Model of Instability of Excess Vacancy System in Metals under Radiation

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Abstract – Swelling of metallic systems associated with the formation and growth of pores under irradiation by fast neutrons, electrons or ions, to a large extent limits the choice of materials for nuclear energy equipment, accelerators etc. Radiation swelling is explained at the qualitative level be the combining of separate, radiation-induced vacancies into pores. The magnitude of radiation swelling in metal is extremely sensitive to the type and concentration of the doping elements. When complex metallic systems are irradiated, segregation of elements occurs. This phenomenon is usually associated with the formation of the vacancy – impurity atom complexes. Diffusion movement of vacancies towards the surface in this case must lead to the segregation of elements.

1. Introduction

Presently available theoretical approaches to the description of the phenomena mentioned above and some others are obviously insufficient. The analysis of the available experimental results lets us make a conclusion regarding the formation of instability in the vacancy system. Instability manifests itself at high concentrations of vacancies [1]. Significant excess in concentration of vacancies of the equilibrium magnitude can be achieved be irradiation or tempering processes.

An assumption can be made that the existence of high concentration vacancies in metal creates the average macroscopic field of stretching elastic strains. In other words, average distances between atoms increase, which is equivalent to all-directional stretch.

In the first approximation a simplified relation between elastic strain σ with concentration of excess vacancies C_{vs} can be used

$$\sigma = A \cdot C_{vs}. \quad (1)$$

Index A is defined by the power of punctual defect and elastic constants of the material. The presence of elastic strains changes the chemical potential of vacancies.

$$\mu_x = \mu_{x0} + k \cdot T \cdot \ln C_{vs} - \sigma \cdot \Omega, \quad (2)$$

where Ω – atom volume. Diffusion flux of vacancies is defined

$$J = D \cdot \left(1 - \frac{\sigma \cdot \Omega}{k \cdot T}\right) \cdot \operatorname{grad} C_{vs} \quad (3)$$

where $D = D_0 \cdot \exp\left(-\frac{E_m - \sigma \cdot \Omega}{k \cdot T}\right)$ – autodiffusion index, E_m – energy of vacancy migration. From the expression (3) follows that at $\sigma \cdot \Omega > k \cdot T$ in the system of excess vacancies there forms an instability that is essentially a change in the direction of diffusion flux. The phenomenon of ascending diffusion in the region of instability can be reduced to the fact that a random increase in the vacancy concentration grows and becomes a pore.

Another important statement of the introduced model is the division of doping impurities into undersize and oversize according to whether they induce elastic strains of stretch or contraction in the matrix lattice. In this case doping changes the condition of instability. On the other hand, undersize impurity can behave as vacancy and lead to segregation of elements.

The introduced model of instability of excess vacancies in metallic systems serves as a base for a theoretical description and calculation of observed phenomena. Among such phenomena where a satisfactory correlation of the calculations with the experimental results the following can be listed.

2. Dependence of radiation swelling on temperature [2]

If the formation of a radiation pore is related to the ascending diffusion of vacancies, then the temperature-dependent part of the index of ascending diffusion must be taken as the function characterizing the temperature interval of swelling. Fig. 1, 2 show the comparison of the calculated temperature dependence with experiment.

3. Formation of a radiation pore [3]

Existing quasithermodynamic theory of the formation of a new phase cannot explain the formation of an embrionic pore bigger than critical. Within the framework of the introduced model this question is resolved in a natural way. The competing processes of ascending vacancy diffusion and the effect of the forces of surface strain are taken into account in the calculations.

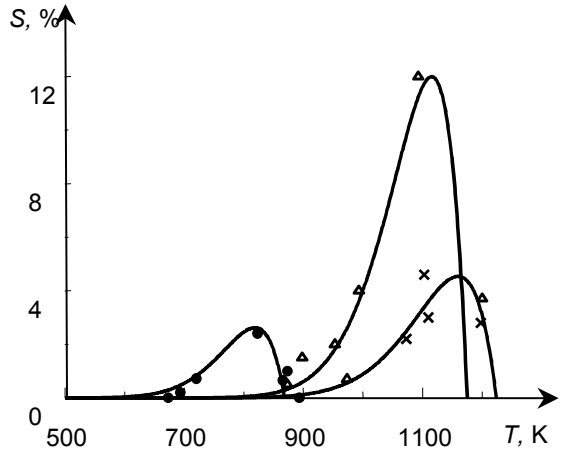

Fig. 1. Function characterizing the interval of radiation swelling, experimental results: • – Ni; Δ – Mo; × – Nb

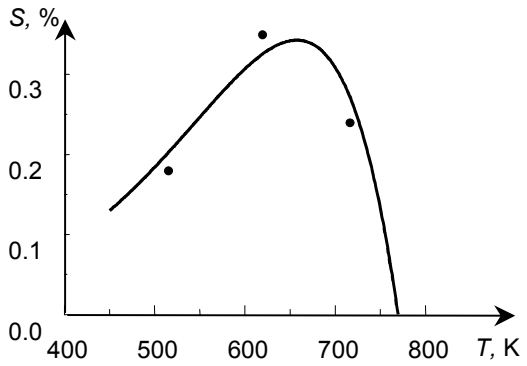

Fig. 2. Function characterizing the interval of radiation swelling for copper, • – experimental results

Fig. 3 shows the results of the calculations of vacancy clusters. When a cluster of vacancies becomesa pore, the latter's size is above critical.

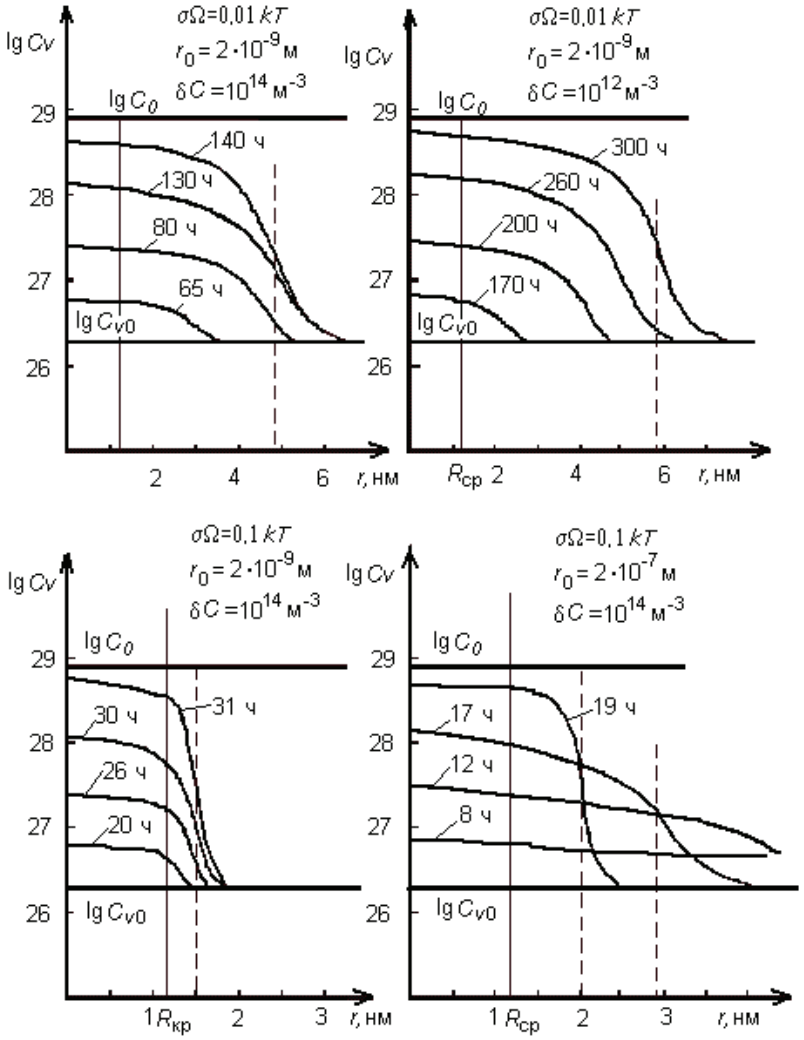

Fig. 3. Profiles of vacancy concentration in clusters at different moments of irradiation process

4. Concentration of radiation pores

Examination of diffusion fluxes forming a pore at given oversatiation allows to calculate the area of influence of a pore and subsequently their concentration. It was supposed that pores form simultaneously. Fig. 4 represents a comparison of the temperature dependence of pore concentration with experiment.

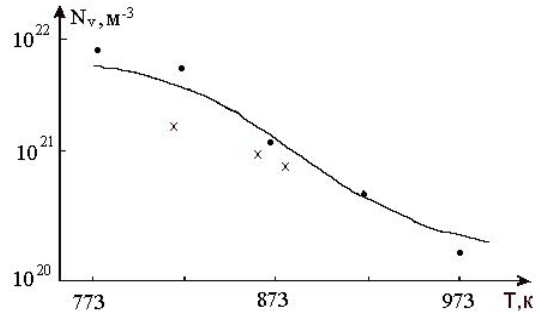

Fig. 4. Temperature dependence of pore concentration for nickel (experimental points: • – irradiation by ions Ni^+, × – reactor irradiation)

5. Parameter of superlattice of radiation pores

In the stationary case the generalized diffusion equation has a space periodic solution. A necessary condition is the presence of additional outflow channels. The periodical character of the equation solution allows the calculation of the constant of superlattice of radiation pores. Fig. 5 shows a comparison of the calculated temperature dependence of pore superlattice with experiment.

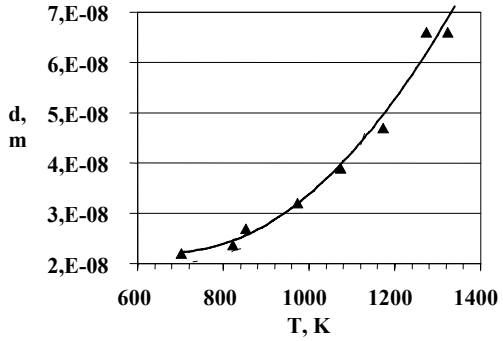

Fig. 5. Temperature dependence of parameter of pore superlattice: calculation at $C_{vs}=3.8\cdot10^{26}$ m^{-3}; ▲ – experimental results

6. Doze dependence of radiation swelling

The experimentally observed linear dependence of the degree of radiation swelling and irradiation doze is well described within the framework of the introduced model for metals with different types of crystal lattice. The additional assumption that in the vicinity of a pore the main outflow channel for interstitial atoms is the pore itself, is made in the calculations.

7. Effect of weak doping on swelling

Introducing impurities of various types into a metallic matrix as a rule lowers the intensity of radiation swelling. Calculations have been made for nickel with the addition of 1% doping element. The change in magnitude of elastic strains associated with the introduction of impurity, and mutual diffusion of atoms of matrix and impurity are taken into account. Fig.6 demonstrates a comparison of the calculated dependence of the disparity of lattices of matrix and impurity with experiment.

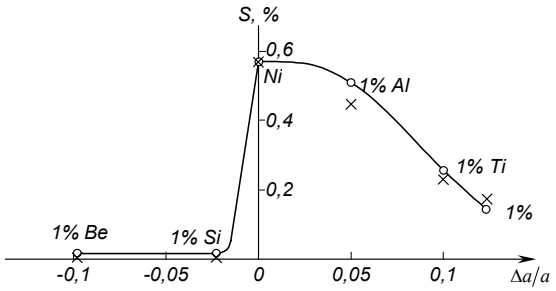

Fig. 6. Dependence between swelling of nickel-based binary alloys and dimensional disparity of atoms of the base and doping element [× – theoretical estimates, o – experimental results (Ni^+, 2 с. н. а., 848 K)]

8. Pore formation and segregation of elements in binary alloy

Alloys Ni-Si with different ratios of components are chosen for calculations. Variation of elastic strains and diffusion of undersize component – silicon was taken into account (Fig. 7, 8). It is shown that pores decorated by silicon form at low concentrations of silicon. At high concentrations of silicon pores don't form, and segregation of elements is observed. Calculations were stopped at the attainment of the composition of intermetallide Ni_3Si stechiometry, i.e. diffusion in the area of γ′ – phase was not considered. The rate of growth of the intermetallide phase was estimated to be ~10^{-11} m/s. Calculation results agree with the experimental data.

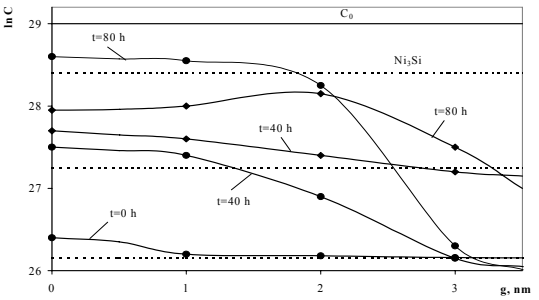

Fig. 7. Profiles of vacancy cluster (●) and concentration of silicon (♦) in alloy Ni+2%Si at different moments in time, Treg=800°C

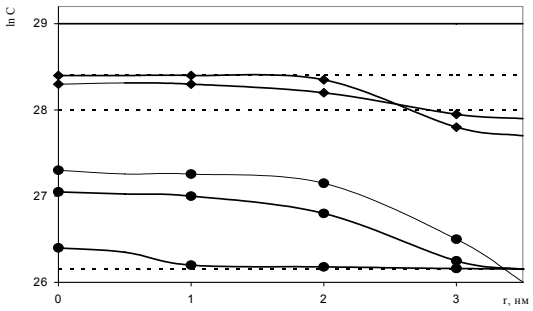

Fig. 8. Profiles of vacancy cluster and concentration of silicon in alloy Ni+10%Si at different moments in time, T_{reg}=800°C

8. Surface bursting of details after tempering

Kinetics of cooling of a detail during tempering is calculated. Thermostrains originating in this process are calculated. It is shown that in the proximity of the surface significant tangent strains arise and the instability condition $\sigma \cdot \Omega > k \cdot T$ is satisfied. Within the volume of the detail the release of contracting strains occurs simultaneously with the decrease in vacancy concentration. Kinetics of crack formation during tempering is calculated. Dependence between time of tempering before cracking of surface and temperature of tempering has the form of a curve with a maximum and agrees with experimental results.

As a result, a conclusion can be made that the model of instability in excess vacancies in metal can be used for the purpose of describing a wide range of observed phenomena. At present the model is used for describing SHS-processes, decay of solid solutions etc.

References

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