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EFFECT OF STRUCTURE AND ALLOYING ELEMENTS ON VOID FORMATION IN AUSTENITIC STEELS AND NICKEL ALLOYS

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INTRODUCTION

One of the major problems in the development of a fast breeder reactor is the phenomenom of metal swelling due to the formation of radiation induced voids. In this field we are faced with two problems : provide estimates of the swelling that must be taken into account in the design of power reactors and develop alloys with very low swelling rates. To cope with these problems on a reasonable time scale a great deal of work has been developped using high flux ion or electron beams to reach the displacement dose levels involved in a power reactor. However the use of either tool for rapid simulation of the fast neutron environment requires the knowledge of experimental and theoretical correlations between the swelling caused by neutrons and the swelling caused by other particles. In the case of pure metals a thorough investigation of the mechanisms involved in void swelling has led the Physical Metallurgy research group of the Commissariat à l'Energie Atomique to elaborate a simulation model which reproduces reasonably well the experimental swelling versus temperature curves for all the irradiating particles if dislocation densities are available ; fig. 1 gives the published results for the swelling of copper (1) (2). The extension of this model to stainless steel is still difficult since one must take into account the evolution of both structure and composition of these alloys during irradiation. Furthermore, in these complex alloys small fluctuations in composition can have a considerable effect on swelling and a great deal of investigation on the effect of both major and minor alloying elements is still needed to be able to predict swelling. To provide more insight to these problems a research program, involving irradiation of both commercial or specially cast alloys by 500 keV Ni^+ ions or 1 MeV electrons, has been developped ; we will outline here the major results.

I - VOID NUCLEATION IN STEELS

A good knowledge of the basic parameters involved in void nucleation has been achieved through experiments performed on pure metals. To try and separate the points who car be extended to steels and those involving strong differences we have repeated on steels the experiments that led to major informations for pure metals.

I.1 - Effect of the irradiating particle

We have irradiated in well defined conditions (temperature, flux, dose) and with different types of particles (electrons - self-ions) the same material namely 316 solution treated stainless steel.
The results are in all aspects similar to those quoted on pure metals (2) (3) (4) and are reported on fig. 2 a, b. One can notice that voids are observed in electron irradiated 316 ST down to temperatures of 350°C whereas one must reach 500°C (to see voids in the Ni^+ bombarded material. Furthermore at a given dose the overall swelling achieved by electrons is much higher than the one achieved with self ions.

I.2 - Effect of gases

In the case of steels it is very difficult to outgas the alloy without altering the composition fluctuations. That is why our results only refer to the effect of a gas preinjection. Three points can be made out of these experiments :

- For a given temperature range no voids are formed in some alloys as Inconel 706 or incolloy 825 probably because of gas trapping or a low gas content. However if 10 ppm helium is injected with a cyclotron prior to irradiation, swelling occurs.

- Both soluble and insoluble gases affect void nucleation. In all the steels and nickel alloys examined, void number always increases with gas content.

- Gas effect is temperature dependent ; an increase in gas concentration always shifts the temperature of maximum swelling to higher temperatures

and increases the void domain. Fig. 3 illustrates this effect for incolloy 825.
Here again the results are in general agreement with those published in pure metals (4) (5).

I.3 - Effect of dislocations

In pure metals voids are seen to nucleate preferentially in the compression side of dislocations with an edge component (6) (7). Both grown in and radiation induced dislocations are involved in the void nucleation process and there is a good correlation between void number and the total dislocation density (2). In the case of steels the problem seems more complex and has been investigated in detail in 1 MeV electron irradiated 316 steels.

The analysis of swelling versus temperature in electron irradiated 316 S T shows that two temperature domains must be considered (fig. 2b) :

- below 570°C the results are reproducible ; in agreement with theory (fig. 4), the maximum swelling corresponds to the maximum electron beam flux (maximum dose), void number increases with dose rate and void size with dose.

- above 570°C, on the contrary, a great dispersion of swelling values is observed ; for instance at 620°C swelling rates range between 0.1 and 0.6 per cent per dpa. In most cases an anomalous distribution of swelling is observed through the electron beam section (fig. 5). This the result of a heterogeneous distribution of voids ; void size only behaves normally with dose.

A careful analysis of the microstructure clearly shows that in the early stages all voids are nucleated in the vicinity of preexisting dislocations (fig. 6). This void distribution remains up to doses as high as 40 dpa although the radiation induced dislocation network is established in the first dpa ; it is only after 40 to 50 dpa that a new void population sometimes occur ; this population accounts in fact only for less than 10 % of the measured overall swelling at high doses.
Recording the dislocation structure prior to irradiation, a correlation between void number and the preexisting dislocation density has been established (fig. 7), it extends to cold worked material.

These results clearly show that in steels an important difference exists between preexisting and radiation induced dislocations, especially for the nucleation process. This effect is probably related to segregation of impurities mainly gas atoms. At the high temperatures involved, void nucleation is difficult and the large gas concentration needed to stabilize the void embryo is probably only found segregated near the preexisting dislocations. This idea is confirmed by the fact that if 10 ppm helium is preinjected in the steel no effect of preexisting dislocations occurs and a random distribution of voids takes place (fig. 8).

I.4 - Discussion

All the work performed on steels point out that as in the case of pure metals the temperature range of swelling is nucleation limited and that gases play a major role. Thus one can reasonably think that the models worked out to explain the final configuration of vacancy clustering in pure metals (1) (2) can be completely extended to steels and the same assumptions used to simulate void nucleation in swelling calculations. However our results point out that the treatment of dislocations in the nucleation model must be modified to take into account segregation effects and that more understanding is still needed in this field to elaborate previsional models.

II - EFFECT OF COMPOSITION ON VOID SWELLING IN STEELS AND NICKEL ALLOYS

It is well established that both major and minor alloying elements affect swelling, and a considerable amount of experimental data has already been accumulated on the swelling of both commercial and simple Fe-Ni-Cr alloys. Most of these results have been reviewed and analysed in a paper by W.G. Johnston and al (8).

We will report here the results of some experiments performed either to understand the behaviour of reactor component materials, or to provide a guidance for how to modify swelling resistance of steels, and serve as a screening tool for materials that will be thoroughly tested in a reactor.

Both commercial and specially cast alloys have been used in this work performed with 500 keV Ni^{+} ions or 1 MeV electrons. In most cases no helium was preinjected in the alloys tested. Swelling was measured by transmission electron microscopy.

II.1 - Effect of major alloying elements

All the results obtained in this field are in good agreement with the results reported previously (8) (9) (10).

II.1.1 - Effect of nickel content

On fig. 9 we have plotted, versus nickel content, the swelling rate of different commercial alloys and three simple alloys, irradiated a 600°C with 1 MeV electrons. This temperature is not far from the swelling peak temperature except for incolloy 825 and inconel 706 were it is shifted to 520*.

As already stated by Harries (9) Watkins and Standring (10) and Johnston and al (8) swelling drops rapidly with increasing nickel content and presents a minimum around 40-50 % nickel in commercial alloys.

II.1.2 - Effect of chromium content

As observed by Johnston and al (8) swelling increases with chromium content. Fig. 10 illustrates this effect for a simple alloy. One must note that the increase of swelling is essentially due to an increase of void size.

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* One must however note that the swelling tendencies of the alloys are not affected by this shift.

II.2 - Effect of minor alloying elements

The effect of minor alloying elements has been investigated by adding dif: .rent amounts of a single element to a given base alloy. Both substitutional (Ti, Mo, Nb, Cu...) and interstitial (C...) elements have been added to base alloys ranging from 316 S T stainless steels to high nickel alloys.

Some of the results are given on table I and II. The analysis of these results leads to the following main informations.

1. Among the elements added to 316 solution treated stainless steel, titanium causes the most appreciable reduction of swelling; for molybdenum the inhibition of swelling is limited wereas, only a small effect can be evidenced for copper for concentrations above 1 %. As for carbon it is most effective at high concentrations or high temperatures and seems to inhibit swelling only if it is in solid solution. That is why any heating prior to irradiations at temperatures where carbide precipitations occurs leads to an increase of swelling.

2. The variation of swelling with titanium content in 316 S T and 16 Cr 40 Ni (fig. 11) alloys indicates as already stated by Johnston and al (8), that in most cases the dependence of swelling on minor element concentration is of the form.

$$\Delta V(Ci) = \Delta V(0) \ \exp. \ (- K\ Ci)$$

Where ΔV (Ci) is the swelling of an alloy with concentration Ci of minor element and ΔV (0) is the swelling of the base alloy and K a constant for the system.

3. The effect of a given element depends very much of the base alloy. Fig. 11 describes the effect of Ti on swelling in 3 different alloys assuming the functional dependence on concentration stated above. On can see that 0.5 % titanium reduces swelling by about two orders of magnitude in 316 S T stainless steel and only by a factor two in a 22 Cr 33 Ni alloy. This difference can be due to the effect of combined additions since we have shown that combined additions of Ti and Mo, Ti and Nb for instance have a stronger inhibiting effect on swelling of

some alloys than a simple combination of the individual effects would suggest ; an identical observation has been demonstrated for silicon and Titanium by Johnston and al (8) and suggested by the results of Bloom and al in 316 steels (11).

4. The inhibition of swelling can be temperature dependent. Fig. 12 illustrates this effect for carbon. As ascribed by Johnston and al (8) the reduction of swelling in this case is more effective at higher temperatures and is probably due to a shift of the swelling peak temperature.

5. The mode of action of the different elements differs widely ; thus carbon in 316 S T reduces only void number , niobium in 16 Cr 40 Ni only void size and titanium void size for low contents and both void size and number a higher concentration levels. This is illustrated in fig. 13 for Ti and Nb in the 16 Cr 40 Ni alloy.

One can anticipate that the elements that act only on nucleation are more effective at high temperature when nucleation is difficult as is the case for carbon . Furthermore a gas preinjection must have a considerable effect on swelling on alloys where only nucleation is difficult, this seems to be the case of Incolloy 825 (fig. 3). On the contrary as anticipated when both void size and number are reduced (ex. Incoloy 706) only a small effect of helium on swelling is observed.

6. The alloys in which the most effective swelling inhibition occurs are those where a very fine precipitation takes places during irradiation. This has already been observed in simple alloys as Ni-Si for instance. The exact mechanism involved for swelling inhibition due to radiation induced precipitation as still obscure and more work needs to be done in this field.

CONCLUSION

To conclude, this work has emphasized that :

- in steels the effect of dislocations is complex and that a considerable difference exists between preexisting and radiation induced dislocations probably in relation with segregation effects.

- void swelling is strongly influenced by the relative amounts of Fe, Cr and Ni ; void swelling is minimum for Ni contents of 40 to 50 %.

- the swelling of simple and complex alloys can be reduced by certain minor element additions. The most effective swelling inhibitors are those who lead to a finely dispersed precipitation during irradiation.

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Minor alloying element	Base alloy	Minor element concentration c wt %	$\frac{\Delta V(c)}{\Delta V(c=o)}$	Irradiation conditions
Mo	316 S T stainless steel	1 2 3 4	0.42 0.72 0.39 0.24	500 keV Ni^+ ions T = 600°C 30 dpa
Cu	316 S T stainless steel	0.5 1 2	1 0.2	500 keV Ni^+ ions 600°C 30 dpa
Ti	316 S T stainless steel	0.25 0.44	0.1 0.02	1 MeV electrons 520°C 10 - 40 dpa
	33 Ni 22 Cr	0.5	0.43	1 MeV electrons 600°C 10 - 40 dpa
	40 Ni 16 Cr	0.5 1.17	0.27 0.04	
Nb	40 Ni 16 Cr	3	0.51	id.

TABLE I - EFFECT OF INDIVIDUAL ADDITIONS ON SWELLING SOME BASE ALLOYS.

Carbon content ppm wt.	ΔV/V %	
	T = 600°C	T = 650°C
210	0.271	1.21
270	0.264	0.31
335	0.254	
537	0.05	0.025

TABLE II - EFFECT OF CARBON ON SWELLING OF 316 S T STAINLESS STEEL
500 keV Ni^{+} ions - 30 dpa,

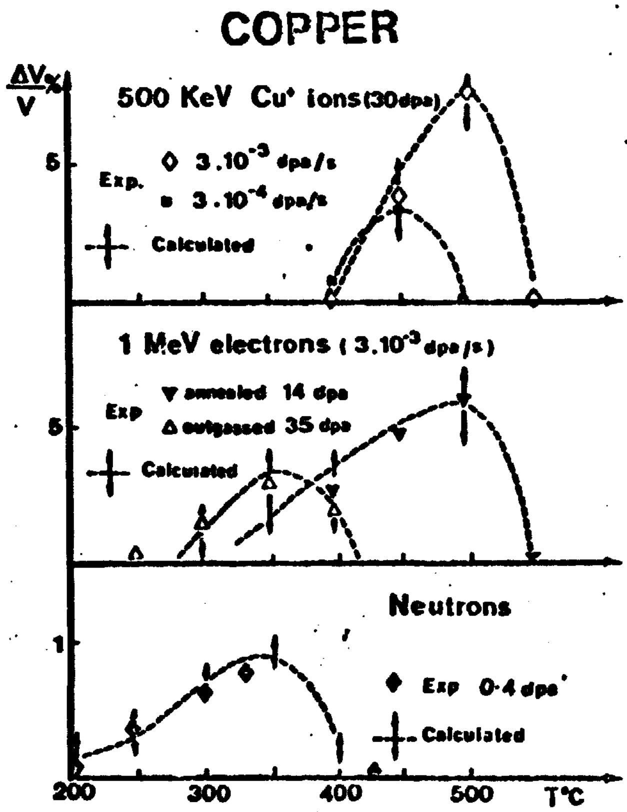


Fig. 1 - Comparison of calculations with experiment

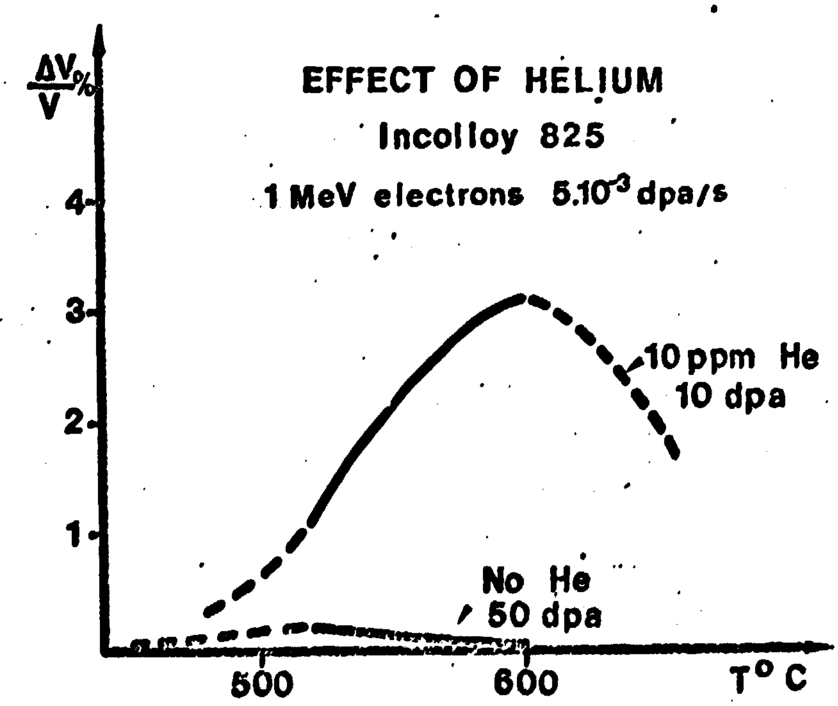


Fig. 3 - Effect of helium preinjection on swelling of Incolloy 825

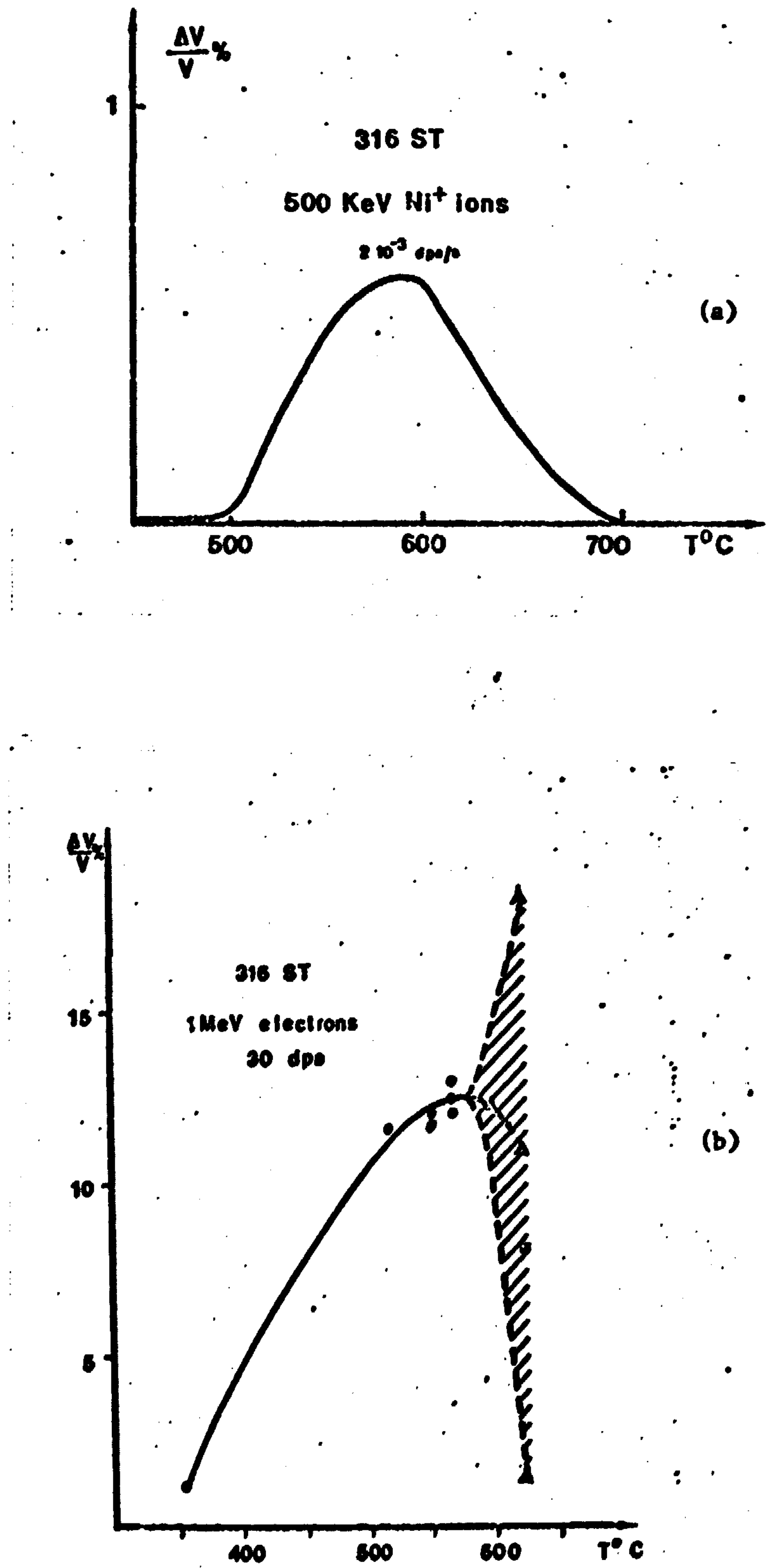
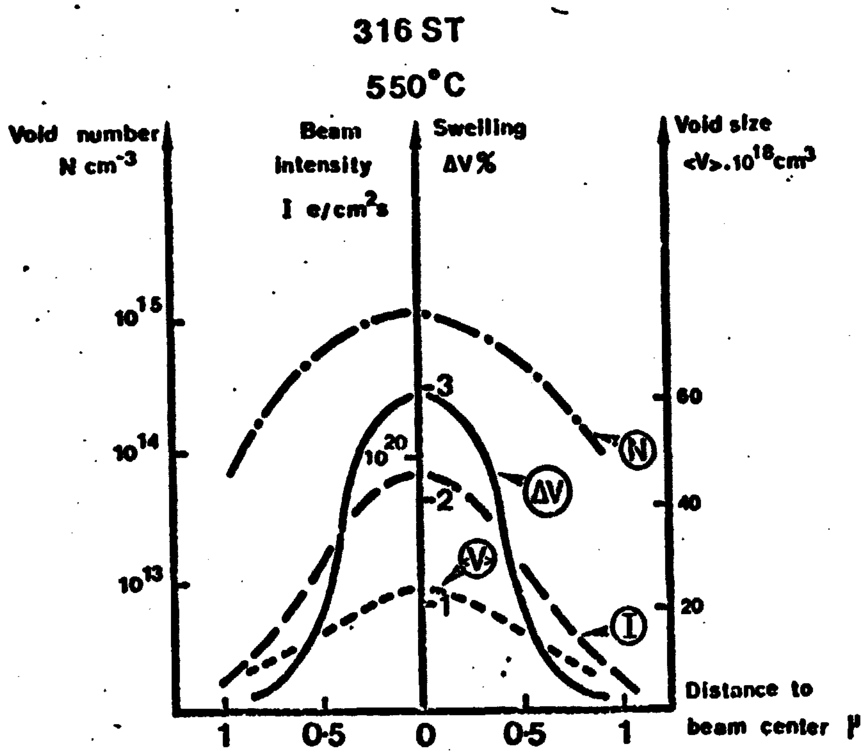
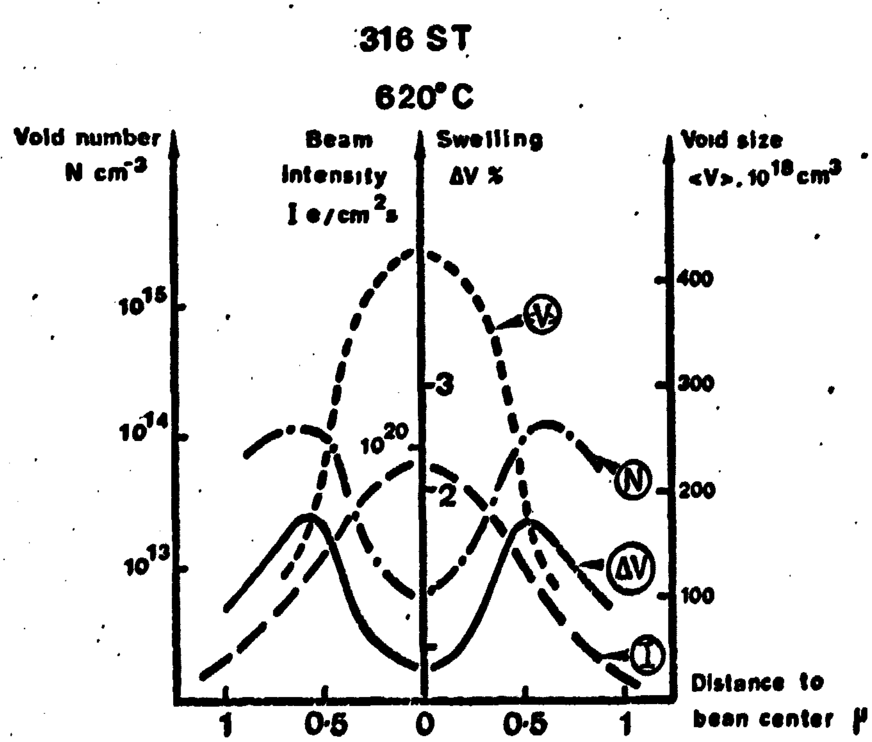


Fig. 2 - Comparison of 500 keV Ni^{+} ion and 1 MeV electron irradiation of 316 S T steel.



<u>Fig. 4</u> - Swelling distribution in the electron beam below 570°C



<u>Fig. 5 -</u> Swelling distribution in the electron beam above 570°C

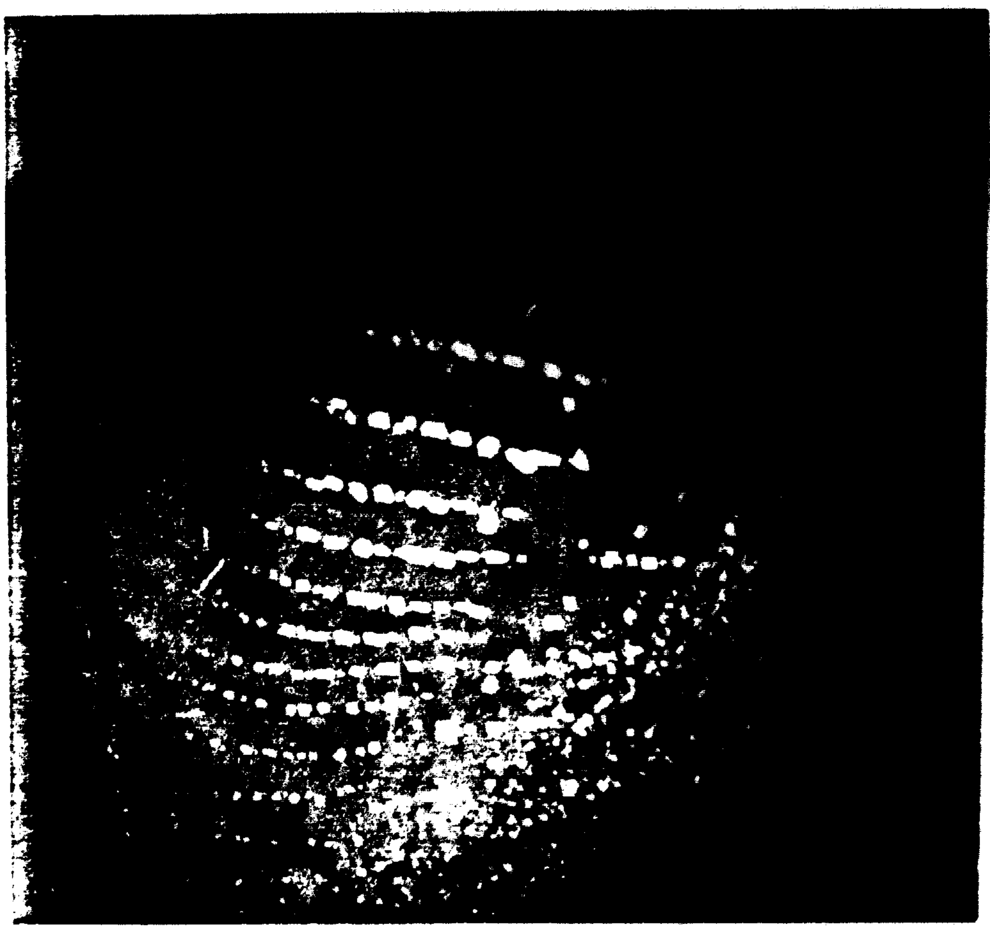


Fig. 6 - 316 ST steel electron irradiated at 620°C
Voids are only seen near preexisting dislocations.

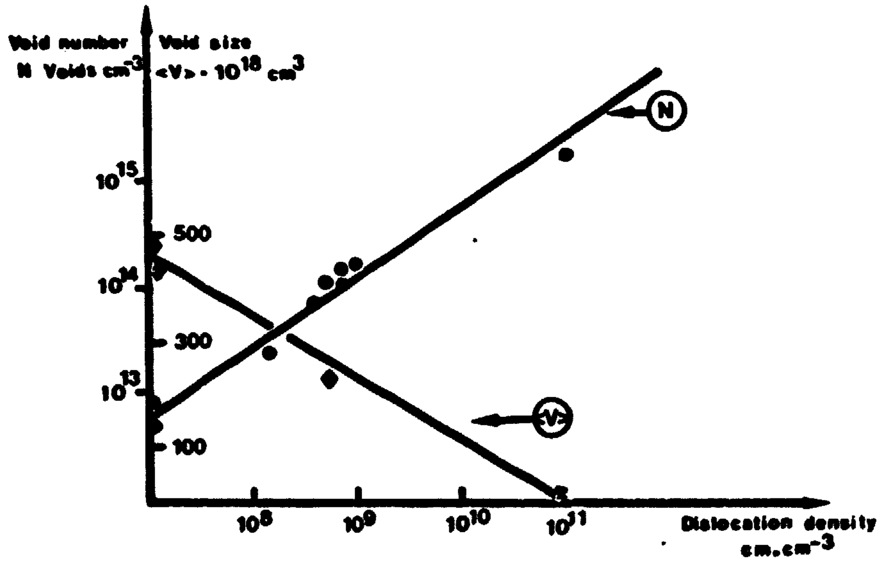


Fig. 7 - Correlation between void number and size and preexisting dislocation density.

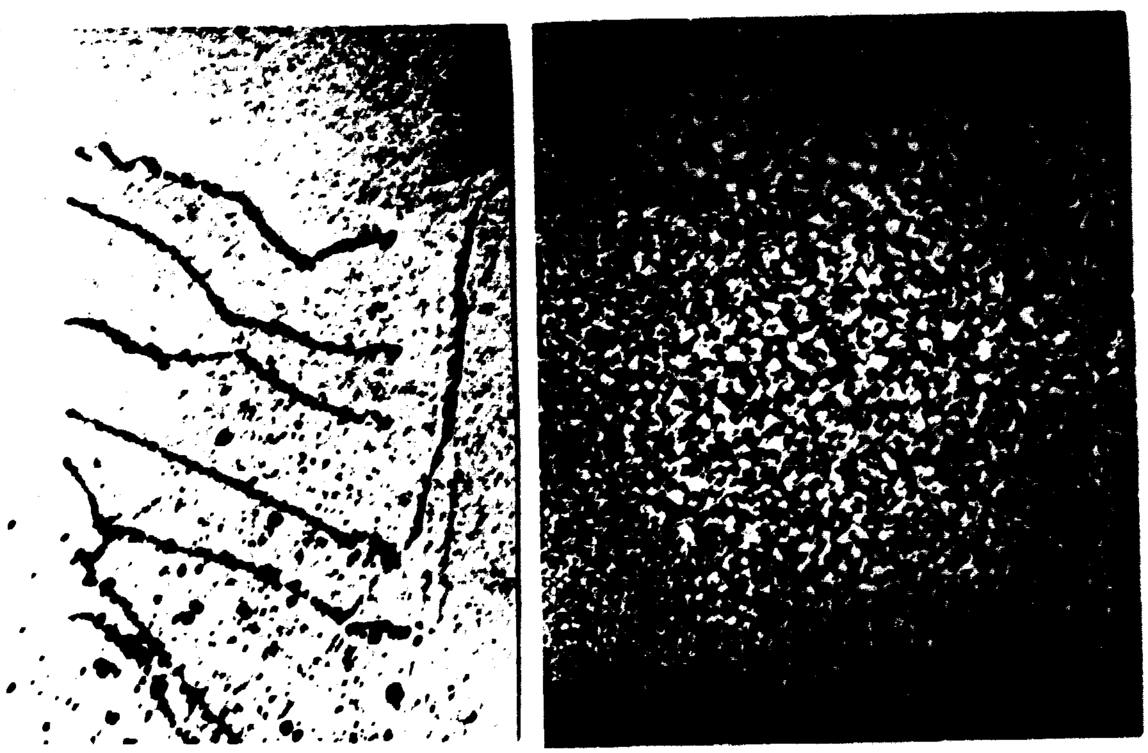


Fig. 8 - 316 S T helium preinjected irradiated at 620°C.

INFLUENCE OF NICKEL CONTENT ON VOID SWELLING

1 MeV electrons 600°C 5.10^{-3} dpa/s

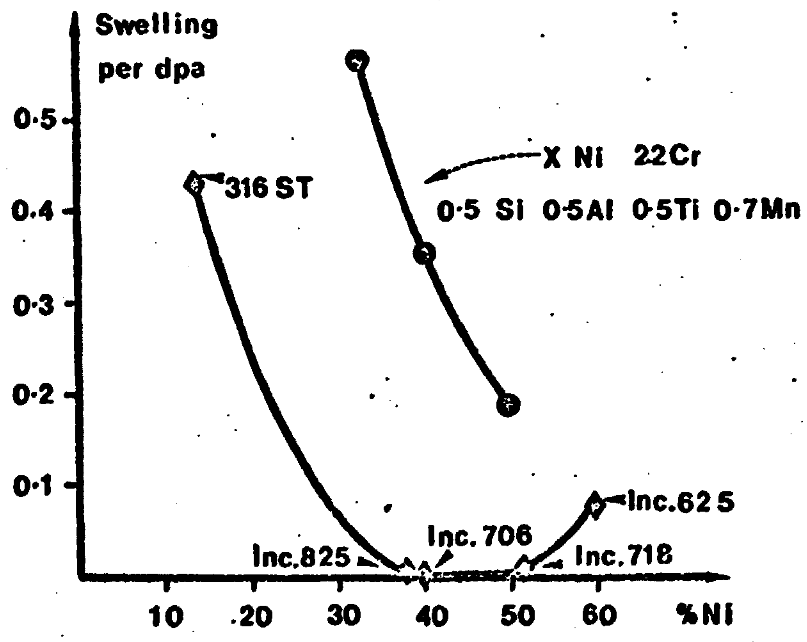


Fig. 9

EFFECT OF CHROMIUM

X Cr 40 Ni 0·5Ti

1 MeV electrons

600°C

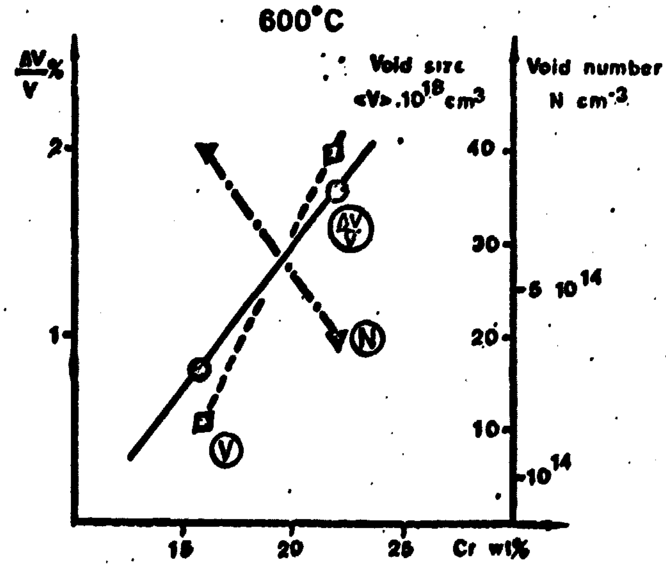


Fig. 10

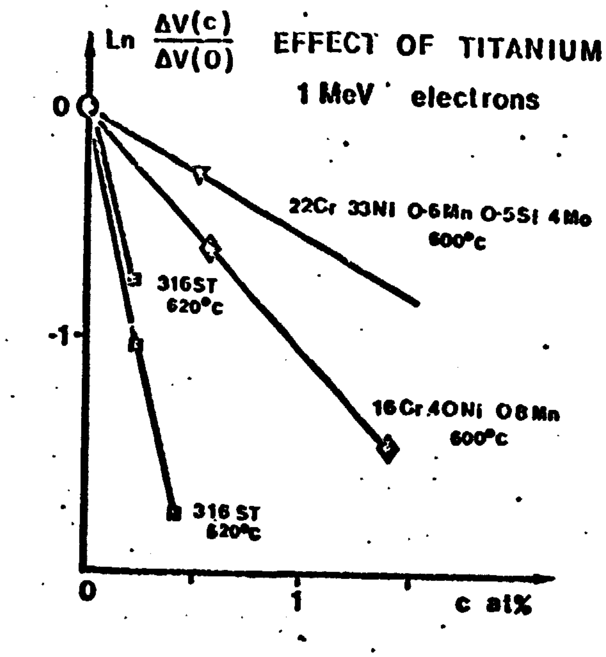


Fig. 11 - Effect of titanium on the swelling of different alloys.

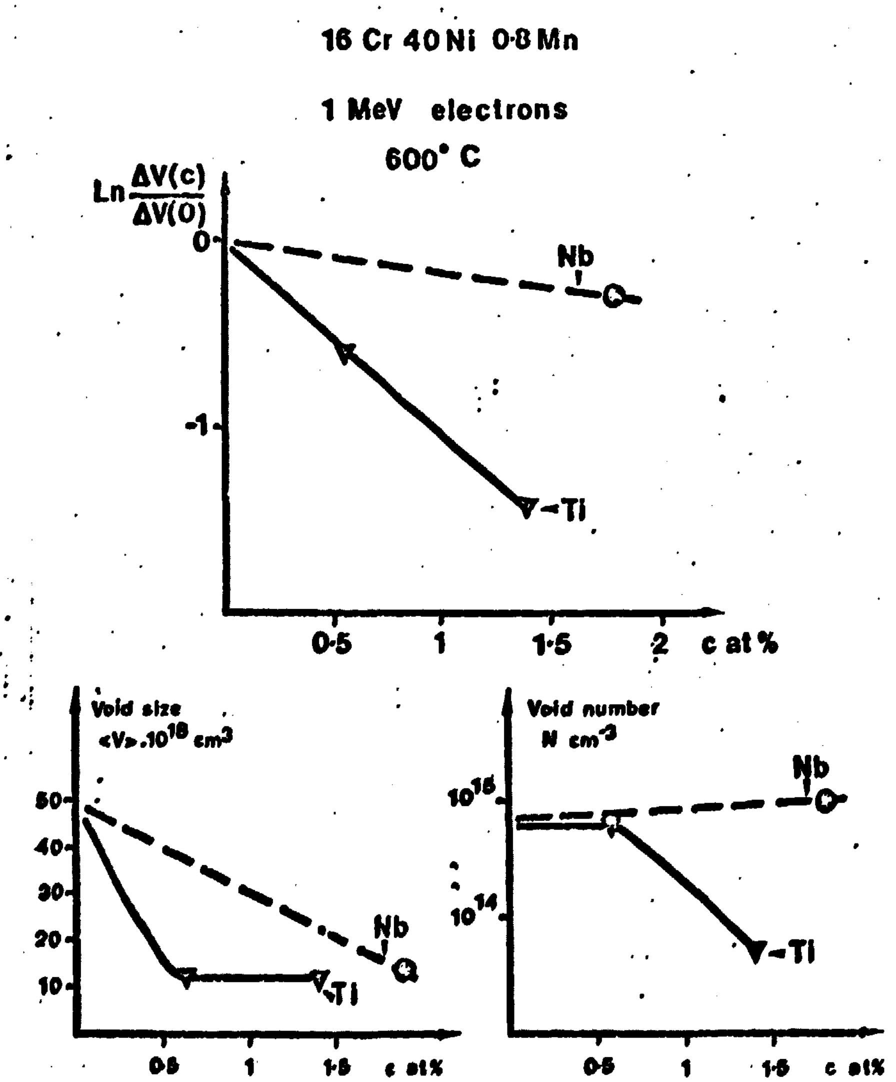


Fig. 13 - Comparison of the effect or Titanium and Niobium on the swelling of a 40 Ni 16 Cr alloy.

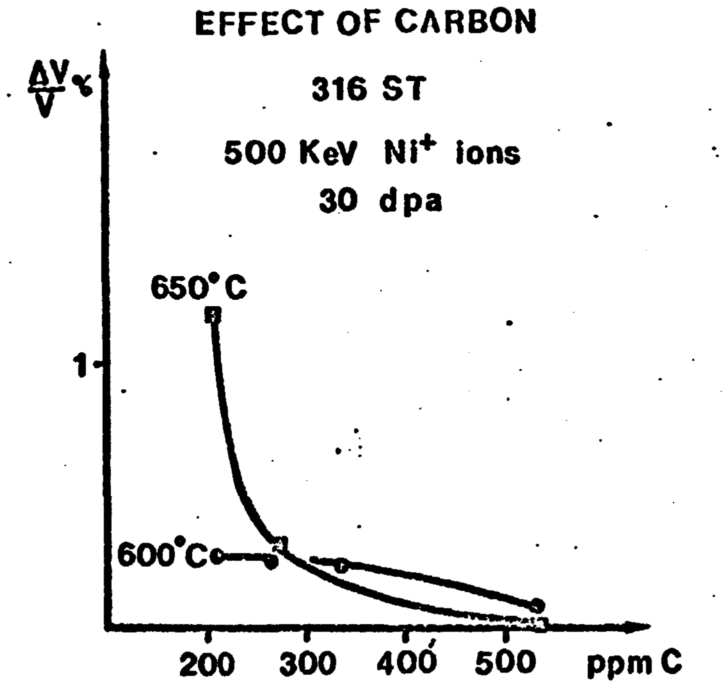


Fig. 12 - Temperature dependence of the effect of carbon.on swelling of 316 steel.