate are slightly shifted. Fig. 10b groups the results obtained at two doses 72 and 82 ± 2 dpa and for each one two average temperatures respectively 480°C and 540°C for the first and 520°C and 540°C for the second. We can see that the swelling of fissiles clads increases in general with ΔT. The resulting exaltation of the swelling is particularly clear at the hot end ($T > 500°C$) when the temperature difference between the external fiber is above 35°C ; this effect of the temperature gradients partly explains the observed differences for a given dose and can be misleading when analysing swelling kinetics since in general high doses are linked to high values of ΔT.

The differences between clads on the one side and wrappers and samples on the other can also be partly due to the temperature gradient since only fissile pins exhibit sufficiente values of ΔT.

We have nct invoqued here the dose rate effect. As above (fig. 5b), its influence is limited and would lead to the opposite effect.

The physical origin of the effect of the temperature gradient is not clearly established. Some assumptions can nevertheless be made. Let us consider first the usual effect due to the importance of the temperature dependence of swelling of 316Ti. Let us for example consider a section of the clad whose average temperature is sufficiently high for its swelling to be low at that temperature (for example 540°C and $\phi t < 100$ dpa). If ΔT is sufficient, there will exist in the thickness of the clad a cold region succeptible to swell. In this case the global swelling of the clad will be inevitably larger than a specimen at the same average temperature but with no ΔT present. According to this, the swelling range will be increased toward the high temperatures, but still the importance of the observed effect is not explained. So, one can then invoque a mechanism where swelling is enhanced by the secondary stresses due to the swelling gradient inside the clad. Such swelling gradient has been observed on T.E.M. samples taken from different part in the thickness of a C1 clad irradiated up to the average incubation dose of the CW 316Ti ($\phi t_{max} = 91$ dpa). At this dose figure 11 shows that the swelling gradient increases with temperature to reach a high value at the average temperature of 547°C where local swelling measured by TEM goes from 0 at the inner part to 10% at the outer part of the clad.

This swelling difference induces a high level of secondary stresses.

Although irradiation creep tends to release them, these stresses can in 316Ti reach much higher values than the primary ones and thus have a non negligible effect on swelling.

5. CONCLUSION.

In the fast breeder subassembly CW 316Ti can sustain doses beyond 100 dpa. This analysis of results obtained on pressurized tubes has evidenced the effect of the main irradiation variables on the swelling and irradiation creep deformations of CW 316Ti pins and wrappers. The irradiation creep associated to the principal mechanical stresses (sodium pressure for the wrapper, fission gas pressure for the clad) explain the plastic deformation observed on the wrappers but not on the clads.

We observe an important effect of temperature of the incubation dose for swelling in CW 316Ti. The maximal steady state swelling rate does not exceed 0.5% per dpa whatever the lot. Fissile pins swell more and the scatter of the results is larger than for wrappers or samples. It does not seem possible to invoque flux or primary stress differences to explain this fact. On the opposite the thermal gradient in the thickness of the components can be important for materials which present an incubation dose increasing very much with temperature (as 316Ti). Thus, in a fissile pin the temperature difference between the internal and external fiber gives rise to a swelling gradient observed by electron microscopy that must be taken into account when comparing different species.

As compared to CW 316Ti, CW 15-15Ti is an important improvement since its incubation dose for swelling is far beyond 100 dpa. Further more since its swelling temperature dependence seems not to be as important as for 316Ti, it should be less sensitive to the effect of thermal gradients.

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Figure captions.

Fig. 1 : Performance of the different Phenix cladding materials.

Fig. 2 : Comparison of the behavior of the two Ti-Mod. SS used as reference material for cladding.

Fig. 3 : Deformation observed on two fuel pins (C2, CW 316Ti) irradiated in a same outer core subassembly.

Fig. 4 : Swelling differences between C2 clad and W1 wrapper irradiated in the same subassembly.

Fig. 5 : Effect of stress and dose-rate observed on pressurized tubes of S1 316Ti.

Fig. 6 : Characterization of the irradiation creep coefficients obtained with the pressurized tubes data.

Fig. 7 : Characterization of the plastic deformation observed on an experimental W1 wrapper (thinned to increase the plastic deformation) and standard C2 fuel pin.

Fig. 8a and 8b : Effect of temperature on the swelling parameters of the CW 316Ti.

Fig. 9 : Effect of temperature on the swelling incubation period of the CW 316Ti.

Fig.10 : Swelling gradient and isodose dispersion observed on the fuel pins of the same outer core subassembly irradiated up to 94 dpa (fig.10a). Effect of the thermal gradient in the thickness of clads on the isodose swelling observed on the fuel pins of the figure 10a (fig.10b).

Fig.11 : Swelling gradient observed par TEM in the thickness of a C1 clad irradiated up to 91 dpa in the Phenix central core.

Table 1 : Metallurgical description of the different nuances of 316Ti under consideration.

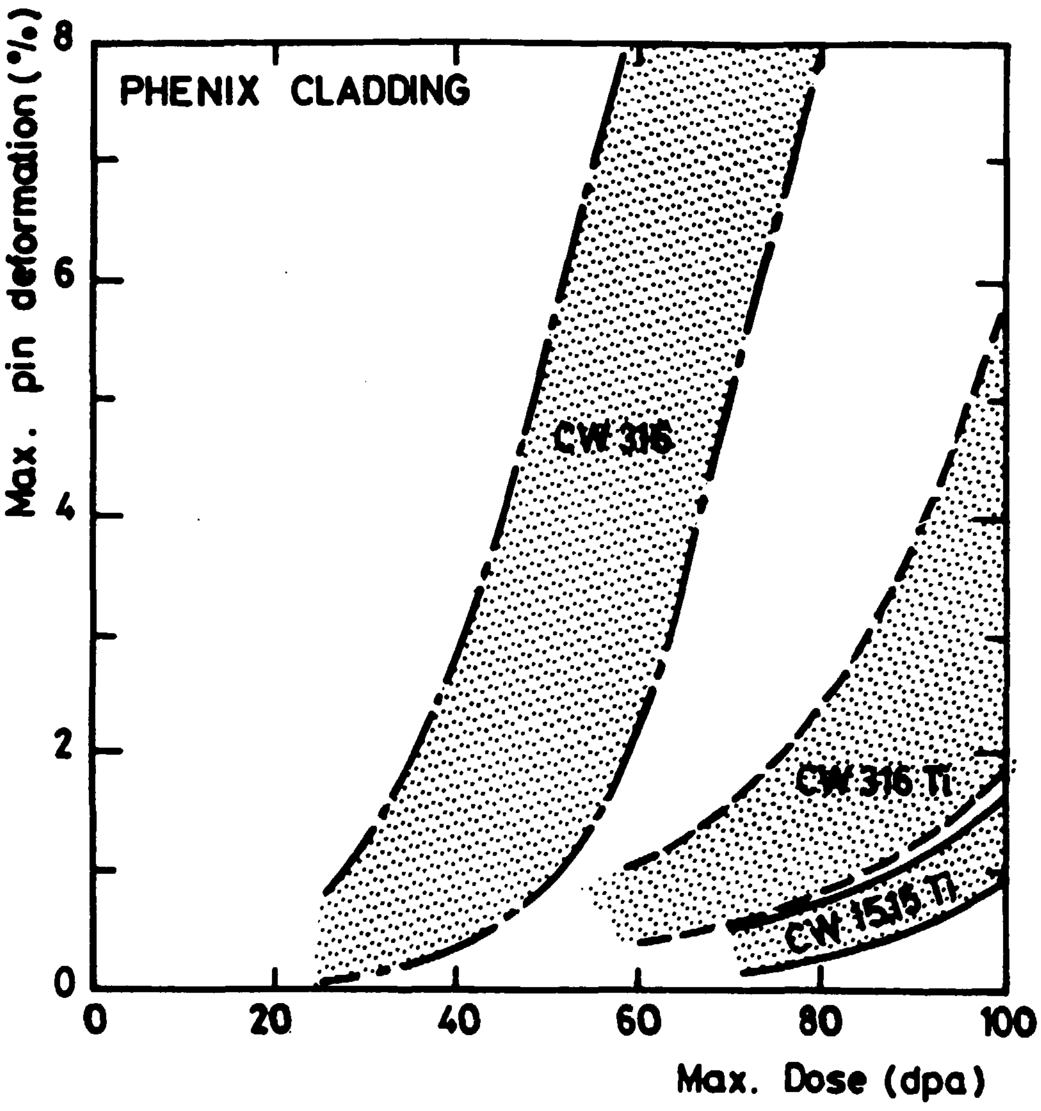

FIG. 1 : Performance of the different Phenix cladding materials.

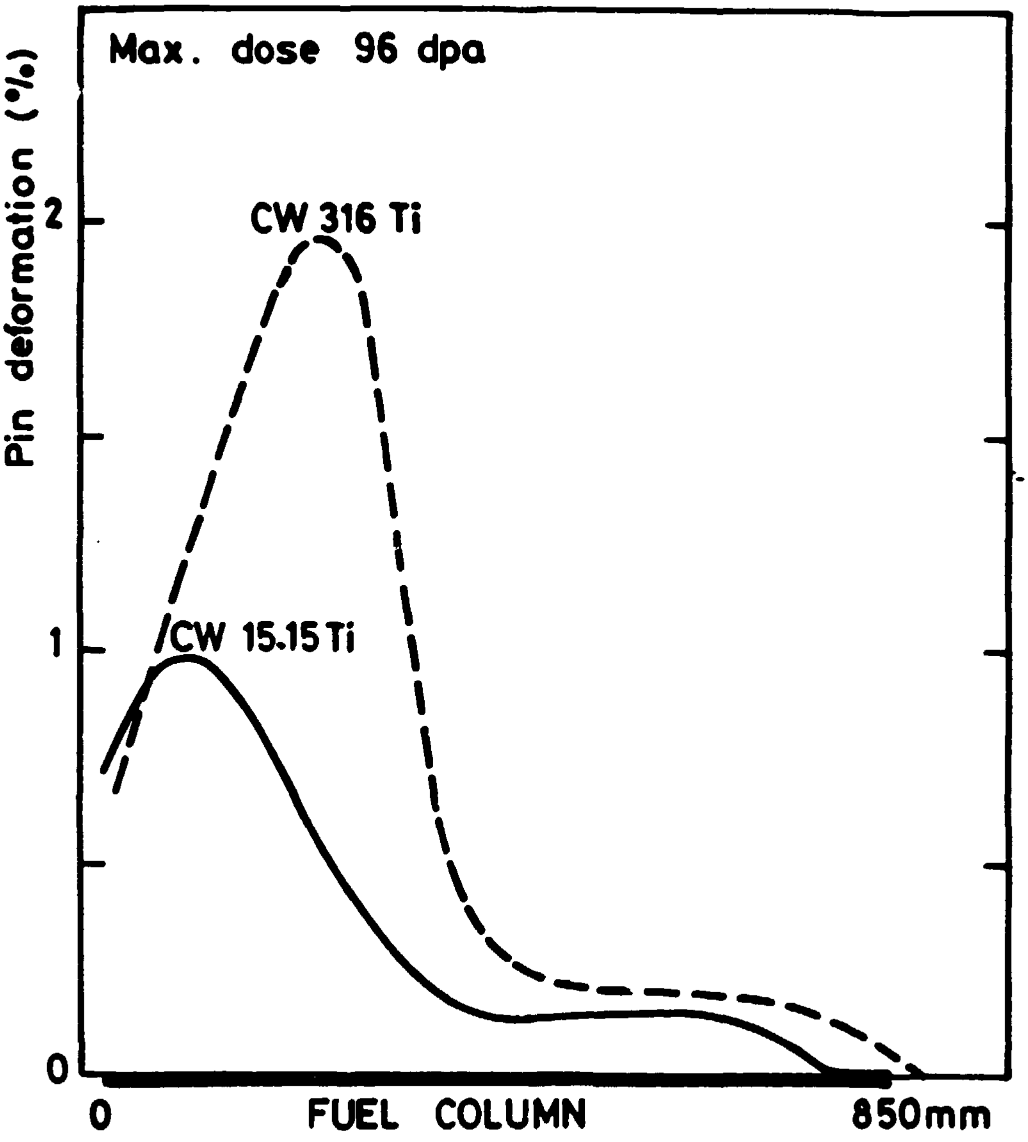

FIG. 2 : Comparison of the behavior of the two Ti-Mod. SS used as reference material for cladding.

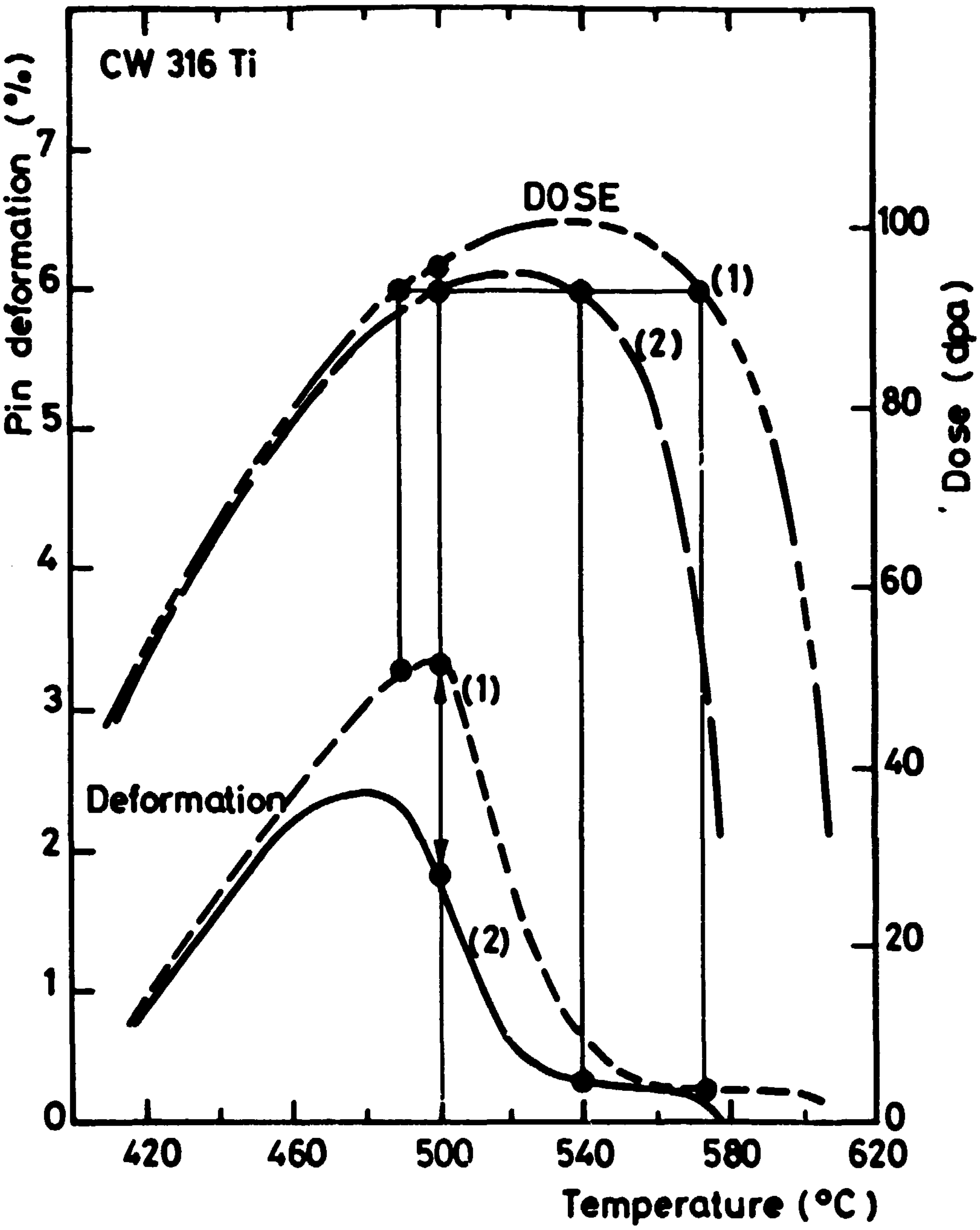

FIG. 3 : Deformation observed on two fuel pins (C2, CW 316Ti) irradiated in a same outer core subassembly.

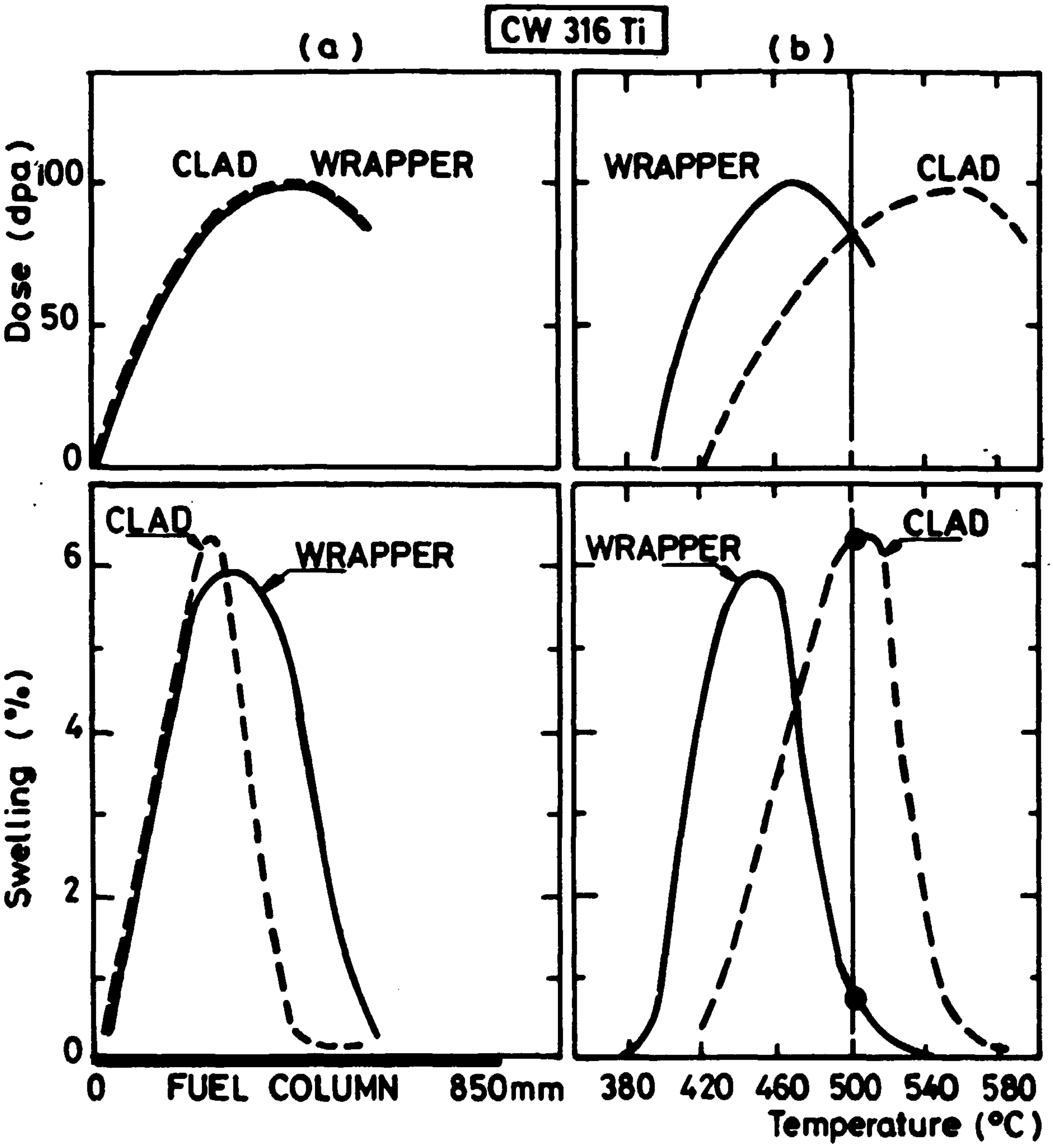

FIG. 4 : Swelling differences between C2 clad and W1 wrapper irradiated in the same subassembly.

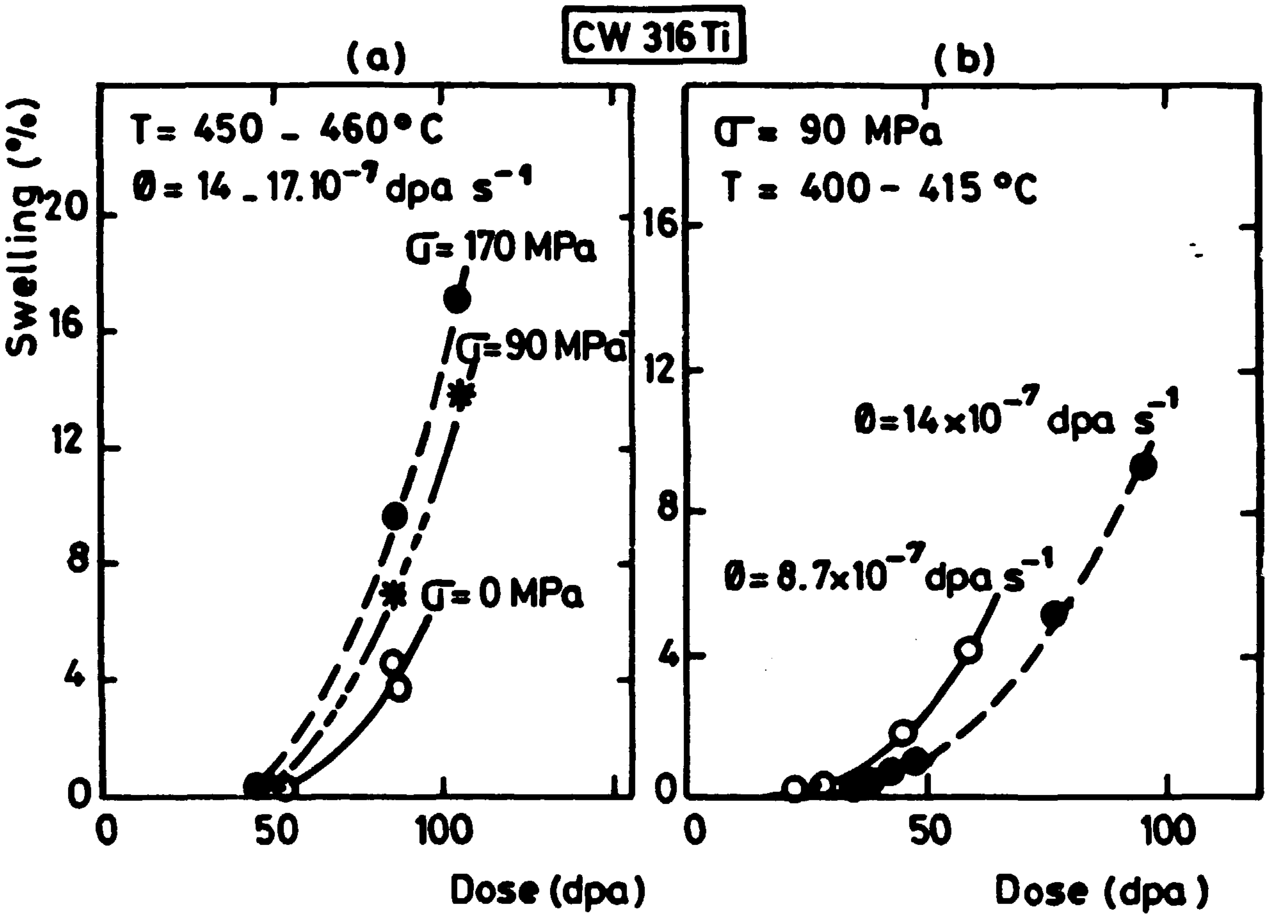

FIG. 5 : Effect of stress and dose-rate observed on pressurized tubes of SI 316Ti.

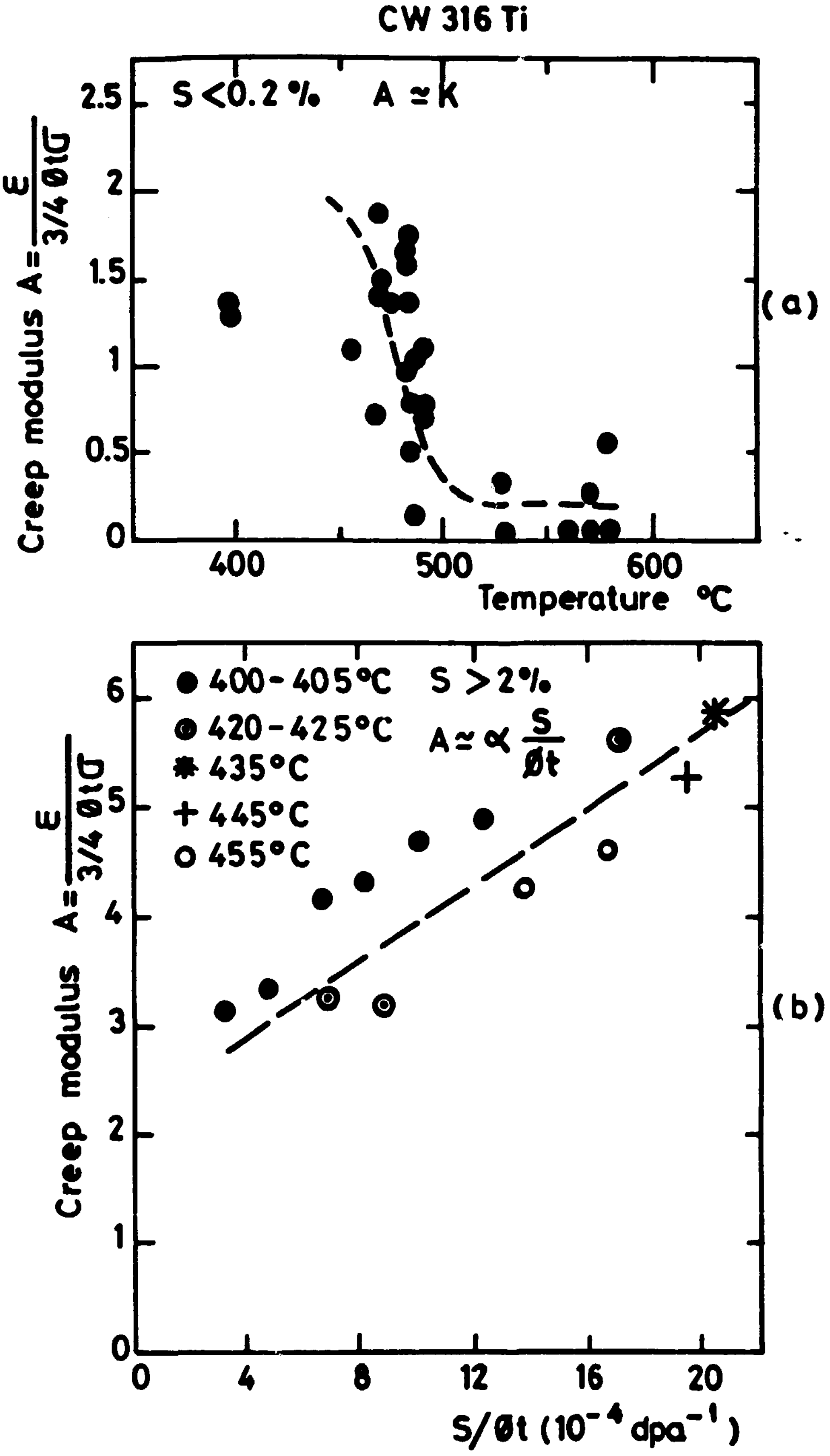

FIG.6 : Characterization of the irradiation creep coefficients obtained with the pressurized tubes data.

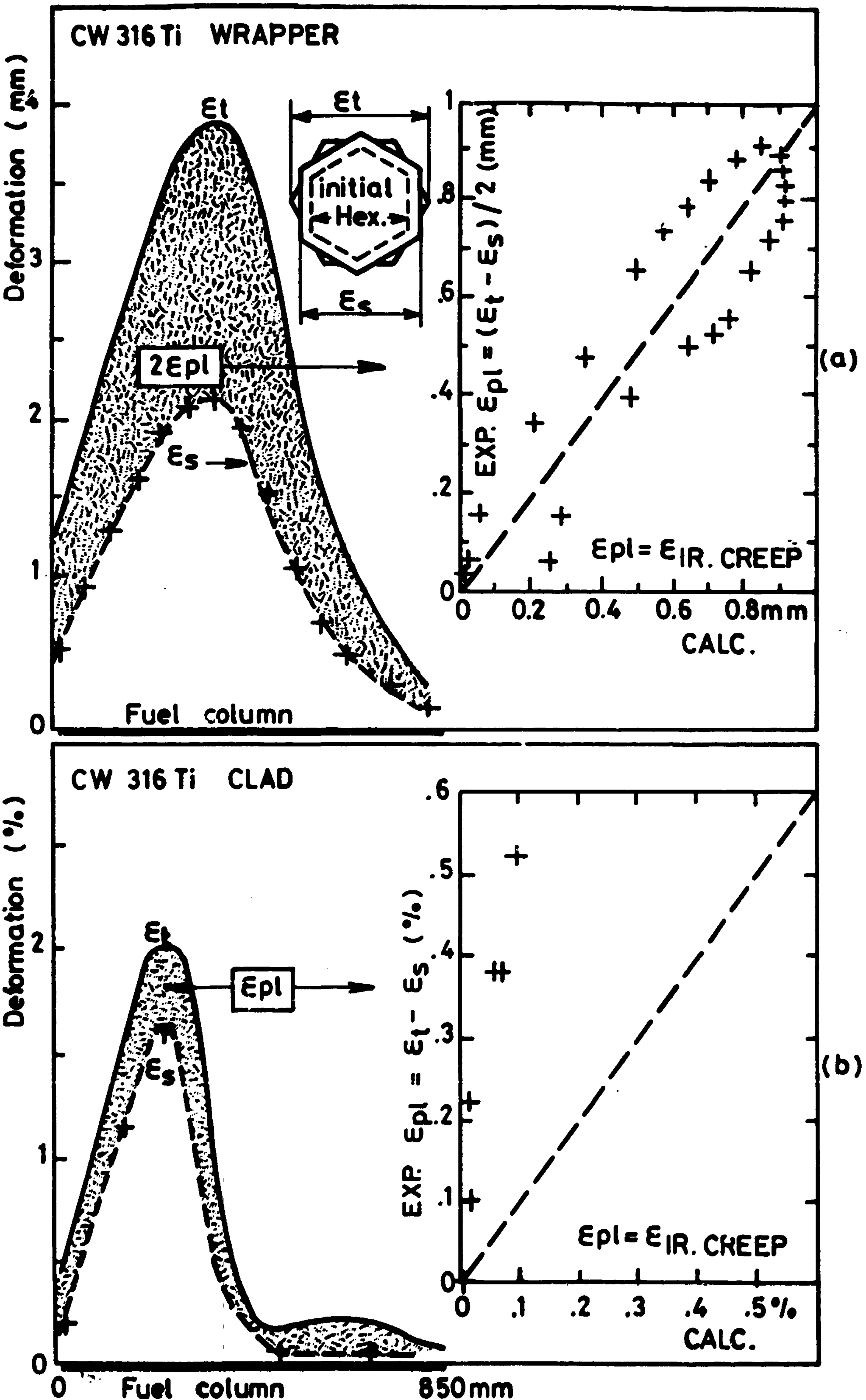

FIG.7 : Characterization of the plastic deformation observed on an experimental WI

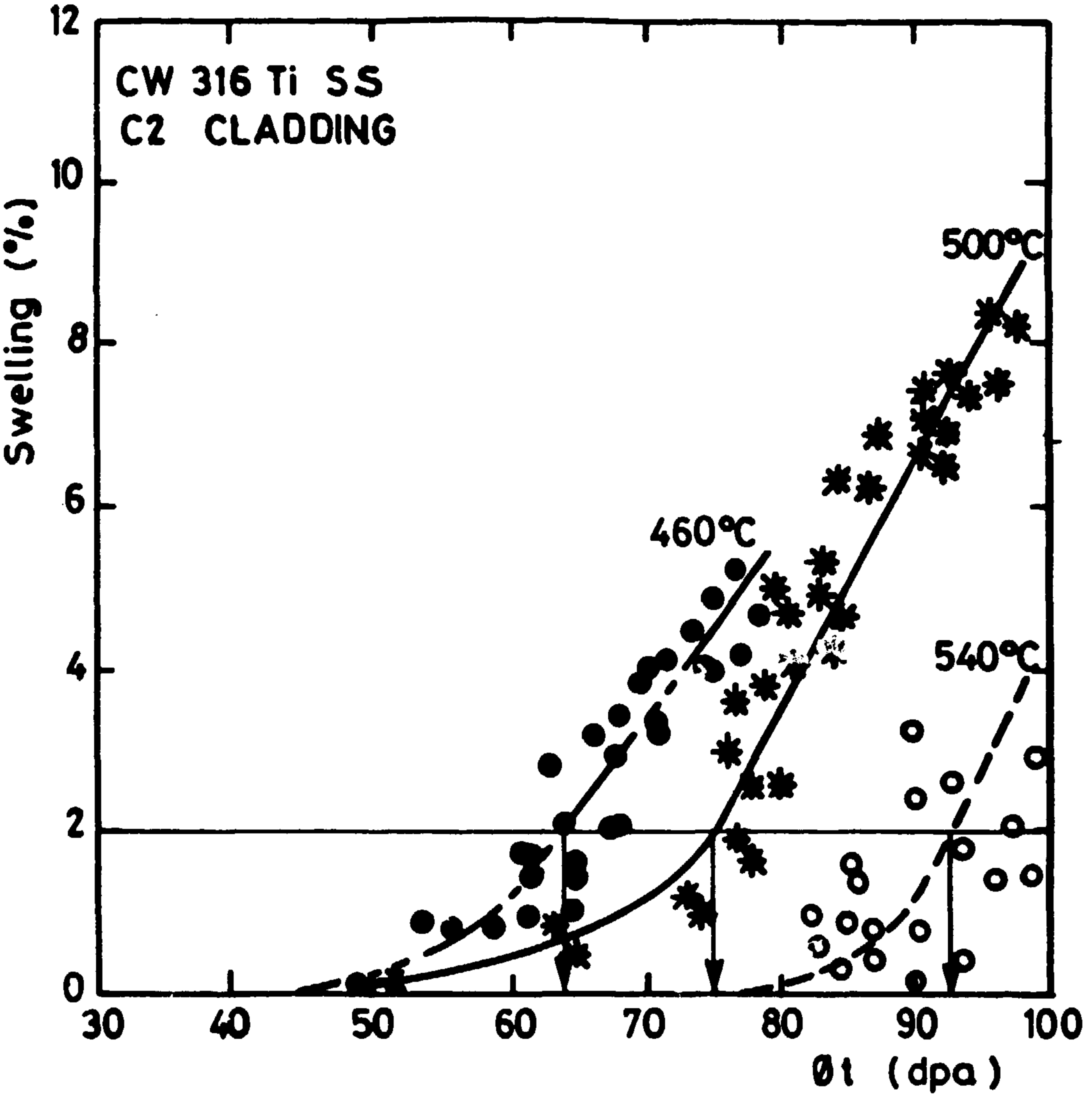

FIG.8a : Effect of temperature on the swelling parameters of the CW 316Ti

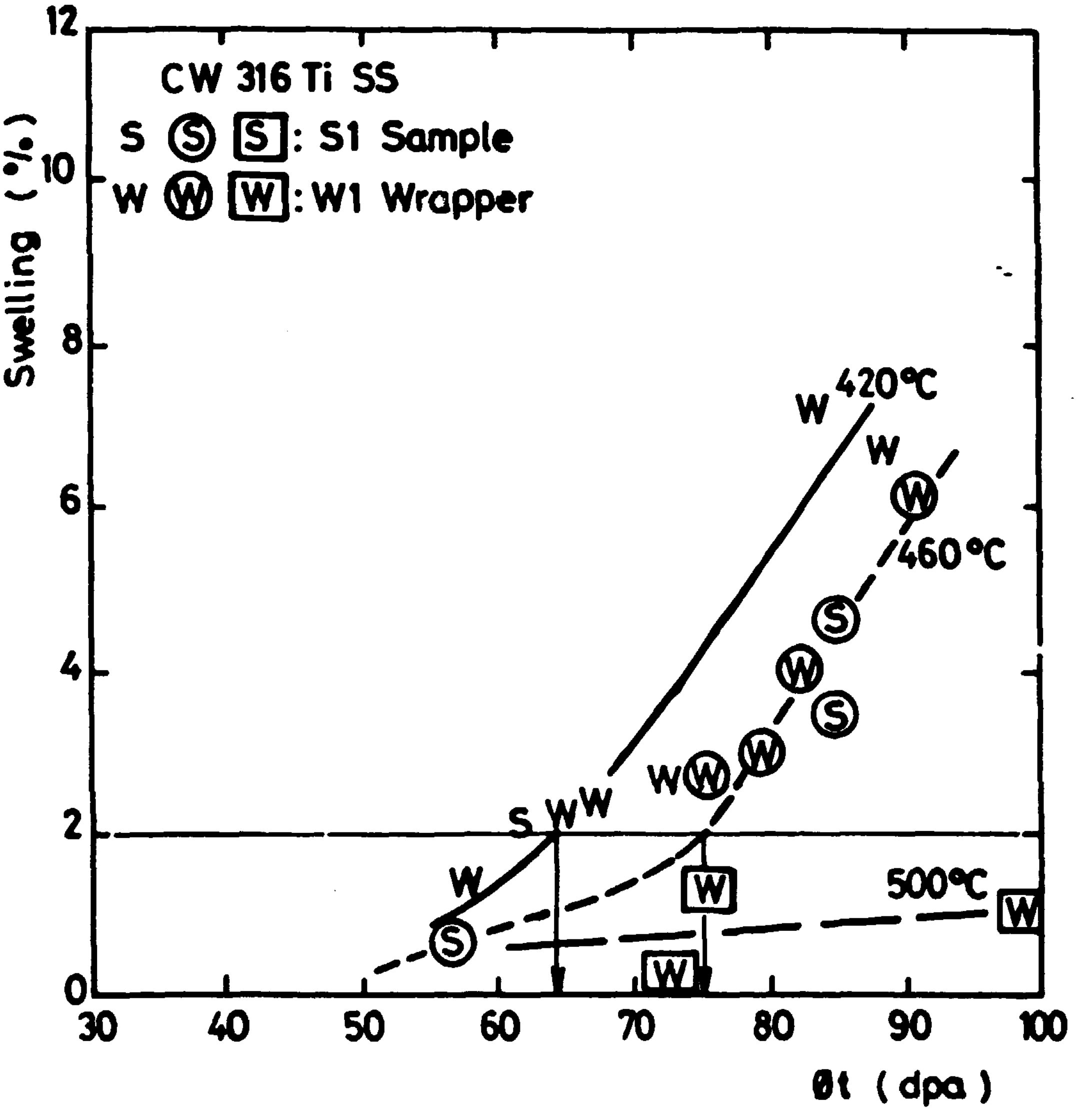

FIG.8b : Effect of temperature on the swelling parameters of the CW 316Ti.

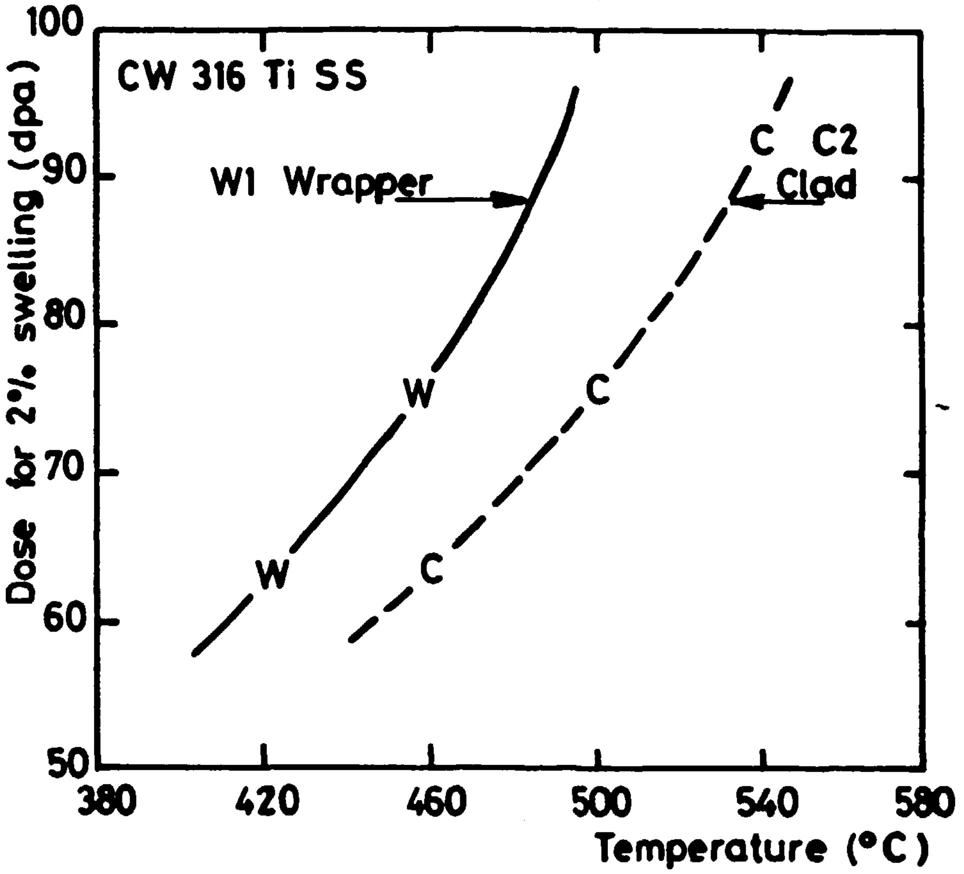

FIG.9 : Effect of temperature on the swelling incubation period of the CW 316Ti.

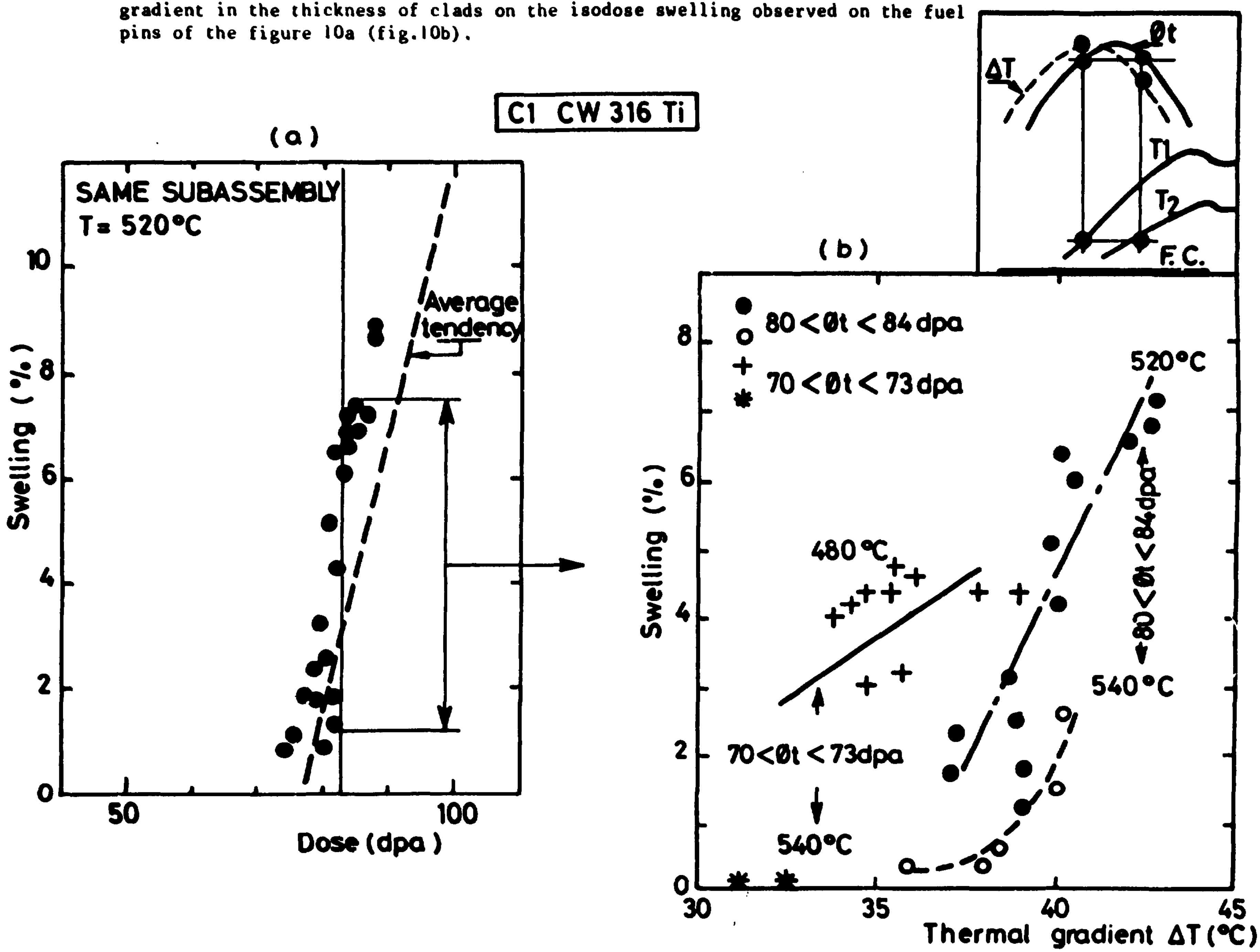

FIG.10 : Swelling gradient, and isodose dispersion observed on the fuel pins of the same outer core subassembly irradiated up to 94 dpa (fig.10a). Effect of the thermal gradient in the thickness of clads on the isodose swelling observed on the fuel pins of the figure 10a (fig.10b).

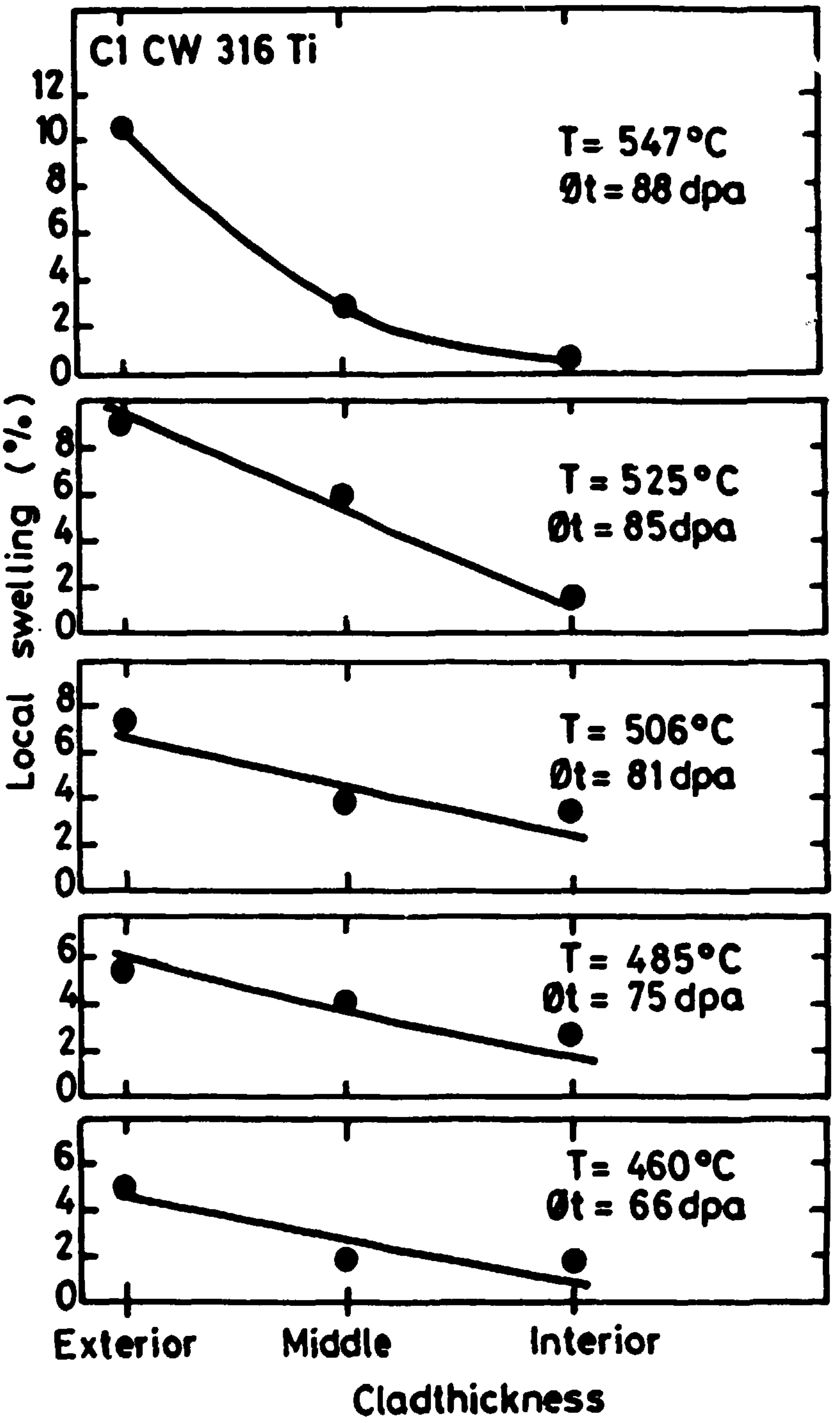

FIG.11 : Swelling gradient observed par TEM in the thickness of a C1 clad irradiated up to 91 dpa in the Phenix central core.