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EVALUATION OF CLADDING ALLOY SWELLING AT HIGH FLUENCES

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**EVALUATION OF CLADDING ALLOY SWELLING
AT HIGH FLUENCES**

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ABSTRACT

Review and analysis of all the available data on the swelling of metals exposed to fast neutrons at high fluence (to $\sim 10^{23}$ n/cm², total) discloses that the mechanisms of void formation and growth are beginning to be understood and that there is real potential for the development of swelling resistant alloys.

Recent results ranging from a volume increase of 10% $\Delta V/V$ in solution annealed Type-347 stainless steel to densification in Inconel-625 are described in detail. Empirical equations describing swelling as a function of neutron fluence and temperature of irradiation are developed in response to the needs of LMFBR core design. Extrapolation of these correlations predicts maximum volume increases of $\sim 15\%$ $\Delta V/V$ for solution annealed Type-321 and cold worked Type-316 compared with $\sim 25\%$ $\Delta V/V$ for solution annealed Type-304 at an LMFBR fluence of 2×10^{23} n/cm², $E > 0.1$ MeV.

It is concluded from data analysis and theoretical treatments that irradiation-induced metal swelling can be substantially controlled by suitable metallurgical pre-treatment of certain alloys to produce finely dispersed second-phase particles or high dislocation densities.

INTRODUCTION

The phenomenon of void formation in metals irradiated in a fast neutron environment was first noticed, together with the attendant volume increase, in stainless steel fuel pin cladding irradiated in the Dounreay Fast Reactor and reported in 1967¹. It was soon realized that volume increases of the extent measured posed new and far reaching problems in the design and performance of fast reactor cores. In the intervening years experimental programs have been started to fully characterize swelling for immediate design needs, and have led to an understanding of the mechanisms of void formation and growth. This effort was also directed toward the objective of attaining swelling resistant materials. These programs have now generated considerable data.

This report reviews the data concerned with stainless steel swelling (by reference only) and describes recent high fluence data which show promising directions for eliminating, or at least limiting, swelling. The use of swelling data to provide engineering information is illustrated by empirical equations which describe the swelling of Types-304 and -321 stainless steel. These equations were developed in response to reactor design needs and are used to predict core component performance in current designs.

1. SUMMARY AND CONCLUSIONS

Recent data on void formation in EBR-II irradiated stainless steels and nickel alloys are described in detail and compared with data obtained in other domestic and foreign experimental programs. A major conclusion is that currently commercially available stainless steel and nickel alloys can be rendered swelling resistant to various degrees by suitable metallurgical pre-treatments. These are cold working of Types-321, -318, -316+Ti, or -316 stainless steel to introduce high dislocation densities, or aging of the nickel-base alloys Inconel-625, or Nimonic PE-16 to produce finely dispersed precipitate particles.

Recent results from immersion density measurements of swelling and transmission electron microscopy (TEM) examinations of void precipitate and dislocation morphology are described in detail. Measured volume changes ranged from 10% increase in volume in solution annealed Type-347 stainless steel to slight densifications in Inconel-625.

In annealed stainless steel it is found that void formation is not continuous as fluence increases; instead, void nucleation ceases at some fluence and thereafter swelling occurs solely by void growth. Comparison of various solution annealed stainless steels showed similar temperature dependencies of swelling although the temperature at which peak swelling occurs can vary.

Possible effects of externally applied stress systems on void swelling are considered in detail. The concept of "stress induced void growth" at high temperatures has not yet been experimentally demonstrated and it is not known whether the effect has a major influence on volumetric swelling. At intermediate cladding temperatures of 500 to 550°C (932 to 1022°F), no effect of stress on void growth was detected, instead strain assisted void nucleation had apparently occurred.

The following equations describing swelling as a function of neutron fluence and temperature of irradiation are developed in response to the needs of LMFBR core design.

(a) Solution annealed Type-321 and cold worked Type-316

$$\% \frac{\Delta V}{V} = 8.9 \times 10^{-2} (\phi t) 1.5 \exp(-3.14 + \frac{5.04 \times 10^3}{T} - \frac{1.66 \times 10^6}{T^2})$$

where ϕt is neutron fluence for $E > 0.1$ MeV $\times 10^{-22}$ n/cm²,
and T is the irradiation temperature in °C.

(b) Solution annealed Type-304

$$\% \frac{\Delta V}{V} = (\phi t) 1.57 \exp(-35.01 + \frac{2.968 \times 10^4}{T} - \frac{6.567 \times 10^6}{T^2})$$

with ϕt and T as in (a) above.

Extrapolation of these correlations predicts maximum volume increases of ~15% $\Delta V/V$ for solution annealed Type-321 and cold worked Type-316 at an LMFBR fluence of 2×10^{23} n/cm², $E > 0.1$ MeV. Whereas at this fluence the maximum swelling of solution annealed Type-304 is calculated to be ~25% $\Delta V/V$. These volume increases are based on the peak swelling occurring at 650°C (1202°F) for solution annealed Type-321 and cold worked Type-316, and 450°C (842°F) for solution annealed Type-304.

The mechanisms of metal swelling are discussed in Section 4. It is pointed out that neutron bombardment of metals in a fast reactor inevitably causes enormous quantities of point defects [vacant lattice sites (vacancies) and interstitial atoms] but that void formation and swelling would not occur unless means were available to preferentially re-combine interstitials and thus cause an excess concentration of vacancies. Subsection 4.1 discusses the initial point defect formation from collision cascades and concludes that precise knowledge of displacement cascade formation is not of primary importance in void nucleation or subsequent void growth.

Experimental data show that void nucleation is substantially enhanced by the presence of impurity gases such as helium, but that the enhancement effect saturates at low impurity levels. During reactor irradiation helium and hydrogen are continuously formed by $n(\alpha)$ and $n(p)$ reactions so that adequate concentrations of gaseous impurities will always be available to assist void nucleation. Thus, given a sufficient vacancy supersaturation, void formation in-reactor is inevitable and swelling can only be inhibited by lowering the excess vacancy concentration through annihilation of point defects by recombination.

The factors causing a point defect imbalance are discussed in subsections 4.2.2 and 4.2.3. Principal causes are considered to be the higher lattice diffusivity of an interstitial rather than a vacancy and the preferential attraction that interstitial and vacancy loops have for interstitials. Thus interstitial loops form and grow readily. By contrast vacancy loops will not grow because their associated strain fields also preferentially attract interstitials which result in mutual annihilation. Only the interstitial loops grow and these rapidly form an irradiation-induced dislocation network. As long as these dislocations are free to move, they continue to absorb more interstitials than vacancies so that an excess vacancy concentration continuously arises and void formation occurs.

Ways to inhibit swelling are discussed in subsection 4.3. They consist of means of providing short-range, unbiased sinks for vacancies and interstitials which limit the formation of a point defect imbalance, lower the overall point defect concentration, and prevent the long range migration and ultimate aggregation of excess vacancies to form voids. The

results described in Section 2 suggest two methods of achieving swelling resistance. One is to provide a sufficiently high concentration of finely dispersed precipitate particles in the alloy matrix—preferably by pre-aging treatments. The particles appear to act as recombination sites for interstitials and vacancies and thus lower the overall point defect concentration, and also to largely prevent dislocation movement so that the production of a vacancy supersaturation is minimized. Alloys behaving in this way are Inconel-625 (subsection 2.2) and the U.K. alloy PE-16. Detailed differences in their irradiation behavior are discussed in subsections 4.2.3 and 4.3. A second method of providing swelling resistance is to form a dense and stable dislocation network in the alloy prior to irradiation. This can be partially achieved by simple cold working as in cold-worked Type-316 (subsection 2.1) but could be improved by pre-aging before cold working to give a dense dislocation network pinned by second phase particles. Critical aspects of this method are that the dislocations must be stable and immobile. As long as they are kept essentially stationary, they cannot accept an excess of interstitial atoms and cause a point defect imbalance. Furthermore, they inhibit long range migration of point defects to other sinks such as grain boundaries so that formation of a vacancy supersaturation is largely prevented.

Further experimental data are required to test these conclusions, particularly at fluences greater than 10^{23} n/cm² and temperatures above 650°C (1202°F).

2. REVIEW OF EXPERIMENTAL DATA

2.1 STAINLESS STEELS

2.1.1 Introduction

A number of reviews of stainless steel swelling are available. Comprelli, et al.,² reviewed the data available up to mid-1969; HEDL workers³ have reviewed the U.S. data up to the Fall of 1970 and Bagley, et al.,⁴ have summarized the British data. Two international conferences held during 1971 contained review papers on various aspects of metal swelling.^{5, 6} In this report it is intended only to review the information necessary to establish trends in overall swelling, void size, number density and roles of microstructural characteristics in the swelling of each alloy. Recent high fluence data are the only ones described in detail.

2.1.2 Type 304

2.1.2.1 Solution Annealed Type-304 Stainless Steel

Solution annealed Type-304 stainless steel is the principal material used for core structural components in EBR-II. For this reason the first U.S. metal swelling data were obtained for this material and at this point in time more data (upwards of 200 data points) are available to describe the swelling of solution annealed Type-304 than any other alloy. The properties of Type-304 stainless steel are such that this alloy is no longer a prime candidate for use as a cladding or core structural material in LMFBRs. Nevertheless, the data for Type-304 are invaluable in illuminating the physical processes involved in metal swelling and in providing guidance concerning the projected swelling behavior of the prime candidate cladding alloys which at this time are Types-321 and -316 stainless steel.

In retrospect it is fortunate that the first large body of data to become available should be for solution annealed Type-304 stainless steel as Type-304 is one of the most simple of the 300 series. The Type-304 components and specimens used to generate the swelling data were generally in the solution annealed condition. The annealing process consists of rapid cooling to room temperature from 1800 to 2100°F (1020 to 1150°C) which retains a fully austenitic structure at room temperature. The resultant structure is simple face centered cubic and single phase in the form of equiaxed and twinned grains. The grain size is determined by the solution annealing temperature and the prior cold work history of the metal. No alloying additions to give solid solution strengthening, stabilization or precipitation hardening have been added. Thus the alloy is essentially single phase and can be considered as such in theoretical treatments. After long times at elevated temperatures some grain boundary $M_{23}C_6$ carbides appear together with occasional small intragranular sigma-phase particles (Fe-Cr intermetallic). The more complex stainless steels such as Type-316 and -347 display more extensive and complex precipitation kinetics. Recent work (described in this report) has shown that second phase particles can influence and even control the swelling characteristics of an alloy. In Type-304 the grain boundary carbides exert little influence on swelling since grain boundaries are denuded of void

formation and the sigma phase is so sparse as to be very much a secondary consideration. Thus the Type-304 gives rise to relatively simple swelling behavior. However, it must be noted that the data exhibit considerable scatter which at this time can be attributed to difficulties in temperature estimation and the secondary effect of variations in composition and metallurgical conditions. This is reinforced by the observation that sets of data are generally consistent in themselves but display divergences when compared with each other.

The Type-304 bulk swelling data show an exponential increase with fluence with volume increases of up to 10% $\Delta V/V$ having been measured to date. A detailed analysis has shown⁷ that this exponential increase in turn increases with temperature. This effect is not wholly clear-cut and a less sophisticated analysis (described in subsection 3.1) still has not revealed this effect clearly. The exponential nature of Type-304 swelling is displayed in Figure 2-1 which is a linear plot of volume increase as a function of fluence at $450 \pm 20^\circ\text{C}$ ($842 \pm 36^\circ\text{F}$).

The temperature dependence of bulk swelling for Type-304 exhibits a bell-shaped curve with peak swelling at $\sim 450^\circ\text{C}$ (842°F) and rapidly approaching zero at 325°C (618°F) and 625°C (1156°F). A transmission electron microscopy examination⁸ has shown that the maximum temperature for any void formation in Type-304 is $650 \pm 15^\circ\text{C}$ ($1202 \pm 27^\circ\text{F}$).

Figure 2-1. Swelling of SA Type 304 at $450 \pm 20^\circ\text{C}$ ($842 \pm 36^\circ\text{F}$)

The average void size increases rapidly with temperature during irradiation while the void number density undergoes a rapid decrease as illustrated in Figures 2-2 and 2-3.⁸ These data are not normalized for fluence. The reason for the data spread in Figure 2-2 is that the GE data from the MT-3 capsule⁸ are from material irradiated to a higher fluence than are the other data. Recent studies^{8,33} have shown that after a certain limiting fluence (which is dependent on alloy type and irradiation temperature), void nucleation essentially ceases and further swelling occurs almost exclusively by void growth.

Figure 2-2. The Temperature Dependence of Void Size in Solution Annealed Type 304 Irradiated in EBR-II

Figure 2-3. The Temperature Dependence of Void Number in SA Type 304 Irradiated to $\sim 5 \times 10^{22} \text{ n/cm}^2$, $E > 0.1 \text{ MeV-GE}$

Harkness and Li⁹ have developed a thermodynamically based model for void formation and growth based on observations of void formation in solution annealed Type-304. The model is a quasi-steady-state approach that attempts to describe void and dislocation loop nucleation and growth rates as a function of temperature, neutron flux, and type and density of point-defect sinks. The model was fitted to experimental observations of void formation by adjustment of material constants "within reasonable limits." The agreement of the model with their own experimental data on void size, number density and bulk swelling is excellent and suggests that their model, which is really semi-empirical, approaches a realistic description of swelling in solution annealed Type-304 stainless steel. The model is based on the hypothesis that some kind of preferential attraction between one type of sink and one type of point defect is necessary for void growth to occur, and the good fit with the data supports the hypothesis. Since neutron bombardment causes equal numbers of vacancy and interstitial point defects, void formation can occur only if an imbalance of point defects exists in some way. Harkness and Li⁹ together with Bullough and Perrin¹⁰ attribute this imbalance to a preferential attraction between the stress field of a dislocation and the misfit strain of an interstitial. This coupled with the ability of interstitial defects to diffuse more rapidly than vacancies means that interstitials are accepted by dislocation lines more readily than vacancies. Thus in the neighborhood of dislocations a high vacancy concentration exists and rapid void growth occurs. This has been demonstrated by Norris¹¹ and Nelson¹² who have obtained motion pictures of void growth in the neighborhood of dislocations in electron irradiated nickel (Norris)¹¹ and cyclotron irradiated (carbon ions) stainless steel (Nelson).¹²

Thus void growth in Type-304 is reasonably well understood. However, the processes of void nucleation are still largely unknown and theoretical treatments in general tend to ignore them and simply assume an empirical dependence of void nucleation on temperature. In Section 4 of this report void nucleation and growth are discussed in some detail.

2.1.2.2 Cold Worked Type-304 Stainless Steel

UK fuel pin data¹³ and more recently HEDL data from materials capsules¹⁴ have shown that cold work can be effective in reducing swelling in Type-316 stainless steel. Few data are available for cold worked Type-304, and those few by contrast show that cold work is relatively ineffective in reducing swelling in Type-304.¹⁴ Indeed at high irradiation temperatures [$T > 600^{\circ}\text{C}$, (1112°F)], cold worked Type-304 appears to swell more than the same material in the solution annealed condition. The HEDL data have been previously tabulated,¹⁴ the present GE-BRD data are listed in Table 2-1. Figure 2-4 is a plot of volume increase as a function of irradiation temperature and shows that the 13% CW GE data are in fair agreement with the HEDL materials specimens irradiated at similar temperatures. The HEDL data¹⁴ are from 50% cold worked Type-304 irradiated as cylindrical specimens in sodium filled materials capsules. The GE data are from encapsulated fuel pin claddings of the F3B series. Two claddings were investigated, 13% cold worked Type-304 and 13% cold worked Type-304 with a subsequent "Garofalo-type" carbide precipitation heat treatment which consisted of 24 hours at 900°F (482°C) followed by 216 hours at 1300°F (704°C) and air cooled.

The HEDL workers were surprised by their observations with 50% cold worked Type-304 since it showed much more swelling than cold worked Type-316 in companion specimens (discussed below in subsection 2.1.3) and also, on occasion, showed more swelling than solution annealed Type-316. Plotting the density change as a function of irradiation temperature (Figure 2-4) showed that starting at the lowest temperature the swelling increased to about 480°C (896°F), decreased to $\sim 580^{\circ}\text{C}$ (1074°F) and then increased through another maximum temperature before finally decreasing to negligible values at the highest irradiation temperatures (above 800°C). It is suggested that the second maximum is associated with cold work recovery. Certainly comparison of data for swelling of cold worked Types-304 and -316 stainless steels (subsection 2.1.3) indicates that the cold worked Type-304 has a greater tendency to swell than does cold worked Type-316 and transmission microscopy indicates that the cold worked structure is more stable in Type-316 than Type-304. Thus in the more complex Type-316 containing molybdenum as a solid solution strengthener and Laves phase former, it is apparently possible to retain a cold worked structure and retain resistance to swelling.

The specimens of 13% cold worked Type-304 fuel cladding show an intermediate degree of swelling between that observed for cold worked Type-316 and solution annealed Types-304 and -316. In the case of the carbide agglomerated material, the final heat treatment appears to have removed the beneficial effect of cold working and the associated swelling resistance and produced a material which swells even more than annealed Type-304—probably by virtue of providing additional void nucleation sites in the form of carbide precipitates.

Table 2-1
IMMERSION DENSITY MEASUREMENTS OF SWELLING IN GE FUEL PIN CLADDINGS

Specimen	Cladding	Axial Location Above Bottom Of EBR-II Core (in.)	Fluence $\times 10^{22}$ $E > 0.1$ MeV	Temperature (°C) (°F)		Density (g.cm ⁻³)	$\Delta\rho / \rho$ (%)
F2G-D	SA 347	2.0-3.0	6.40	487	909	7.4886	-5.49
F2G-H	SA 347	6.25-7.5	7.00	526	980	7.2007	-9.12
F2G-M	SA 347	11.0-12.0	5.50	541	1007	7.5114	-5.21
F2G-P	SA 347	15.0-16.25				7.8803	-0.56
F2G-R	SA 347	29.0-30.25				7.9245	(Control)
F2V-C	SA 316	0.75-2.0	6.15	471	881	7.6354	-3.80
F2V-E	SA 316	3.0-4.25	7.03	495	923	7.3834	-6.96
F2V-F	SA 316	4.25-5.5	7.32	508	946	7.3651	-7.20
F2V-J	SA 316	6.5-7.75	7.32	523	975	7.3031	-7.98
F2V-M	SA 316	9.5-10.75	6.40	534	994	7.4811	-5.74
F2V-P	SA 316	11.75-13.0	5.28	537	998	7.7077	-2.89
F2V-S	SA 316	15.0-16.25				7.9276	-0.10
F2V-U	SA 316	29.0-30.25				7.9369	(Control)
F2X-D	SA 316	1.0-2.25	5.58	460	861	7.7159	-2.78
F2X-J	SA 316	6.25-7.5	6.56	505	941	7.4964	-5.54
F2X-M	SA 316	9.75-11.0	5.60	516	960	7.6770	-3.26
F2X-R	SA 316	15.0-16.25				7.9370	-0.45
F2X-T	SA 316	29.0-30.25				7.9362	(Control)
F2H-4	SA 316	3.6-4.6	3.85	512	954	7.8173	-1.41
F2H-7B	SA 316	6.6-7.8	3.93	536	997	7.8119	-1.47
F2H-7A	SA 316	7.8-9.1	3.77	542	1007	7.8197	-1.38
F2H-8C	SA 316	10.9-11.9	3.10	546	1015	7.8746	-0.68
F2T-8	SA 347	7.1-8.1	3.92	538	1000	7.7235	-2.25
F2T-9	SA 347	8.1-9.3	3.76	543	1009	7.7488	-1.99
F2T-11	SA 347	10.3-11.3	3.27	546	1014	7.7947	-1.35
F3B-2-D2	SA 316	4.0 (midlength)	3.16	468	875	7.9359	-0.34
F3B-2-G	SA 316	5.7 (midlength)	3.26	494	922	7.9472	-0.19
F3B-2-K1	SA 316	11.8 (midlength)	2.52	558	1035	7.9504	-0.15
F3B Control	SA 316		Unirradiated			7.9626	
E1H-26-D	SA 316	1.85-3.35	4.53	505	940	7.8088	-1.96
E1H-26-I	SA 316	7.1-8.85	4.84	573	1065	7.5916	-4.69
EO33	SA 316		Unirradiated			7.9652	
E1H-29-B	13% CW 304	1.8-2.8	4.74	500	930	7.8343	-0.85
E1H-29-H	13% CW 304	10.8-11.8	4.19	593	1100	7.8847	-0.21
H-043	13% CW 304		Unirradiated			7.9014	
E1H-30-B	CW 304 + HT	1.9-2.9	4.71	503	935	7.5401	-4.32
E1H-30-H	CW 304 + HT	10.9-11.9	4.10	593	1100	7.8405	-0.51

Table 2-1 (continued)

H-102	CW 304 + HT						7.8808	
F5M-B	SA 321	2.8-3.8	3.12	527	980	7.8556	-0.52	
F5M-F	SA 321	7.5-8.5	3.21	595	1103	7.8196	-0.98	
F5M-J	SA 321	12.0-13.0	2.32	627	1160	7.8711	-0.32	
21-020	SA 321						7.8966	
F5F-B	SA 316	3.0-4.0	3.20	526	979	7.9204	-0.38	
F5F-E	SA 316	7.2-8.2	3.33	592	1098	7.9135	-0.47	
F5F-H	SA 316	12.0-13.0	2.36	625	1157	7.9376	-0.17	
16-028	SA 316						7.9507	
F5Q-B		2.5-3.5	3.08	526	979	7.9499	+0.07	
F5Q-E	50% CW 316	6.8-7.8	3.29	592	1098	7.9467	+0.03	
F5Q-I	50% CW 316	11.8-12.8	2.38	625	1157	7.9506	+0.07	
16-566	50% CW 316						7.9447	

Archive densities:— F2H 316 clad ballized and swaged once 7.9288 g.cm^{-3}
 F2T 347 ballized twice, swaged twice 7.9014 g.cm^{-3}

SA - denotes solution annealed

CW - denotes cold worked as measured by percent reduction in area

HT - denotes 24 hours at 900°F and 216 hours at 1300°F with air quenching.

Figure 2-4. Swelling in CW Type 304 After Fluence of $3.1\text{-}5.1 \times 10^{22} \text{ n/cm}^2$, $E > 0.1 \text{ MeV}$

2.1.3 Type-316 Stainless Steel

2.1.3.1 Solution Annealed Type-316 Stainless Steel

U.K. swelling data for Type-316 have been recently reviewed⁴ as have the U.S. data to the Fall of 1970.³ Two important sets of data have been obtained in the U.S.A. since the latter report; these are measurements of swelling in claddings from GE fuel pin irradiation experiments¹⁵ and data from materials capsules irradiated by HEDL.¹⁶ Table 2-1 summarizes the GE data and the void morphology data are listed in Table 2-2. The analyses of the alloys are given in Table 2-3.

In Figure 2-5 the swelling data for solution annealed Type-316 fuel cladding from the GE F2, F3A, F3B and F5 series pins are plotted as a linear function of fluence illustrating the non-linear nature of the volume increase. These data are unique in that they report swelling of solution annealed Type-316 irradiated under stress, and thus afford an opportunity to evaluate, at least in a preliminary manner, the effects of stress on swelling. This aspect of the data is described in subsection 2.3.

Table 2-2
SUMMARY OF IRRADIATION INDUCED VOID FORMATION
DATA FROM F2 CLADDING

Specimen Designation	Cladding Type	Fluence ($n/cm^2 \times 10^{22}$ E>0.1 MeV-GE)	Irradiation Temperature (°C) (°F)		Swelling ^(a) $\Delta V/V$ (%)	Mean Void Diameter Å	TEM Results Void Number Density ($cm^{-3} \times 10^{14}$)	Calculated ^(b) Swelling $\Delta V/V$ (%)
F2H-4	SA 316	3.85	512	954	1.41			
F2H-7B	SA 316	3.93	536	997	1.47			
F2H-7A	SA 316	3.77	542	1007	1.38			
F2H-8C	SA 316	3.10	546	1015	0.68	365	2.1	0.64
F2V-C	SA 316	6.15	471	881	3.95	285	13.3	2.0
F2V-E	SA 316	7.03	495	923	7.48			
F2V-F	SA 316	7.32	508	946	7.76	454	12.6	7.05
F2V-J	SA 316	7.32	523	975	8.68	640	4.8	7.85
F2V-M	SA 316	6.40	534	994	5.98			
F2V-P	SA 316	5.28	537	998	2.98	535	3.2	3.07
F2X-D	SA 316	5.58	460	861	2.86	262	10.6	1.3
F2X-J	SA 316	6.56	505	941	5.86	464	8.9	5.5
F2X-M	SA 316	5.60	516	960	3.37			
F2T-8	SA 347	3.92	538	1000	2.25	251	15.3	1.89
F2T-9	SA 347	3.76	543	1009	1.99			
F2T-11	SA 347	3.27	546	1014	1.35			
F2G-D	SA 347	6.40	487	909	5.80	270	30.0	4.3
F2G-H	SA 347	7.00	526	980	10.02	470	13.0	9.3
F2G-M	SA 347	5.50	541	1007	5.49			

(a) - Measured by immersion density.

(b) - Calculated from void size and number density data.

SA - Denotes Solution Annealed.

Table 2-3
COMPARISON OF VARIOUS COMPOSITIONS OF TYPE 316 STAINLESS STEELS

Element	GE (F2 Series)	GE (F3A Series) F3B-2	GE (F3B Series) E1H-26	GE (F5F)	HEDL (a) (Ref 17)	HEDL (b) (Ref 17)	FFTF Specification (Ref 17)	Uk M316 (Ref 17)
Chromium	17.40	17.31	17.63	17.82	17.30	16.45	17.00-18.00	16.50-17.50
Nickel	12.53	13.53	13.16	13.24	13.30	13.38	13.00-14.00	13.00-14.00
Carbon	0.05	0.07	0.04	0.05	0.06	0.056	0.040-0.060	0.03-0.06
Molybdenum	2.34	2.93	2.81	2.25	2.33	2.41	2.00-3.00	2.00-2.75
Manganese	1.66	1.71	1.90	1.80	1.72	1.51	1.50-2.00	1.50-2.00
Nitrogen	N.S.	N.S.	N.S.	0.051	0.048	0.007	0.010 Max	0.06
Aluminum	N.S.	N.S.	N.S.	0.10	N.S.	N.S.	0.050 Max	0.05
Arsenic	N.S.	N.S.	N.S.	N.S.	N.S.	N.S.	0.030 Max	0.035
Boron	N.S.	N.S.	N.S.	0.001	5ppm	<0.001	0.0010 Max	0.003
Cobalt	0.08	N.S.	N.S.	N.S.	N.S.	N.S.	0.050 Max	0.03
Niobium	N.S.	N.S.	N.S.	0.03	N.S.	N.S.	0.050 Max	0.05
Copper	0.15	N.S.	0.15	0.16	0.065	0.08	0.100 Max	N.S.
Phosphorus	0.018	0.018	0.021	0.017	0.012	0.011	0.020 Max	0.02
Silicon	0.68	0.67	0.52	0.60	0.40	0.46	0.750 Max	0.06
Sulfur	0.019	0.014	0.006	0.011	0.007	0.006	0.010 Max	0.02
Tantalum	N.S.	N.S.	N.S.	<0.001	N.S.	N.S.	0.020 Max	N.S.
Vanadium	N.S.	N.S.	N.S.	N.S.	N.S.	N.S.	0.200 Max	0.15
Titanium	N.S.	N.S.	N.S.	0.01	N.S.	N.S.	N.S.	N.S.
Iron	Balance	Balance	Balance	Balance	Balance	Balance	Balance	Balance

(a) - HEDL "Program Heat" (used for 27% and 75% swaged rod plus solution-annealed specimens)

(b) - HEDL "Car Tech Lot N" (used for prototypic 20% CW FFTF fuel pins)

N.S. - Not Specified

Figure 2-5. SA Type 316 Swelling (Fuel Pin Claddings)

The data for solution annealed Type-316 from the GE pins and the HEDL materials capsules show that Type-316 undergoes a longer threshold fluence prior to swelling than does Type-304 and then increases with fluence at a more rapid rate. Comparison of Figures 2-1 and 2-5 illustrates this on linear plots and Figures 2-6 and 2-7 compare the Type-316 data at 450 to 500°C and 500 to 550°C with a correlation for solution annealed Type-304. (The correlation is described in Section 3.) The reason for this difference is most likely linked to the increased complexity of Type-316 which has molybdenum added as a solid solution strengthener. The molybdenum may effectively impede point defect mobility so that in turn the agglomeration of vacancies and void nucleation is delayed.

The F2 fuel cladding data have provided insight concerning void nucleation and growth.

Comparison of quantitative void data for the same tube lots of solution annealed Types-316 and -347 fuel cladding irradiated in different pins to different fluence levels has shown that at 500 to 550°C (930 to 1020°F) void nucleation ceases at $3.4 \times 10^{22} \text{ n/cm}^2 E > 0.1 \text{ MeV}$ and that further swelling is due solely to the growth of existing voids. The data presented in Table 2-4 illustrate this behavior. The Type-316 data from pins F2H and F2V (Figures 2-9 and 2-10) show that little nucleation has occurred between 3.1 and $7.3 \times 10^{22} \text{ n/cm}^2 E > 0.1 \text{ MeV}$ (void number density doubled), whereas the mean void diameter has increased from 365 to 640 Å resulting in an increase in swelling from 0.7 to 8.7% $\Delta V/V$. The volume change due to the presence of voids is linear with number density and cubic with void diameter. The effect is more clear cut in the 347 data from pins F2T and F2G (Figures 2-11 and 2-12) which show almost precisely the same void number densities at 3.9 and $7.0 \times 10^{22} \text{ n/cm}^2 E > 0.1 \text{ MeV}$ while the mean void diameter has almost doubled giving an increase in overall swelling from 2.3 to 10.0%.

The data in Table 2-4 also show that the extent of void nucleation is substantially assisted by the presence of intragranular precipitate particles. Comparison of Figures 2-8 (Type-304), 2-9 (Type-316) and 2-11 (Type-347) all irradiated at similar fluences and temperatures show that the degree of intragranular precipitation increases markedly from Type-304 (light) to Type-316 (heavy) and Type-347 (very heavy), and inspection of Table 2-4 shows that the void number density increases by two orders of magnitude from Type-304 ($2 \times 10^{13} \text{ voids/cm}^3$) to Type-347 ($1.5 \times 10^{15} \text{ voids/cm}^3$). Additionally voids tend to be associated with the precipitate particles.

In the U.K. fuel pin results, solution annealed Type-316L is reported to be less susceptible to swelling than is regular Type-316.¹³ This behavior has been cited as being due to the availability of fewer precipitate particles in the Type-316L to act as void nucleation sites. This may well be a contributory factor in the case of fuel pin experiments running at high temperatures where the cladding suffers some strain and precipitation is enhanced, as found in the Type-316 clad F2 series fuel pins (subsection 2.3). However, at lower irradiation temperatures where precipitation is sluggish, little effect of carbon content on void nucleation would be anticipated; similarly, in unstressed specimens in which no enhanced precipitation occurs, the differential might be small. Data from the GE L-2 series materials test tend to confirm this. The test has been fully described elsewhere¹⁵ and was intended to test austenitic alloys in stressed and unstressed conditions. Tubular specimens were filled with sodium under a pressure of helium contained in a reservoir to provide the stress during irradiation. However, the specimens ran hotter than designed, were thus over-pressurized and failed. Additionally, the Type-316L specimens were unpressurized from start of life. The swelling data obtained from the L-2 test are listed in Table 2-5 with the analyses of the alloys in Table 2-6. In Figure 2-13 the Type-316L data are compared to the Type-316 data from the F2 fuel pin data after normalizing to an intermediate fluence of $5 \times 10^{22} \text{ n/cm}^2 E > 0.1 \text{ MeV}$ assuming a fluence dependence of $(\phi t)^{1/8}$. It is found that there is no apparent difference in the overall swelling behavior of these alloys irradiated under these conditions.

The data described in the previous paragraph suggest that when variations in void number density exist, they may only marginally influence the extent of overall swelling. A further observation from Table 2-4 fortifies this conclusion. The Type-304 specimen and the F2T Type-347 specimen were each irradiated at similar temperatures to similar fluences but had widely differing void morphologies. The 304 specimen had relatively few ($2 \times 10^{13} \text{ /cm}^3$) large voids (mean diameter 1000 Å), whereas the Type-347 had almost 100 times as many smaller voids. However, the overall swelling calculated by void counting in each case was similar (1.8% and 1.9% $\Delta V/V$). This shows that the vacancy supersaturation in each alloy was also similar since similar numbers of vacancies were employed in producing the measured swelling. The Type-347 contained many more nucleation sites (Figure 2-12) and thus many more voids were generated.

Figure 2-6. SA Type 316 Swelling at 450 to 500°C (842 to 932°F)

Figure 2-7. SA Type 316 Swelling at 500 to 550°C (932 to 1022°F)

Table 2-4
DEPENDENCE OF VOID FORMATION ON INTRAGRANULAR PRECIPITATION

Alloy	Sample Source	EBR-II Fluence $E > 0.1$ MeV $n/cm^2 \times 10^{22}$, GE	Calculated Temperature During Irradiation (°C) (°F)		Void Number Density (Cm^{-3})	Void Mean Diameter (Å)	Swelling $\Delta V/V$ (%)	Extent of Intragranular Precipitation
304	Materials Capsule MT3	3.7	555	1032	2.1×10^{13}	1000	1.8 ^(a)	Light
316	Fuel Pin F2H	3.1	545	1013	2.1×10^{14}	365	0.7 ^(b)	Heavy
316	Fuel Pin F2V	7.3	523	975	4.4×10^{14}	640	8.7 ^(b)	Heavy
347	Fuel Pin F2T	3.9	538	1000	1.5×10^{15}	250	2.3 ^(b)	Very heavy
347	Fuel Pin F2G	7.0	526	980	1.3×10^{15}	470	10.0 ^(b)	Very heavy

(a) Void volume fraction from TEM examination.

(b) Swelling from immersion density measurement.

Table 2-5
SWELLING DATA FROM L-2 SERIES CAPSULES¹⁵

Specimen	Alloy	Fluence $n/cm^2 \times 10^{22}$, $E > 0.1$ MeV-GE	Nominal Calculated Temperatures (°F) (°D)		Swelling $\Delta \rho / \rho \%$
L2A-1A	I-800 ↓	4.03	870	465	5.8
L2A-2A1		5.22	1025	550	5.4
L2A-2A2		5.5	1170	632	5.2
L2A-3A1		7.34	730	388	4.4
L2A-3A2		7.31	710	376	3.5
L2C-1A	316L ↓	4.03	840	448	0.3
L2C-2A1		5.22	995	535	3.7
L2C-2A2		5.5	1140	616	1.4
L2C-3A1		7.34	730	388	3.0
L2C-3A2		7.31	710	376	2.6
L2E-1A	347 ↓	3.73	870	465	1.5
L2E-2A1		4.8	1025	550	1.7
L2E-2A2		5.05	1175	635	1.9
L2E-3A1		6.72	730	388	2.5
L2E-3A2		6.7	710	376	0.4
L2G-1A	304 ↓	3.73	870	465	4.3
L2G-2A1		4.8	1025	550	6.6
L2G-2A2		5.05	1165	630	6.8
L2G-3A1		6.72	730	388	4.2
L2G-3A2		6.7	710	376	3.1
L2I-1A	321 ↓	2.28	878	470	0.9
L2I-2A		3.98	1076	580	1.1
L2I-2C		3.67	1184	640	1.6
L2I-2E		3.14	1004	540	0.7
L2I-3A		3.44	1022	550	0.6
L2I-3C		3.84	1166	630	1.3
L2I-3E		4.04	1049	565	1.7

Figure 2-8. Type-304 Stainless Steel from MT-3 (micrograph)

Figure 2-9. Type-316 Stainless Steel from Pin Γ2II (micrograph)

Figure 2-10. Type-316 Stainless Steel from Pin F2V (micrograph)

Figure 2 11. Type-347 Stainless Steel from Pin F2T (micrograph)

Figure 2-12. Type-347 Stainless Steel from Pin F2G (micrograph)

Table 2-6
ANALYSES OF L-2 SERIES TEST ALLOYS¹⁵

Alloy	Incoloy-800	Type 316L	Type 347	Type 304	Type 321
Capsule	L2A	L2C	L2E	L2G	L2I
Heat Number	4167	133310	99973	125653	800738
Element					
Ni (wt %)	32.00	13.31	9.32	9.15	9.53
Cr (wt %)	20.44	17.39	18.20	18.42	17.13
Fe (wt %)	46.09	Balance	Balance	Balance	Balance
C (wt %)	0.05	0.02	0.05	0.06	0.05
Mn (wt %)	0.99	1.80	1.16	1.55	1.35
Si (wt %)	0.25	0.66	0.72	0.41	0.64
Cu (wt %)	0.33	0.07	0.20	0.27	0.16
Mo (wt %)	ND	2.20	0.14	0.50	0.29
P (wt %)	ND	0.017	0.023	0.025	0.019
S (wt %)	0.007	0.008	0.009	0.010	0.011
B (ppm)	12	9.5	12	16	18.5
N (ppm)	930	500	560	540	110
Other	0.51 Ti+ 0.40	A1	0.91 (Nb + Ta)		0.53 Ti
Grain Size (ASTM)	10	6-7	10	6-7	10

ND - not determined

Figure 2-13. Types 316 and 316L Immersion Density Data

2.1.3.2 Cold Worked Type-316 Stainless Steel

Cold worked Type-316 has attracted much attention as a consequence of U.K. observations from fuel pin claddings^{4,13} which showed 20% cold worked Type-316 to exhibit substantially less swelling than the solution annealed variety of the alloy. These results substantially influenced the specification of 20% cold worked Type-316 as the fuel cladding and structural material for initial FFTF cores and have generated extensive U.S. experimental programs to investigate the irradiation behavior of the alloy. The initial results from these programs suggest even more promise of good swelling resistance in the alloy than do the U.K. data. However, some aspects of the U.S. and U.K. data are in conflict without any wholly satisfactory explanations, so that the swelling behavior of cold worked Type-316 is by no means established and understood.

The U.K. pin data¹³ showed maximum swelling of about 3% $\Delta V/V$ in cold worked Type-316 compared to up to 9% $\Delta V/V$ in solution treated cladding for fluences of approximately $6 \times 10^{22} \text{ n/cm}^2$ $E > 0.1 \text{ MeV}$. Transmission electron microscopy examinations¹⁷ showed that the cold work had not significantly changed the void number density but had caused reduced swelling by reducing the void growth rate.

In contrast the U.S. data show essentially no swelling in cold worked Type-316 for fluences up to $\sim 5 \times 10^{22} \text{ n/cm}^2$, $E > 0.1 \text{ MeV}$, and TEM reveals that where voids have appeared they are of similar size to those observed in solution annealed Type-316 irradiated under similar conditions, but there are drastically fewer of them.¹⁴

Thus in the case of the U.K. data, swelling resistance is apparently imparted by cold work causing a reduced void growth rate whereas in the U.S. experience, resistance to swelling is a consequence of a lowered nucleation rate. These differences cannot be obviously reconciled but may be a consequence of the differing manners of irradiation of the U.K. and U.S. material. This is discussed later. Figure 2-15 shows the U.S. data and the range of the U.K. data plotted as a function of fluence. Only marginal swelling has so far been observed in CW Type-316 irradiated in the U.S.

Some differences in behavior due to differing metallurgical pre-treatments and degrees of cold work cannot be completely discounted, but are likely to be minor. The U.K. data are nominally from 20% cold worked material but little else is known. The U.S. data^{14,16} are from materials having 15% to 75% cold work. However, HEDL workers have shown (Figure 2-14) that variations of 15 to 25% cold work have very little effect on swelling, at least at low fluences.¹⁶

No significant or consistent variations are apparent in the chemical composition of the alloys (Table 2-3). The U.K. data are all from Type-M316 which differs from U.S. AISI Type-316 only in a tightening of the allowable nickel content.¹³

DFR and EBR-II are similar-sized experimental fast reactors with similar in-core flux levels so that effects of different neutron environments can only be minor. However, DFR is a down-flow reactor so that the hot end of an experimental fuel pin is located at the bottom of the core with the plenum in the upper part of the pin separated from the hottest fuel by the fuel column. It is significant that all of the U.K. data for cold worked Type-316 swelling are from fuel pin claddings whereas the very different U.S. data with only two exceptions are from materials test specimens. These exceptions are (a) the data showing zero swelling from the GE F5Q fuel pin clad in 50% cold worked Type-316 and irradiated to $3.3 \times 10^{22} \text{ n/cm}^2$ $E > 0.1 \text{ MeV}$ at mid-wall cladding temperatures to 625°C (1160°F) and (b) the data from the prototypical FFTF pin (20% cold worked Type-316 cladding irradiated in the PNL-17 experiment in EBR-II to $3.0 \times 10^{22} \text{ n/cm}^2$ $E > 0.1 \text{ MeV}$ at mid-wall cladding temperatures to 570°C (1060°F).¹⁶ This latter pin exhibited slight void formation at 470°C (880°F) of $\sim 10^{1.3} \text{ voids/cm}^3$ of $\sim 200 \text{ \AA}$ average diameter. It is possible that the stress system existing in a fuel pin could lead to more swelling in fuel cladding specimens than in materials specimens and that the larger swelling found in the U.K. claddings may be due to differences in fuel rod geometry and mode of operation. It is equally plausible that the U.K. and U.S. volume change data are compatible and that cold worked Type-316 has a long incubation period before swelling appears, but the subsequent increase in swelling with fluence is very rapid. It should be noted that Bullough has suggested that this should occur in cold worked Type-316¹⁰ and that ion bombardment studies at 525°C (975°F) have shown similar behavior.¹⁸

Figure 2-14. Swelling in Type-316 Stainless Steel

Figure 2-15. Swelling in Cold Worked Type 316 After Irradiation at 370 to 600°C (700 to 1110°F)

2.1.4 Stabilized Stainless Steels

Elements with a high propensity for the formation of carbides and nitrides are commonly added to stainless steels to inhibit the formation of $M_{23}C_6$ which is rich in chromium. Chromium depletion of the austenite matrix in this way leads to the condition classically named "sensitization" in which the locally depleted regions are rendered susceptible to corrosion attack. The stabilizing elements are commonly niobium (AISI Types-318, -347 and -348, U.K. FV548) or titanium (AISI Type-321, U.K. EN58B).

The only data describing Type-347 swelling are from the GE F2 series fuel pin claddings (Table 2-1) and the GE L2 series materials irradiation (Table 2-5 and Reference 19). These data show that solution annealed Type-347 swells in a very similar manner to solution annealed Type-316. Transmission electron microscopy (Figure 2-12) shows that Type-347 tends to form more intergranular precipitation than Type-316 irradiated under similar conditions (Figure 2-10) and hence nucleate more voids. However, a comparison of the immersion density data (Figure 2-16) shows that Type-347, if anything, swells only slightly more than Type-316. The comparative swellings of the austenitic alloys irradiated in the L2 test are shown in Figure 2-17 which illustrates the similarity in swelling of Types-347 and -316.

Relevant U.K. data are from a fuel pin clad in solution treated and aged FV548.¹⁷ The swelling was similar to that observed in solution treated Type-316. Electron microscopy revealed that voids tended to be associated with very small precipitates of NbC.

Swelling measurements from cold worked FV548 are extremely low¹³ and suggest that cold working may be more effective in inhibiting swelling in stabilized stainless steels than has been shown in Type-316. Thermomechanically treated Type-348 has been examined²⁰ with a view to providing a swelling resistant alloy. However, results to date suggest that the onset void formation has been delayed but not eliminated.

Data describing swelling in titanium stabilized stainless steels have been generated by GE (Type-321 this report), ORNL (Ti modified Types-316 and -304^{21,22,23}) and U.K. (EN58B).¹³ These data suggest that there is an optimum titanium content that will prove swelling resistance. Together with the probability that cold working will lead to increased swelling resistance, this has, in part, led to the specification of Type-321 as a candidate for first core cladding for the GE LMFBR Demonstration Plant. The GE data are from an L2 materials capsule (Table 2-5) and from a fuel pin (F5M, Table 2-1) irradiated to $3.2 \times 10^{22} \text{ n/cm}^2$, $E > 0.1 \text{ MeV}$ at mid-wall cladding temperatures to 627°C (1160°F). The data generally show significantly less swelling than solution annealed Type-316 data at comparable fluences and are very similar to volume increases found in the U.K. cold worked Type-316 data at comparable fluence levels.

Workers at ORNL have irradiated standard Type-316 and Ti-modified Type-316^{21,22,23} (0.23% Ti) in the annealed and 10% cold worked conditions to fluences of 1.9 to $2.7 \times 10^{22} \text{ n/cm}^2$, $E > 0.1 \text{ MeV}$ over a wide range of temperatures. Inspection of Figure 2-15 shows that very little swelling would be expected in Type-316 after these exposures so the results must be approached with caution. Nevertheless the immersion density data show consistent detectable volume increases of up to $0.16\% \Delta V/V$ in the annealed Type-316 compared to no measurable change in the Ti-modified alloy.²² TEM examinations of specimens irradiated at 580°C showed that the reason for the reduced swelling in the annealed modified alloy is a large reduction in the concentration of voids.²³ The concentration of voids in the standard steel was about $1 \times 10^{13} / \text{cm}^3$ and all voids were associated with precipitate particles. No precipitate particles were found in the modified alloy and the void concentration was less than $10^{12} / \text{cm}^3$.

U.K. workers have suggested that the inhibition of $M_{23}C_6$ precipitation by deliberate additions of carbide forming elements is effective in reducing stainless steel swelling and have cited data showing low swelling in FV-548 (Nb-stabilized stainless) and EN58B (Ti-stabilized 18-8 stainless) as evidence.¹³

2.2 NICKEL ALLOYS

Nickel alloys offer excellent promise as longer range candidate materials for fuel pin claddings and core structural materials because of their high strength and their potential ability to resist swelling. Results described here will be confined to commercial alloys relevant to LMFBR application. Various purities of nickel as well as some alloy compositions have been extensively investigated, particularly in ion bombardment studies with the objective of providing basic information on void formation and growth. These results are fully described and reviewed in two recent conference proceedings.^{5,6}

Figure 2-16. Clad Volume Increase Versus Neutron Fluence in F2 Series Fuel Pins

Figure 2-17. Comparative Swelling of Austenitic Alloys Based on Results from L2 Tests

U. K. work²⁴ has shown that Nimonic PE-16, an age-hardening nickel alloy, potentially offers excellent resistance to swelling. PE-16 is a nickel base alloy containing about 42% Ni, 35% Fe, 17% Cr, 3% Mo, 1.2% Ti, 1.2% Al and 0.08% C. The Ti and Al content results in a precipitation hardening alloy which gives it excellent strength properties. It is age-hardenable by solution-heat-treatment in the range 1020 to 1150°C (1870 to 2100°F) and subsequent precipitation of gamma prime [in this case Ni₃(Al, Ti)] by holding in the range of 700 to 800°C (1290 to 1470°F). Carbide formation is limited by the low carbon content but discrete grain boundary carbides are formed during cooling from the solution treatment temperature.

The likelihood of resistance to swelling is based upon results from ion-bombardment experiments²⁴ using 20 MeV carbon ions or protons as the bombarding species. These tests were conducted at 525°C (975°F) and showed maximum swelling of 0.5% $\Delta V/V$ after exposure of 300 displacements/atom (equivalent to approximately 4×10^{23} n/cm², $E > 0.1$ MeV in EBR-II). Similar tests showed swelling of $\sim 15\%$ $\Delta V/V$ in nickel and solution annealed Type-316 after the same exposure. Results from fuel pin claddings irradiated in DFR are much less dramatic^{13,17} but also show superior swelling resistance in PE-16 as compared to cold worked or solution treated stainless steels, although it is noteworthy that a DFR irradiated cold worked FV548 ($\equiv$ AISI 318) clad pin showed similar swelling resistance to the PE-16.¹³ The swelling resistance of PE-16 is ascribed to the presence of the finely distributed Ni₃(Al, Ti) gamma prime precipitate; coherence and small interparticle size are considered critical. Recent work at GE-BRD with Inconel-625²⁵ and described below strongly suggests that full coherence is not a prerequisite in obtaining swelling resistance. An experimental duplex alloy IN-744 irradiated by ANL showed very low void swelling²⁶ in the ferritic phase which contained a fine coherent precipitate providing additional support for the use of small precipitates to control swelling.

Tensile specimens and pre-cut electron microscopy discs of Incoloy-800, Hastelloy-X and Inconel-625 were irradiated in a GE materials capsule (MT-3) at 600 to 650°C (1100 to 1200°F) to a maximum fluence of 5.5×10^{22} n/cm², $E > 0.1$ MeV. Table 2-7 lists the compositions of the alloys.

Table 2-7
CHEMICAL COMPOSITIONS OF NICKEL-BASE ALLOYS (MT3 CAPSULE)
(REFERENCES 15 AND 34)

Elements	Incoloy 800 (HH-9696A) ^(a)	Inconel 625 (Y8668) ^(a)	Hastelloy X (X1-4370) ^(a)
C	0.06	0.03	0.10
Mn	0.89	0.12	0.60
Fe	45.70	1.86	18.7
S	0.007	0.007	0.006
Si	0.41	0.25	0.85
Cu	0.13	0.03	ND
Ni	32.03	62.31	Bal
Cr	20.75	21.98	21.68
Al	0.10	0.17	ND
Ti	0.21	0.23	ND
Mo	ND	8.75	9.17
Nb + Ta	ND	4.24	ND
B	0.0030	0.0065	0.0040
W	ND	ND	0.44
Co	ND	ND	1.87

(a) - Heat Numbers

ND - Not Determined

Immersion density measurements were made of swelling in the tensile specimens. It was found that the Incoloy-800 showed 0.05 to 0.5% swelling, the Hastelloy-X showed 0.6 to 2.1% swelling, while the Inconel-625 showed densifications of up to 0.6%. The only other swelling data for Incoloy-800 were obtained from immersion density measurements of specimens irradiated in the L2A materials capsule. These data as shown in Figure 2-18 show more swelling than found in solution annealed stainless steels irradiated under similar conditions (Figure 2-17).

The transmission microscopy results from the MT-3 capsule are equally dramatic. The Incoloy-800 discs contain voids with an average diameter of about 850 Å and a number density of less than 10^{13} per cm^3 . Figure 2-19 shows a typical area. Some very small pores were detected, especially at twin boundaries. These pores are probably helium bubbles. The microstructure is generally clean, except for a few carbide particles (M_{23}C_6) of similar size to the voids which frequently have voids associated with them. The general microstructure is very similar to that of solution annealed Type-304 stainless steel irradiated at the same temperature, i.e., a normal austenitic structure containing little precipitation. By contrast the Hastelloy-X discs contain very large voids and a considerable quantity of large rod-like precipitates as seen in Figure 2-20. The voids are so large (2500 to 10,000 Å diameter on the micrographs) that it is obvious that most of them have been enlarged during thinning. However, examination of very thick areas of the foil which were otherwise opaque showed clearly defined voids with diameters up to 3000 Å. Also the void number density is very small (approximately 1.3×10^{12} voids/ cm^3) and an average void size of ~ 2000 Å is required to achieve the volumetric swellings of up to 2.1% measured by liquid immersion in the tensile specimens. Thus it is evident that the swelling measured in the Hastelloy-X is due to a small number of very large voids. Examination of unirradiated Hastelloy-X shows that the rod-shaped precipitates seen in Figure 2-2 have appeared during irradiation, and from the work of Comprelli and Lewis²⁶, they are most probably M_6C .

Figure 2-18. Incoloy-800 Immersion Density Data from L2A

Figure 2-19. Incoloy-800 (TEM micrograph)

Figure 2-20. Hastelloy-X (TEM micrograph)

Figure 2-21. Inconel-625 (TEM micrograph)

Figure 2-22. Inconel-625 (TEM micrograph)

Micrographs of the Inconel-625 are presented in Figures 2-21 and 2-22 and it is seen that no voids have been generated. Instead, the microstructure contains a very high number density of finely dispersed precipitates in the form of semi-coherent discs 1000 to 2000 Å in diameter and 100 to 200 Å thick. The habit planes of the discs are [100] matrix planes, and the interlamellar spacing is ~ 500 Å as shown in Figure 2-22. The matrix precipitate is not the stable orthorhombic Ni_3Nb which had been identified in an earlier study²⁷ of unirradiated Inconel-625 heat treated for ~ 1000 hours at 1200°F. Electron diffraction patterns obtained from the irradiated Inconel-625 were indicative of the metastable tetragonal Ni_3Nb phase described by Royer and Gantois²⁸ with unit cell dimensions $a = 3.57 \text{ Å}$, $c = 7.39 \text{ Å}$. With this unit cell, there is nearly perfect coherency in the [100] and [010] directions of the precipitate phase but a 3% deviation in the [001] direction. These facts account for the platelet shape and for the coherency strains observed. Coherency strains are invisible when the platelets are seen edge-on.

The structure of the unirradiated solution-annealed material contained no Ni_3Nb but only larger equiaxed carbide precipitates which are also present in the post-irradiation structure. The metastable Ni_3Nb was precipitated during in-reactor aging and apparently it has inhibited void formation and caused a slight densification of the alloy.

The vast differences in swelling behavior of these alloys is attributed to their different aging characteristics. Incoloy-800 for example with Ti content below 0.4 wt % is a fully austenitic solid solution alloy with no purposeful additions for second-phase strengthening, and thus behaves much as would be expected on the basis of results from solution annealed Type-304 stainless steel (i.e., showing moderate swelling at 630°C (1165°F)). Limited M_{23}C_6 appears and assists void nucleation. Otherwise void formation is random throughout the structure except at the grain boundary areas.

The Hastelloy-X has formed a relatively coarse carbide precipitate of rods ranging from 7000 to 10,000 Å in length and by about 1000 Å thick. These rods were sufficiently spaced for dislocations to have appeared between and among them and this is the likely cause of the appearance of the very large voids. The dislocations were able to move to and fro during irradiation and accepted more of the interstitials than the less mobile vacancies arising from the neutron bombardment. The resultant excess vacancies nucleated and grew voids on the surfaces of the precipitates. Once nucleated the voids continued to grow from the excess vacancies arising from the imbalance of point defects caused by the moving dislocations.

By contrast the Ni_3Nb precipitate in the Inconel-625 is on a much finer scale as well as being semi-coherent. The structure shows very few dislocations after irradiation and it appears that the closely spaced second-phase particles lower the overall point defect concentration by providing sites for the mutual annihilation of vacancies and interstitials. Interstitials have a higher lattice diffusivity than vacancies. If, during their random movements, they encounter a precipitate particle, they will adhere to the precipitate-matrix interface which represents a lower energy state than interstitial sites in the metal matrix. This causes a compressive stress field to result which repels further interstitials but which attracts vacancies causing mutual annihilation and thus lowers the overall point defect concentration and reduces the ability of the system to produce any excess vacancy concentrations. The precipitate particles are spaced closely enough together to prevent major interstitial loop formation and essentially inhibit dislocation motion such that large excess vacancy concentrations do not occur and no voids nucleate.

The PE-16 swelling resistance is ascribed to the formation of a coherent gamma-prime precipitate of $\text{Ni}_3(\text{Al},\text{Ti})$.²⁴ The coherence and interparticle spacing are considered critical. In the ion bombardment studies, the PE-16 had been aged to give gamma-prime precipitates of about 100 Å diameter in a concentration of about $2 \times 10^{16}/\text{cm}^3$.

However, in the case of the Inconel-625 precipitate, no voids appeared even though complete coherence was not maintained. Thus, incoherent or partially coherent particles, if sufficiently finely dispersed, may also effectively inhibit swelling. Whether void formation will eventually occur in the over-aged structure is not yet known. The answer should be forthcoming by examination of the companion capsule MT-4 which is currently in-reactor undergoing irradiation to a fluence in excess of 10^{23} n/cm^2 .

2.3 EFFECTS OF EXTERNALLY APPLIED STRESS

Solution annealed Type-316 clad fuel pins irradiated in DFR have frequently shown anomalously high clad swellings. Approximately 9% volume increase has been measured after the moderate exposure of $5 \times 10^{22} \text{ n/cm}^2$, $E > 0.1 \text{ MeV}$.¹³ These swellings, which occurred for mean mid-wall temperatures greater than 500°C (930°F), have generated considerable speculation concerning the possible existence of enhanced void growth under stress (stress induced void growth), and its theoretical treatment has already been attempted.^{29,16} To date the effect has not been verified experimentally because specimens irradiated under the necessary conditions have not been available for examination in the U.S.A. However, the detailed investigation of the void morphology in the F2 series fuel cladding has provided insight on the effects of an externally applied stress on void formation and growth for mean cladding temperatures to 550°C (1025°F). The F2 fuel pin profilometry and clad swelling measurements had shown that the three high burnup pins had undergone varying amounts of plastic deformation.³⁰ The maximum plastic deformation values were 1.5%, 0.8% and 0.1% $\Delta D/D$ for fuel pins F2G, F2V and F2X respectively.

A stress system could affect void formation and growth at high cladding temperatures in at least two ways: (a) by the formation of grain boundary creep cavities in the manner described by Hull and Rimmer³¹ which would be a minor effect, or (b) by causing an increase in void growth kinetics as described by Gulden and Kaae³² which could have a major influence on the extent of swelling. Examination of the grain boundaries in the F2G, F2V and F2X specimens showed no evidence of grain boundary creep cavities. Conversely, zones denuded of voids were apparent at the grain boundaries. Figure 2-23 shows a typical grain boundary area of a specimen (F2G-H) which had experienced 1.5% $\Delta D/D$ plastic deformation during irradiation.

The question of a stress effect on void size is more difficult to resolve experimentally. The void size is acutely dependent on irradiation temperature as was shown for Type-304.^{8,33} Thus any comparison of void size must be made at the same temperature. Precise knowledge of sample temperature during irradiation in EBR-II is subject to uncertainties, especially between differing specimen geometries. Void size is also dependent on neutron fluence so that comparisons must also be made at similar fluence levels in order to minimize the uncertainties. Unfortunately no void size data are available from unstressed solution annealed Type-316 for fluences as high as those of pins F2V and F2X. Recently reported data on swelling of unstressed Type-316¹⁶ are at fluences comparable to that obtained by F2H. These specimens (HEDL) irradiated at 1000 to 1100°F (538 to 593°C) to 2.63 and $4.47 \times 10^{22} \text{ n/cm}^2$, $E > 0.1 \text{ MeV}$ showed average void diameters of 274 and $385 \text{ \AA}$ respectively with a void number density of 5×10^{13} to $10^{14} \text{ voids/cm}^3$.¹⁶ Specimen F2H-8C (Table 2-4) irradiated in the same temperature range to an intermediate fluence level showed an average void diameter of $365 \text{ \AA}$ and a number density of $\sim 2 \times 10^{14} \text{ voids/cm}^3$. Considering the temperature and fluence uncertainties, the void sizes in the stressed and unstressed specimens are equivalent. Void number density is difficult to determine, a factor of two can be within the experimental error. However, the swelling in the HEDL specimens was low ($\sim 0.1\% \Delta V/V$) compared to $0.68\% \Delta V/V$ for F2H-8C. Thus if the void sizes are similar then the higher void concentration in F2H-8C is real.

A comparison of Type-316 specimens F2X-J and F2V-F (irradiated at 500 to 510°C (932 to 950°F)) is shown in Figure 2-24. Specimen F2V-F underwent $0.7\% \Delta D/D$ plastic deformation compared to $0.1\% \Delta D/D$ for F2X-J. It can be assumed that F2V-F was subjected to higher sustained stresses while being irradiated at the same temperature as F2X-J. The measured mean void diameters are almost exactly equal but the void number density is higher in F2V-F. As in the previous comparison the measured swelling also was greater in the specimen with the higher void number density. Since the void sizes are equivalent, the observed difference in void concentration is considered real and leads to the conclusion that the stressed specimens of solution annealed Type-316 stainless steel irradiated at $\sim 500^\circ\text{C}$ (932°F) contain more voids than the unstressed specimens. Inspection of the intragranular precipitation in F2V-F and F2X-J suggests that the extent of matrix precipitation is also greater in the stressed specimens as can be seen in Figure 2-24. These data suggest that the effect of an external stress system on void formation in this temperature range is not to generate creep cavities or enlarge existing voids but rather to increase the degree of intragranular precipitation which in turn increases the void concentration and the overall swelling. This explanation appears to be consistent with the F2 series data described here, the HEDL unstressed data¹⁶ and the DFR data¹³ in which the higher swelling of Type-316 compared to Type-316L was attributed to the more extensive matrix precipitate formation in Type-316.

SPECIMEN IRRADIATED AT 525°C TO $7.0 \times 10^{22} \text{ n/cm}^2$.
 $E > 0.1 \text{ MeV-GE}$. MEAN VOID DIAMETER = 470 Å, VOID
NUMBER DENSITY = $1.3 \times 10^{15} \text{ /cm}^3$. SWELLING = 10%

Figure 2-23. TEM of Type 347 Cladding from Pin F2G Showing Void Denuded Grain Boundaries

(a) SECTION F2V-F IRRADIATED AT 510°C (950°F) TO $7.3 \times 10^{22} \text{ n/cm}^2$, $E > 0.1 \text{ MeV-GE}$. $1.3 \times 10^{15} \text{ VOIDS/cm}^3$
 MEAN DIAMETER – 454 Å. DENSITY SWELLING – 7.8%
 PLASTIC DEFORMATION – 0.7%

(b) SECTION F2X-J IRRADIATED AT 510°C (950°F) TO $6.6 \times 10^{22} \text{ n/cm}^2$, $E > 0.1 \text{ MeV-GE}$. $9 \times 10^{14} \text{ VOIDS/cm}^3$.
 MEAN DIAMETER – 464 Å. DENSITY SWELLING – 5.9%
 PLASTIC DEFORMATION = 0.1%

Figure 2-24. TEM of Type 316 Cladding from F2 Fuel Pins Showing Difference in Number Density of Matrix Precipitates

At higher clad temperatures, stress induced void growth may well occur. A recent observation at GE-BRD may eventually confirm this. A solution annealed Type-316 cladding pin (E1H-26) was irradiated to $\sim 80,000$ MWd/Te (peak fluence $\sim 4.9 \times 10^{22}$ n/cm², $E > 0.1$ MeV) at mid-wall cladding temperatures to 625°C (1157°F). To date two immersion density measurements have been made and are shown in Figure 2-5 together with other solution annealed Type-316 cladding data. The 505°C (941°F) specimen shows a volume increase that is consistent with the previous data. However, the 573°C (1063°F) specimen shows a volume increase of 4.7% which is substantially more than expected especially remembering that peak swelling for solution annealed Type-316 is thought to be 520°C (968°F). It is possible that stress enhanced swelling has occurred in this section.

U. K. workers³⁴ have looked for stress effects in the swelling of nickel irradiated at 500°C (932°F) under tensile loads of 34 to 2500 psi. To date no stress effect has been observed on either the void size or concentration. The experiment is continuing at stress levels of ~ 7500 psi. In view of the fuel pin data suggesting little effect to 550°C, the experiment should also be performed at higher temperatures.

At low irradiation temperatures ($\sim 430^\circ\text{C}$, 800°F), evidence has been cited³⁵ to show that stress can reduce swelling. Immersion density measurements of swelling were made in the dimple region of an EBR-II hexagonal-can (Type-304 stainless steel). This particular dimple was stressed by its contact with its neighbor while it swelled and the measurements showed less swelling than anticipated in the dimple region. The stress analysis is complex and open to question but it was concluded that in this case a tensile stress had reduced swelling. It should be noted that an equally plausible explanation is that the reduced swelling was due to cold working of the area around the dimple during fabrication.

Reduced swelling under a tensile stress was considered to be due to the stress field initiated around the voids which leads to increased interstitial flux to the void and thus a decreased growth rate. A compressive stress was held to have the opposite effect. At high irradiation temperatures and low vacancy supersaturations, the effect might be expected to be swamped out by the stress changing the probability of a vacancy leaving the void and leading to further void growth.³²

Stress induced void growth has been considered analogous to Herring-Nabarro creep.^{29,36} Li and Nolfi³⁶ maintain that if stress induced void growth occurs due to a non-hydrostatic stress system (e.g., biaxial tension in fuel pin cladding) then linear dimensional change in the component will no longer be given by the commonly used

$$\frac{\Delta \ell}{\ell} = \frac{1}{3} \frac{\Delta V}{V}$$

where ℓ is the linear dimension and V is the volume. Essentially the appearance of elongated voids is predicted. These elongated voids would have the effect of causing preferred growth in the bulk material and in the elongation direction. Such voids have not been reported to date.

3. EMPIRICAL ANALYSIS OF STAINLESS STEEL DATA FOR DESIGN REQUIREMENTS

3.1 TYPE-304 (SOLUTION ANNEALED)

A review of experimental data (subsection 2.1) has indicated that the increase-in-volume has a power law dependence on neutron fluence (Figure 2-1). Data analysis has reinforced the theoretical prediction⁹ that at a given fluence the temperature dependence of fluence should exhibit data in the shape of a "bell-shaped curve." The further prediction that swelling should be flux dependent rather than fluence dependent was not found in the data. Thus the data were correlated in terms of fluence and temperature of irradiation alone. Over 150 data points obtained from immersion density and profilometry measurements on unstressed components irradiated in EBR-II were utilized in the correlation. Most of these, however, are from EBR-II structural components exposed to the EBR-II coolant and thus irradiated in the narrow temperature range of 370 to 470°C (700 to 800°F). Data from the GE-BRD fuel pin capsule tubes were most useful in providing a large body of data at higher temperatures and fluences.

Correlation of the measured swelling as a function of fluence and temperature resulted in the following expression

$$\frac{\% \Delta V}{V} = (\phi t)^{1.57} \exp(-35.01 + \frac{2.968 \times 10^4}{T} - \frac{6.567 \times 10^6}{T^2})$$

where: ϕt is fluence for $E > 0.1$ MeV $\times 10^{-22}$ n/cm², GE.

and T is the irradiation temperature in °C.

The data used to generate this equation covered the temperature range of 370 to 700°C (700 to 1292°F) with fluences up to 7.5×10^{22} n/cm², $E > 0.1$ MeV.

3.2 TYPES 316 AND 321

Swelling data for Type-321 stainless steel are sparse. However, the data that do exist (Tables 2-2 and 2-6) suggest that solution annealed Type-321 swelling is very similar to that of 20% cold worked Type-316 used in DFR fuel pin claddings. It was assumed that the data could be described by the same form of equation as used for solution annealed Type-304 and a fit of the data yielded the following correlation for cold worked Type-316 stainless steel and solution annealed Type-321 stainless steel.

$$\frac{\% \Delta V}{V} = 8.9 \times 10^{-2} (\phi t)^{1.5} \exp(-3.14 + \frac{5.04 \times 10^3}{T} - \frac{1.66 \times 10^6}{T^2})$$

where ϕt is fluence $\times 10^{22}$ for $E > 0.1$ MeV

and T is the temperature in °C.

Data used in generating this equation covered the temperature range from 280 to 650°C (536 to 1202°F) with fluences up to $\sim 5 \times 10^{22}$ n/cm², $E > 0.1$ MeV.

4. THE MECHANISMS OF SWELLING

4.1 FORMATION OF POINT DEFECTS

During irradiation the metal lattice is subject to a continuous bombardment by high energy neutrons. Each neutron causes a "collision cascade" in the lattice. That is a number of atoms are knocked out of their equilibrium positions while atoms further from the primary impact are displaced somewhat. The extent of displacement is reduced with distance from the primary impact until eventually no further effect is noticed. The majority of these displaced atoms will rapidly find themselves new equilibrium lattice positions. However, a small number will remain displaced and will give rise to an overall defect concentration consisting of equal numbers of vacancies (vacant lattice sites) and interstitials (atoms in nonlattice positions). These point defects are free to move and form voids and/or loops as described in subsection 4.2. The present discussion is confined to the collision cascades caused by the neutron bombardment and the question of how these influence the eventual void formation.

The number of point defects arising from a collision cascade is dependent on the energy of the bombarding neutron. However, the eventual swelling is less influenced by the total number of point defects produced than by the excess of vacancies arising from preferential recombination of interstitials. As discussed in subsection 4.3, the preferential interstitial recombination is highly dependent on the dislocation density and precipitate morphology of the alloy. Thus knowledge of the precise nature of the collision cascade is not a prerequisite to the understanding of void growth and of methods to inhibit swelling.

The mechanisms by which voids nucleate are less well defined and it has been hypothesized that the initial displacement cascades act as nuclei or, alternatively, that dissolved impurity gas atoms act as nucleation sites. High voltage electron microscope investigations¹¹ appear to have demonstrated that collision cascades are not necessary for void nucleation. During electron irradiation no cascades or impurities are formed. Experiments with electron irradiation of nickel produced no voids, only interstitial and vacancy loops. However, samples pre-injected with gas atoms and annealed to remove the accompanying displacement damage and then irradiated with electrons, did exhibit void formation. Conclusion is that the precise knowledge of displacement cascade formation is not of first importance in void nucleation and therefore will not be considered further.

4.2 VOID FORMATION AND GROWTH

4.2.1 The Role of Impurity Gases

Aspects of the influence of gaseous impurities on void nucleation were briefly discussed in the previous section in which evidence was cited to show that voids can nucleate in the absence of collision cascades but apparently not in the absence of implanted gaseous impurities. Experiments to demonstrate unequivocally a necessity for the presence of gaseous impurities in order that void nucleation can occur are very difficult to perform, since gaseous impurities can never be wholly eliminated. Nevertheless, ion bombardment and electron irradiation studies have shown that void concentrations increase as the amount of pre-injected helium is increased which shows a direct relationship between gas content and void number density. Additionally, ion bombardment studies on a titanium-modified stainless steel (U.K. EN58B) have shown no void formation unless helium was pre-injected. In this experiment¹² oxygen was thought to be present as an impurity but presumed to have been taken up by the titanium. Thus it is evident that gaseous impurities play a most important role in assisting void nucleation. Since during reactor irradiation helium and hydrogen are formed by $n(\alpha)$ and $n(p)$ reactions, it is evident that adequate concentrations of gaseous impurities will inevitably be available to assist void nucleation. Thus it would appear that given a sufficient vacancy supersaturation, void formation in-reactor is inevitable and attempts to inhibit swelling must attack the mechanism producing the supersaturation rather than attempting to influence void nucleation by trying to limit the formation of gaseous impurities.

4.2.2 The Role of Dislocations

The collision cascades caused by the bombarding neutrons give rise to equal numbers of vacancies and interstitials. If no means were available to produce an excess concentration of vacancies, then no voids could form and the point defects which did not recombine would agglomerate only as interstitial and vacancy loops. Such loop formation would cause no more than a trivial volume increase. That an excess vacancy concentration does occur is due to two factors: first, the slightly higher diffusivity of an interstitial than a vacancy; and second, the preferential attraction that an interstitial loop has for further interstitials. The higher diffusivity means that interstitials more readily form loops (essentially an extra-plane of atoms at some location in the overall matrix). These interstitial loops have a characteristic dislocation strain field around them that tends to attract further interstitials. Thus these loops rapidly grow and form an irradiation-induced dislocation network. This dislocation network is the prime source of the point defect imbalance causing swelling. As long as the dislocations are free to move, they will continue to absorb more interstitials than vacancies so that an excess vacancy concentration is created. Pre-existing dislocations will behave in the same manner. The excess vacancies may attempt to form vacancy loops but the associated strain field again tends to attract interstitials so that vacancy loops tend to be annihilated. Then the excess vacancies apparently form stable three dimensional nuclei with the aid of impurity gas atoms, and these grow by accepting further excess vacancies to form the voids commonly seen in transmission micrographs. Thus it is evident that maximum control of swelling can be achieved by control of the dislocation density and mobility; this is discussed further in subsection 4.4.

Results from cold worked stainless steels point to a potentially different aspect of dislocation behavior. Cold working causes a very high density of dislocations which have frequent intersections among them. Their ability to move is much less than that of an irradiation induced dislocation network (which is generally less dense) or of the dislocations

which exist in nominally annealed material. Thus their contribution to the overall vacancy concentration is less for three reasons; first, their relative inability to move means that they cannot accept an excess of interstitials, but rather must accept more nearly equal numbers of vacancies and interstitials. Acceptance of an interstitial causes a jog, which could be reversed by accepting a vacancy. Second, the inter-dislocation spacing is so small that the growth of interstitial loops with its commensurate production of excess vacancies is restricted. A third aspect is that the dislocation network has the effect of isolating collections of vacancies by preventing long range movement; this retards nucleation until sufficient vacancy concentrations can form in conjunction with impurity gas atoms in the compartmented structure.

A further effect which applies both to dislocation networks and to second phase particles and which is discussed in the next section is that the dislocation network retards point defect migration to grain boundaries. Grain boundaries are unbiased sinks for point defects; that is, they are equally receptive to interstitials and vacancies. However, because of the higher lattice mobility of interstitials, more interstitials than vacancies are absorbed at grain boundaries so that grain boundaries contribute to a vacancy supersaturation. Work at ORNL^{2,3} has shown that ultra-fine grain size Type-304 with its large grain boundary area swells more than Type-304 of "normal" grain size. In a cold worked structure or a structure containing dense precipitates, the dislocation network and/or the precipitate particles inhibit interstitial migration to the grain boundaries and thus lower the excess vacancy concentration and reduce the propensity for void formation.

4.2.3 The Role of Second-Phase Particles

Early observations of void formation in Type-304 stainless steel include occasional voids in conjunction with precipitate particles,³ and it was speculated that these particles were in some way nucleation sites. More recent observations described in this report have shown that in the more complex stainless steels (Types-316 and -347) and in nickel alloys (Inconel-625, Incoloy-800, Hastelloy-X, Nimonic PE-16) second-phase morphology can be crucial to void formation and growth and in the optimum condition can actually control the degree of swelling.

Investigations of annealed Types-316 and -347 stainless steels (subsection 2.1.3) confirmed that second-phase particles can act as nucleation sites and enhance the void number density (without necessarily enhancing the degree of swelling). Investigations of nickel-based alloys showed that very heavy finely dispersed precipitation can inhibit swelling (Inconel-625, Nimonic PE-16) while coarser precipitation led to gross swelling in Hastelloy-X (subsection 2.2).

The critical aspect of second-phase particles appears to be their inter-particle spacing. This spacing exerts a controlling influence on the eventual extent of swelling. In alloys which produce well spaced second phases, interstitial loops can grow and move freely and thus contribute to an excess vacancy concentration. Impurity gas atoms are probably attracted to the second phase particles which also collect vacancies from the excess concentration and thus nucleate voids. On the other hand, if the second-phase is sufficiently fine and closely spaced, the growth of interstitial loops is severely limited and those that do appear are effectively pinned by the second-phase particles. Thus a large excess vacancy concentration is largely prevented from appearing. Additionally the particles can act as recombination sites and thus contribute to a lowering of the overall point defect concentration. An excess annihilation of interstitials would cause a compressive stress system around the precipitate which would attract vacancies while repelling further interstitials. Bullough¹⁰ maintains that particles must be coherent to achieve this but the theoretical bases are not universally accepted. The recent GE-BRD data (subsection 2.2) suggest that coherence is not a prerequisite for inhibition of void formation although irradiation to higher fluences is necessary to demonstrate this. It should be noted that intragranular precipitation is most likely the only nonbiased sink for point defects. As already discussed, dislocations have a preferential attraction for interstitials. Grain boundaries would be nonbiased if vacancies and interstitials were equally mobile, but the higher diffusivity of the interstitials means that in net terms more interstitials than vacancies will "sink" at grain boundaries and thus contribute to the overall excess vacancy concentration.

4.3 WAYS TO INHIBIT SWELLING

The discussions in subsection 4.2 suggest the following techniques to inhibit or at least limit metal swelling:

1. *Produce a sufficiently fine second-phase constituent.* The Inconel-625 data cited in subsection 2.2 and the U.K. PE-16 data²⁴ demonstrate that finely divided second-phase particles can effectively limit swelling. In the case of the Inconel-625 at least, the particles appeared to perform two functions. First, they acted as recombination sites for interstitials and vacancies and thus lowered the overall point defect concentration.

Second, they were sufficiently close together to severely limit interstitial loop formation and subsequent dislocation movement so that vacancy supersaturation was minimized and the vacancy density was insufficient to nucleate voids.

The PE-16 behavior was somewhat different in that interstitial loops apparently were able to form and produce a complex dislocation network. However, the second phase particles effectively pinned these dislocations, prevented their movement and thus inhibited further preferential interstitial trapping by dislocation sweeping. The vacancy supersaturation caused by the interstitial loop formation was sufficient to form isolated voids but they did not grow because the pinned dislocations could not produce further free vacancies.

2. *Cold working.* This process has been shown to be effective in largely inhibiting swelling in Type-316 stainless steel for fluences to $\sim 5 \times 10^{22} \text{ n/cm}^2$, $E > 0.1 \text{ MeV}$.¹⁴ In this case the dislocation network is so complex as to be largely immobile and thus does not itself contribute to an excess vacancy concentration. Additionally the dislocation network keeps the structure compartmented so that excess vacancies arising from interstitial loop formation and impurity gases from transmutations are prevented from diffusing over long distances, so that the vacancy and gas aggregates necessary for void nucleation may be difficult to accumulate. The large unknown, of course, is whether the cold worked material will show a rapid rate of volume increase at higher fluences as suggested by Bullough¹⁰ and which the U.K. data can be interpreted as showing^{13,18} (Figure 2-15), or whether swelling remains minimal as the present HEDL and GE data show. This will be dependent on the ability of the dislocations to remain immobilized and thus depends on the time-temperature stability of the cold working. To date the stability of the cold work in the U.S. irradiated Type-316 has been remarkably good. By contrast cold worked Type-304 has shown large swelling values and TEM examinations showed that the material had recovered. Thus if the dislocation structure remains stable and immobile the swelling resistance should be maintained. This suggests excellent promise for swelling resistance in cold worked stabilized alloys in which the particles of TiC , NbC or other precipitates should effectively pin the dislocation network and inhibit its recovery at high temperatures. It is also noteworthy that the solid solution strengthening molybdenum may be of assistance in retaining cold work in Type-316. It is unlikely that total swelling resistance can be achieved in stabilized alloys without inducing cold work, because second-phase particles are probably too difficult to produce on the fine scale required.

The most promising swelling resistant alloys are: CW Type-321, CW Type-318, CW Type-316+Ti, and CW Type-316 austenitic stainless steels, and the nickel base alloys Inconel-625, Inconel-718, Inco-901 and Nimonic PE-16. With respect to swelling resistance, the degree of cold work in the austenitics is unlikely to be critical¹⁶ (although it must be as uniform as possible). From the ease-of-fabrication and mechanical properties viewpoints, a level of cold work in the range of 15 to 25% as measured by reduction in area would be preferred.

The nickel alloys are all precipitation hardening materials and must be aged prior to use to optimize the precipitate size and spacing. Inconel-625 and -718 both have high nickel contents of $\sim 65\% \text{ Ni}$ and are probably impractical for cladding alloys because of the sodium corrosion problem. However, they are excellent candidate materials for core structural elements. The -901 and PE-16 contain only $\sim 42\% \text{ Ni}$ and thus can be considered as long range potential cladding alloys; in particular their high strength properties could lead to savings on plenum lengths in nonvented fuel pin designs.

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