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DU COMBUSTIBLE NUCLÉAIRE AUX DÉCHETS : RECHERCHES ACTUELLES

FROM NUCLEAR FUELS TO WASTE: CURRENT RESEARCH

Metallic structural materials in the nuclear environment: some problems illustrating new methods

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Abstract The structural components of the nuclear industry are submitted to a number of aggressions, mechanical, chemical and physical (irradiation). As a consequence, the problem of durability and ageing of such structures is a key issue. The understanding of the phenomena involved implies the description and modelling of atomic scale events (irradiation point defects) resulting in fluxes of matter (diffusion under irradiation), in the dynamic evolution of structural defects (dislocation loops, cavities, ...), with major consequences on mechanical properties (yield stress, fracture behaviour), with, in addition, phenomena coupled between mechanical behaviour and chemical environment. It is therefore the totality of materials science which is involved in understanding the behaviour of metallic structural materials in the nuclear environment. The aim of the present paper is to illustrate some examples currently under investigation, and some of the new approaches involved in the understanding of mechanical behaviour (a scale transition from the atomic to the macroscopic). The input from large computer simulations as well as the value of simple 'back of the envelope' calculations, plus the need for cautious experimental studies will be illustrated. The theme of the ageing of materials, central to this paper, finds applications in many industrial situations. ***To cite this article: Y. Bréchet, C. R. Physique 3 (2002) 915–922.***

metallurgy / defects / irradiation / mechanical properties

Matériaux de structure métalliques dans le nucléaire : quelques problèmes, quelques nouvelles méthodes

Résumé Les composants structuraux de l'industrie nucléaire sont soumis à des sollicitations mécaniques, chimiques et physiques très agressives. En conséquence le problème de la durabilité et du vieillissement de ces composants est d'une importance majeure. La compréhension de ces phénomènes fait intervenir des mécanismes à l'échelle atomique (défauts d'irradiation), des flux de matière (diffusion et diffusion sous irradiation), le développement de défauts structuraux (boucles de dislocations, cavités), avec des conséquences importantes sur les comportements mécaniques (limite d'élasticité et comportement à rupture) et des couplages entre la mécanique et l'environnement. C'est donc l'ensemble de la science des matériaux qui est impliquée dans la compréhension et la modélisation de ces comporte-

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ments. On se propose de brosser un tableau de quelques questions actuellement à l'étude, du point de vue des changements d'échelle (de l'atomique au macroscopique). L'apport des simulations numériques aussi bien que les vertus de simples calculs de « dos d'enveloppe », et la nécessité d'études expérimentales soignées seront au centre de cet exposé. La problématique du vieillissement des structures ainsi présentée est importante bien au delà du domaine nucléaire et les recherches menées dans ce cadre peuvent trouver des applications dans de multiples domaines industriels. ***Pour citer cet article : Y. Bréchet, C. R. Physique 3 (2002) 915–922.***

métallurgie / défauts / irradiation / propriétés mécaniques

1. Introduction

Metallic structural materials in the nuclear industry, and especially the steels used in the main components apart from the very core of a nuclear reactor, present a number of specific features which have motivated, in the last decades, numerous research programs. The initial driving force for the studies of materials' behaviour under irradiation was to build nuclear power plants in the first place [1]. However, with the increasing age of the production units, and the problem of waste management, the key concept has become the ageing of materials under irradiation [2,3].

The environment under which structural materials are operating is very aggressive: it is aggressive in terms of temperature (leading to creep, to precipitation evolution), in terms of chemical interactions (corrosion, oxidation), and in terms of irradiation damage. Operating a nuclear power plant safely requires, in principle, a proper understanding of the properties' evolution with time, just like designing a safe turbine reactor for an airplane requires a good evaluation of the damage due to oxidation or creep. In case of doubt, the lazy way to ensure security is the 'safety factor' which amounts to overdimensioning the structures, or to replace the components earlier than really needed. For airplanes, the trend toward weight saving has more and more disqualified the use of comfortable safety factors. For nuclear power plants, the need to minimize the amount of radioactive waste leads to the same tendency: more and more it will be necessary to account for the time evolution of properties with a sufficient accuracy, and that cannot be done by purely empirical knowledge. A deep understanding of the underlying mechanisms is required in order to derive reliable 'constitutive laws' able to predict materials' behaviour for durations much longer that the duration of standard experiments. The aim of this paper is to present some recent developments in this direction, and the conceptual tools needed for a safe operation of nuclear power plants in the long term.

2. Basic steps for the understanding of the mechanical behaviour of metallic alloys in a nuclear environment

A number of phenomena specific to irradiation damage with matter are now well identified [4–8]. Irradiation is known to shatter the very basic features of matter: it can destroy the crystalline order, it can introduce structural point defects whose migration will lead to mass fluxes and possible segregation of chemical species, it can even change the chemical composition of an alloy through atomic transmutation. The variety of metallurgical phenomena which may occur under irradiation and lead to a microstructural evolution is far beyond the scope of the present paper (for a review, see for instance [7,11]). It will be enough for us to remember that the basic phenomena occurring under irradiation take place at the atomic scale (a few Angströms).

These microstructural events at the atomic level have consequences at the level of the substructure of the material: the state of precipitation can evolve (accelerated coarsening or dissolution of precipitates, appearance of out-of-equilibrium phases), the initial dislocation substructure may be thoroughly modified (creation of dislocation loops, evaporation of the dislocation network, ...), inner cavities (voids or gas bubbles) may appear. These evolutions take place in the range of scales between 10 nm to 10 µm, and this evolution in the mesostructure is crucial to the understanding of the consequences of irradiation on the plastic behaviour of metallic alloys.

The dislocations responsible for the ability to deform plastically (they are the carriers of plasticity just as electrons in metals are the carriers for electricity) will interact strongly with this mesostructure. The defects created will impede their movement, therefore the ability to deform plastically will decrease, and one expects qualitatively that the alloys will become harder and more brittle.

Assuming we understand this modification of the behaviour of dislocations due to the presence of this microstructure, our problem is far from being solved. The macroscopic plastic behaviour of the sample is a consequence of the collective behaviour of dislocations interacting with a population of irradiation defects. Therefore a key issue is on one hand to understand the collective behaviour of the dislocation population, and on the other hand to understand the collective behaviour of irradiation defects leading to the formation of the mesostructure.

Once these two problems are solved, we can hope to understand the mechanical behaviour of a grain of alloy. A sample on which mechanical tests are performed is never a single grain: it is a polycrystal formed by a collection of grains loosely packed along grain boundaries. Since metals are crystalline, their plastic behaviour depends on the respective orientation of the crystal reference frame and of the loading directions. The different grains will deform differently and thus the overall behaviour will depend not only on the behaviour of each grain, but also on their interactions.

Then we come to the understanding of the behaviour of a 'real sample', at the millimetre scale. However, the story is not over .... A real component in a nuclear power plant has a complex geometry, loadings in terms of temperature and stress fields are not homogeneous. Possible fracture of the component depends on the presence of defects in the materials and is thus probabilistic in nature. In the previous steps, physical metallurgy was the discipline in charge, now mechanics has to take over. The local approach to failure developed in the French school of fracture mechanics has provided a powerful tool for the transition from the local behaviour of the material to the macroscopic behaviour of the component.

The key issues specific to metallic materials in a nuclear environment are the transition from the mechanisms occurring at the atomic level (the very cause of irradiation damage) to the materials' behaviour at the scale of the grains. The problems posed by the transition from the grain behaviour to the component are far from being all solved, but they are not specific to nuclear mechanical metallurgy (for a recent review of the question, see [4]). In the next section, we will illustrate by an example the conceptual tools available for the understanding of irradiation effect on plastic behaviour.

3. An example of scale transition from the atomic level to grain behaviour: Irradiation hardening

Fig. 1 shows strain curves of a 316 stainless steel after irradiation at 20 °C. The main effects are a very large increase of the yield stress (multiplied by 2.5), a decrease in the ductility (divided by a factor 4) and a pronounced softening which is very likely to lead to a very damageable strain localisation.

The alloy 316 is a complex material. In order to understand the mechanisms, it is necessary to study model materials. The different steps in the understanding reflect the 'scale transition' outlined in the previous section.

- Irradiation damage takes place at the atomistic level, and the atoms are removed from their sites on the crystalline lattice. The mechanisms are collective effects, and the modelling tool adapted to this problem is Molecular Dynamics (MD) [9,10]. It allows, for a given input energy, to predict the amount of free vacancies and interstitial created. It also identifies more complex defects resulting from the interactions between the point defects. The key step here is to have a reliable interatomic potential, and the progress of ab-initio methods in recent years are now on the way of providing such potentials, both for pure materials and for simple binary alloys.

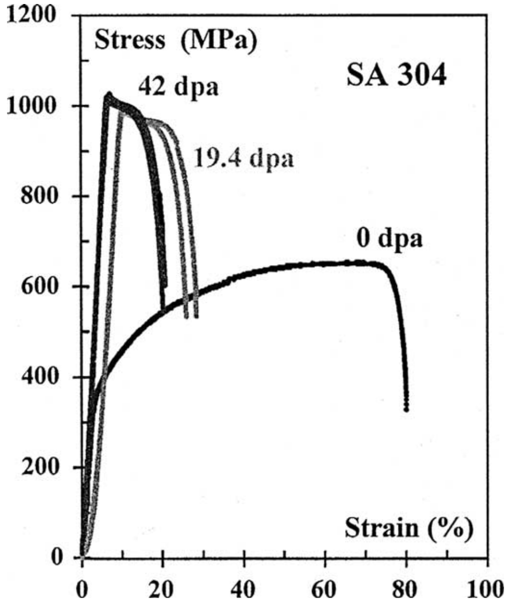

Figure 1. Stress strain curve of an irradiated Stainless Steel with different doses (expressed in dpa, displacement per atom) (courtesy of C. Pokor, P. Dubuisson, J.P. Massoud).

- The point defects created by irradiation are mobile (diffusion) and they interact between them, and with the other structural defects (dislocations, grain boundaries, ...). They tend to coalesce into dislocation loops which become strong obstacles to the motion of other dislocations. Modelling this phenomenon is done using 'cluster dynamics' [11]: a population of interstitial or vacancy loops (a cluster of n defects) evolves by absorbing or rejection a point defect. The master equation ruling this system (taking into account the bias of each cluster for each type of point defects) is solved either discretely, or using the continuous version, or the Fokker Planck equation. This model allows one to predict the size and densities of loops for a given irradiation dose and a given irradiation temperature. The comparison between predicted quantities and TEM observations on irradiated materials allows one to identify the adjustable parameters of the model (whose magnitudes are known from physical arguments) and to test its predictive ability. A key issue here is to have the experimental observations allowing for this comparison: this requires irradiation reactors and special TEM equipment which are crucial to a serious study on irradiation effects.
- When the microstructure resulting from irradiation point defect interaction is well described, the pinning efficiency for other moving dislocations has to be investigated [12]. When the Frank loops are large enough (more than 10 nm) the classical elasticity theory of dislocation-dislocation interaction is sufficient: it allows one to calculate the pinning strength of the defect, and the theory of collective pinning of a dislocation by obstacles, well developed for precipitation hardening, applies directly, allowing to predict the evolution of the yield stress as a function of the loop population. When the loops are much smaller, of the order of the dissociation width of the dislocations (1 nm), elastic theory is dubious, and again the Molecular Dynamics technique is well suited [13]: it allows one to calculate the pinning strength of the obstacle, but also to show its mobility; the effect of irradiation defects is not only to pin the dislocations, but also to make their motion more sluggish. The moving defects can also be annihilated by the moving dislocation. An additional subtlety is the possibility of defaulting the Frank loop, and to decrease the pinning strength of this obstacle that way. From these two mechanisms, it appears that the pinning efficiency of a population of irradiation defects decreases while deformation proceeds: this is the physical origin of the strain softening observed experimentally in Fig. 1.
- However, experimentally, it is observed that the strain softening effect appears only when the irradiation dose exceeds a threshold. This comes from the competing effect of work hardening

(a strained material hardens due to dislocation interactions) and the sweeping out of irradiation defects. In order to quantify this effect, it is necessary to couple the classical analytical work hardening theories (which in essence are nothing but 'chemical kinetics' of a dislocation population) [14] with the theory of hardening by irradiation defects. The parameters introduced in this coupled analytical model (which describe the evolution of dislocation density and irradiation defect population when deformation proceeds) are the ones identified in the molecular dynamics simulations. This is very typical of scale transition in materials science: the purpose is not to 'compute a bridge' right from Schrödinger equation, but to use as an input to a model the outputs of a model at a smaller scale. At this step, again with an experimental comparison, one is able to describe the stress strain behaviour of an irradiated material.

- This behaviour is to be understood as a prediction at the grain level: the transition to the polycristal is delicate [15]. Current models for this transition are either analytical (Self Consistent model) but they assume that deformation is homogeneous within a grain, or they rely on a multicrystal description using FEM codes coupled with crystal plasticity. The work softening behaviour leads to numerical

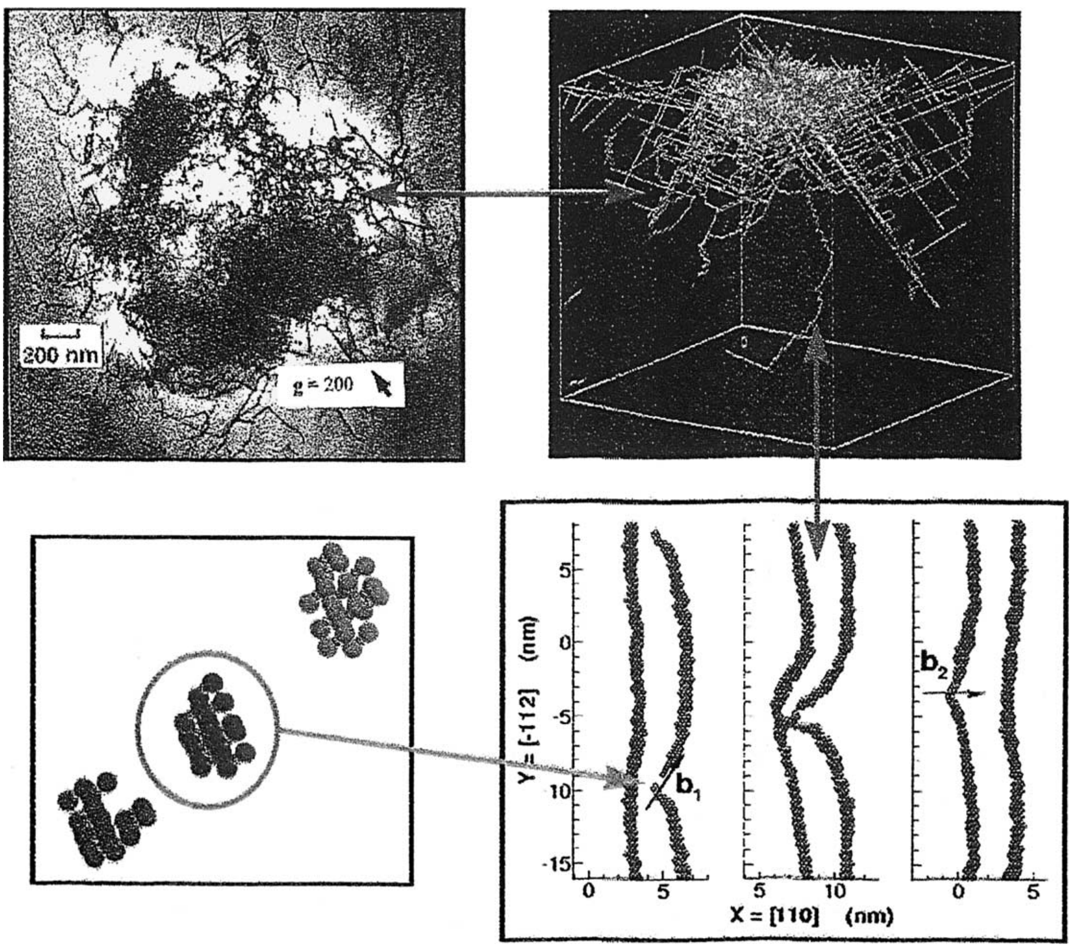

Figure 2. Scale transition in irradiation hardening (see text for details) (courtesy of D. Rodney, M. Fivel, C. Robertson, G. Martin).

instabilities difficult to deal with in FEM models, and the strain is heterogeneous, which contradicts the basic hypothesis of self-consistent models.

- The need to measure mechanical properties on small materials quantities has led to a renewed interest for nanoindentation techniques [16]. The modelling of these tests at the dislocation level has been made possible by the development of intensive computer simulations for a population of interactive dislocations [17,18]. Dislocations are non-conservative, out-of-equilibrium linear defects with long-range interactions. The 3D computer simulation of dislocations has been a major challenge of physical metallurgy in the last decade, and the strains and sample sizes reached are still deceivingly small. But the tool is ideal for modelling a nanoindentation test. Each dislocation moves when the local stress reaches a threshold, and this threshold can evolve with strain. Implementing in the 3D dislocation code local rules for dislocation nucleation under the indentor, and the constitutive behaviour predicted for the dislocation/irradiation defects interaction, allows one to predict accurately the loading curve of an irradiated Cu. The TEM observations of the imprint reveal the dislocation structure and these observations can also be compared, at least qualitatively, with the simulated structures.

Fig. 2 schematises this 'scale transition exercise': from the MD simulation of irradiation induced point defects to the modelling of the indentation test. It is worth noting in this example that the tools used range from intensive atomic simulations relying on advanced solid state physics to develop the interatomic potentials, to the classical theory of dislocation, or the analytical phenomenology of work hardening. Intensive computer simulations, as well as the back of the envelope calculations, contribute to the modelling in these questions.

The research used to illustrate this strategy for understanding macroscopic behaviour of irradiated materials relies on a strong coupling between computer simulations and experimental observations. Materials suitable for this comparison are pure metals such as Nickel, Copper. The 316 alloy is of course beyond the reach of these methods, but the elementary mechanisms identified in simple materials are thought to be relevant to the more complex systems. Additional effects, such as irradiation induced segregation of chemical elements, irradiation driven precipitation, also contribute to the hardening behaviour, and the tools to model these phenomena are also available, but beyond the scope of the present paper.

4. The need for understanding, and the intrinsic difficulties of nuclear materials

From what has been written above, it seems pretty clear that the understanding of the evolution of mechanical properties under irradiation damage is far from being a resolved question. The situation with respect to fracture behaviour and coupling with environmental aggression (stress corrosion cracking) is even more confusing. The embrittlement of austenitic stainless steels is often attributed to the artificial ageing of ferrite, but the relative contribution of the increased yield stress, of the decreased work hardening, and of the strain localisation are still to be quantitatively understood. The relation between grain boundary effective viscosity and corrosion crack propagation in NiCr alloys is well established, but the mechanistic interpretation is still missing. In a word, we need far more than we know at present in order to have an exhaustive understanding of the effects of irradiation on mechanical properties of alloys. In front of these difficulties, a radical solution is to forget about microscopic understanding and to focus on a mere phenomenological description of the effects. Since the engineer apparently needs only this level of description, one may then wonder about the real need for a basic understanding and modelling of irradiation effects . . . .

Even from an engineering viewpoint (not to speak of intellectual satisfaction), this strategy seems totally flawed and inoperative for practical purposes. We have insisted in the introduction on the necessity of a sound understanding of the mechanisms to derive laws which can be extrapolated safely for longer times, higher temperatures, more intense irradiation. Another equivalent view point is the validation (or the invalidation) of accelerated testing procedures. To qualify a materials for use during 50 years, one definitely needs more rapid testing procedures: increasing temperature, or irradiation flux is a method commonly used. It is reliable only when it has been proven that the mechanisms are similar to those operating in real functioning conditions. In addition, irradiated materials are difficult to manipulate, and as a result, ion or electron irradiations are often used to get access to irradiated materials with similar damage. Again, a comparison with materials irradiated in real conditions, as well as a fundamental understanding of the mechanisms is necessary to ensure the validity of such experiments. Irradiated materials are all the more inconvenient since they are in larger quantities: the mechanical testing is extremely expensive and difficult to perform (the very limited amount of available data for toughness of irradiated materials is a consequence of these difficulties). It is necessary to develop testing procedures on small quantities of matter, such as an indentation test. Again, modelling is necessary to extract from the results the constitutive behaviour which will be used in FEM codes for dimensioning macroscopic components.

As Jean Rostand said: “Attendre d’avoir tout compris pour faire, c’est se condamner à l’inaction”. Fortunately, nuclear power plants have been safely designed before the behaviour of materials under irradiation has been understood. A number of phenomena (such as the swelling of the fuel) have been discovered ‘on the spot’, and empirical solutions and design rules have been found. The problems listed above, in spite of many years of research and very significant progresses in the understanding as well as in the experimental investigations are far from being solved. However, they are the key steps toward a more efficient design, for the long term life, of nuclear power plants.

5. Conclusions

The key issues emerging from the examples in physical metallurgy of structural metallic nuclear materials are the need for a ‘scale transition’ to understand the macroscopic mechanical properties, and the need for a clear understanding of the kinetic path for structure evolution in order to be able to predict ‘long time behaviour’. The specificities of radiation damage has led nuclear metallurgy to start from the atomic level much earlier than in other fields of application. Conversely the meso/macro transitions have received a considerable interest, for instance in aeronautic or automotive materials. It is timely now to benefit from the two cultures, and gain from a cross fertilisation between these various industrial domains. The remarkable development of computer simulations at different scales, and the emerging possibility to couple a number of them, are very promising for the future, provided the difficult experiments required to validate the models are developing at the same path.

Let us illustrate the possible input of ‘nuclear metallurgy’ in other fields of application by two recent examples. The developments of the local approach to fracture in the study of nuclear materials are now being applied to optimise design of aluminium alloys for aircraft structures [19]. The studies of phase separation by Monte Carlo simulations have led to important discoveries for the first stages of precipitation of Niobium Carbides in steels [20]. These two very recent examples illustrate that the ‘French exception’ in nuclear metallurgy may also be an asset in other industrial domains.

On the pure research field, the scientific questions raised by the understanding of irradiation effects on microstructure evolution and on mechanical properties open or re-open a number of issues in fundamental materials science (statistical physics of driven systems, atomistic mechanisms of dislocation thermally activated motion) [7].

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