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PHYSICAL METALLURGY RESEARCH LABORATORIES

A SYMPOSIUM ON THE IN-REACTOR CREEP OF ZIRCONIUM ALLOYS AND RELATED TOPICS, PINAWA, MANITOBA, 13-14 MAY, 1975

by

M. J. GODDEN

This symposium was intended as a forum for the exchange of current information on zirconium creep with a view to understanding and possibly alleviating the current and near-future problems associated with this phenomenon in on-line reactors and also to point out the principal features of the phenomenon to designers of future reactors.

John Dunn of Power Products gave the initial talk on reactor design aspects of zirconium reactor creep. He mentioned the designer's dilemma of the exceptionally long time from starting work to actual in-reactor experience with these alloys but in the meantime further reactors must be designed. Reactor operators are learning from experience; the recent heavy water leaks at Pickering 4 were located in a few days by acoustic emission compared to the several weeks needed to find the earlier leaks in Pickering 3. There are four areas to be considered with zirconium creep in reactor design:

(a) Diametral creep of the pressure tube. In the low temperature regime (below 280°C) of the earlier reactors, results predict 4.2% creep in 30 years. However these results do not apply to Zr-2.5% Nb or to the higher temperature now used (now at 312°C and rising). The pressure tubes are designed for strength, which is obtained by cold work, not for creep. To allow for creep in the design, designers are now having to assume an 11% strengthening due to irradiation.

Diametral creep will eventually lock-up the garter spring spacers. Although these spacers only have marginal strength now, future designs call for smaller spacers.

(b) Pressure tube axial creep. This appears to be a major problem although it has only been under intensive investigation for the last two years. Current designs call for 6 in. of creep of the fuel channel, but not all channels behave the same. If a 10% design factor on this 6 in. is allowed, the reactor becomes correspondingly bigger and much more heavy water is required. Bearing rings and bellows must be much longer (3.6 in. compared to the current 0.6 in.) and these cannot be manufactured in these sizes at present. The feeder pipes become more highly stressed and now pre-sprung alloy steel pipes will have to be used. Fretting is also a problem as the channels creep.

(c) Calandria tube axial creep. This aspect had been ignored until recently and there is no design basis for this at all. There is now some evidence of this phenomenon at Pickering 1. There is 1 in. available for calandria axial creep in the 600 MW design and only 0.75 in. in the Bruce design. In some respects it compensates for fuel channel creep.

(d) Creep sag of the whole reactor assembly. This appears to be a potential problem at Bruce in a few years. To avoid two tubes touching, four spacers instead of two are now being used.

Another of the current problems is at Douglas Point where the tubes are loading the end shield which is forcing on the concrete wall. The operators are now waiting for cracks in the concrete. A similar problem of the tubes loading the end shield is being experienced at Pickering 1. Despite these problems, the building of Bruce A is going ahead because the power is needed, although various of the above problems are likely.

Two papers were then presented attempting to fit current in-reactor creep data to empirical equations, basically modifications of the equation from CNRL-49 [$\dot{\varepsilon}_t = k.10^{-25}\ \sigma_t.\ \phi.$ (T-160)]. Ted Ibrahim showed that even after 23,000 hr, a steady state did not appear to have been reached, making any fit to this equation difficult. The effect of cold work on creep is not well documented but it appears that cold working Zr-2.5 Nb before ageing slightly increases the creep rate compared to the unworked condition. As results are obtained for longer times it appears that the results for Zircaloy 2 and Zr-2.5 Nb are tending towards each other and the better creep rate of Zr-2.5 Nb is only an initial transient. Ted Thexton is attempting to extend the equation to wider ranges of temperatures, fluxes and stresses, using the versatility of WR-1. Reasonable agreement has been obtained for the centre of the tube particularly at lower temperatures and neutron fluxes. There is still poor agreement towards the end of the tube, however. Pressurisation of the gas annulus up to 1 MPa is being investigated in an attempt to reduce the creep rate. End restraint is also being investigated and here a spring restraint appears better than a hard stop.

Coleman discussed failure of zirconium alloys by either reaching a tertiary creep condition or by fracture. Failure by fracture does not seem likely as there is no evidence for porosity or internal wedge cracks. For Zircaloy 2, only high stresses (of the order of 100 MPa) will produce a stress exponent large enough to produce necking and hence tertiary creep. Thus under the normal stresses experienced strains of 10% are possible without necking. There is less information on Zr-2.5 Nb but a similar trend is expected. While this work has been done in the laboratory under uniaxial stress, in-reactor creep is biaxial and thus the reported results may be optimistic.

Bill Fidleris discussed some in-reactor creep tests particularly with respect to the effect of flux on creep rate. There appears to be a temperature dependent effect, with three temperature regimes (a) below 450 K (b) 450 to 800 K and (c) above 800 K. The effect of flux on primary creep is small.

Stewart McEwen quickly summarised all theories of the flux enhancement of climb. He divided the theories into two groups after saying that thermal spike and loop alignment theories appeared somewhat irrelevant as they don't agree well with the practical results. He examined the yielding creep theories originally due to Cottrell but further modified by Hasketh and also Gittus. He was particularly critical of the Gittus model in which he claimed the meaning of the symbols used changed as the theory developed. He also examined the climb and glide models in which the rate controlling step is flux enhanced climb. Basically these theories appear to have some relevance (although he admitted to having a strong bias). Here the flux introduces new obstacles and produces climb which produces recovery and allows negotiation of the obstacles. The major problem with such theories is the requirement of a strain-producing step and it was on this score that the Heald model was criticised as being inadequate. He declined to expand much on his own theory based on this type of model.

John Jonas then discussed some aspects of the classical type of empirical creep equation. He pointed out that a major problem seemed to be that the activation energy term was not a constant as generally assumed but was stress dependent and could also be influenced by alloying. He felt that the value of more significance should be the activation energy at zero stress which would have to be obtained by extrapolation.

In a second paper Bill Fidleris discussed in-reactor growth of zirconium alloys in the absence of stress. Earlier work had suggested that interstitials migrate to the prism planes causing growth and vacancies migrate to basal planes causing shrinkage.

However, this is now seen as only part of the explanation. With cold-worked material, growth is linear in the longitudinal direction, but reaches saturation in the transverse direction. Annealed material shows saturation at about 0.1% strain which is all recoverable on post-irradiation annealing. All of the growth of cold-worked material is not recoverable by annealing. Frequently initial transient behaviour does not relate to final behaviour and makes the interpretation of results more difficult. A particularly interesting observation was that it had recently been observed that there was a factor of two difference in the growth rates of nominally identical tubes from different manufacturers.

Craig Simpson of Ontario Hydro described how they were intending to examine creep of zirconium in producing reactors by examining the cladding used for the cobalt adjuster rods used to flatten the flux at the centre of the reactor. As these are only in the reactor 18 months then discarded, it should be possible to examine many variables on the growth in that time. However the stress on these is small (about 500 psi) and the temperature only 70°C. He felt that the flux should be only slightly less than for the pressure tube as the structure is quite open around these cobalt rods. He also gave an explanation of how creep of the pressure tube was causing tensile stresses in the calandria tube due to the floating end design of these reactors. At present there is only 1/4 in. adjustment to allow for this before a hard stop is reached.

Jack Northwood then presented some transmission electron microscopy observations on irradiation damage. He showed the individual effects of neutron fluence, temperature, alloying and stress. The loops formed appear to be about 60% interstitial and 40% vacancy, and have a habit plane close to the prism planes. The loops tend to be in rows which are parallel to the basal planes. The growth strains possible for the observed damage are very small, about 0.02%, compared to the 0.3% actually observed.

Graham Carpenter then attempted to relate these observations to the theory of growth. He examined Buckley's model for the growth of uranium and showed that in many respects the growth of zirconium was not similar. Growth seems to be due to the annihilation of defects at various sinks. Interstitials are attracted to edge dislocations by the surrounding stress fields and vacancies migrate to grain boundaries and screw dislocations. Thus, the growth rate is reduced as the grain size is reduced. Using these assumptions, the predicted growth rate is an order of magnitude too small, so probably the vacancies are short-circuited to the grain boundaries.

Dave Faulkner then spoke about simulating irradiation damage and what could be learned from this. He showed that many parameters could be better controlled by simulation outside the reactor and direct observation of the production of damage was possible. In the high-voltage electron microscope, electrons can be used to create damage. However, even at 10^6V one electron will only displace one zirconium atom so cascade damage is not produced. To produce cascade-type damage bombardment with heavy ions is used. In order to produce voids in the high-voltage electron microscope, pre-injection with an inert gas is necessary. Recent studies of irradiation damage of Zr_3Al has shown that at room temperature and at a dosage equivalent to 1 displacement per atom this material disordered. There is less disordering as the temperature is raised and probably none above 320°C.

Akhtat then spoke on the creep of zirconium single crystals. This was similar to the presentation given at PMRL earlier this year and will not be repeated here.

Mike Luton then spoke about the correlation of high-temperature strength with stacking fault energy in a series of zirconium-tin alloys. These temperatures were higher than experienced in reactors and it was disputed that the strength measured was related to the stacking fault energy on the basal planes. It was felt by some that the stacking fault energy on the prism planes should have been measured.

Keith Nuttall then discussed the effect of hydrogen on the creep of zirconium during thermal cycling. A permanent strain of 0.1% per cycle can be obtained on a zirconium-200 ppm hydrogen alloy by cycling from room temperature to 425°C at a stress of 5000 psi. The hydrides tend to orient themselves perpendicular to the stress axis. The strain increases linearly with the number of cycles. In the absence of hydrogen there is no strain. The effect is attributed to the volume change on hydride precipitation at low temperatures and its re-solution at high temperatures. He said that the anisotropic thermal expansion of zirconium was a contributing factor to this effect, although to this writer there is no obvious physical explanation since the effect is absent in the absence of hydrogen. He claimed that the strain could be fitted to a simple formula due to Greenwood and Johnson relating this to the volume change on precipitation, applied stress, and yield strength of the hydride. The original formula was derived to explain growth due to an allotropic transformation and its application to the present case was not theoretically sound, although it provided a suitable empirical formula.

Ivan Dickson spoke of the strain-ageing effects observed in zirconium and some other materials. He showed many results most of which could be explained satisfactorily by a Snoek ordering mechanism of oxygen atoms for zirconium.

Ian Ritchie then discussed some internal friction results for zirconium single crystals. He outlined the technique and said that the results could only be very tenously related to the creep of zirconium. Solute-interstitial and interstitial-interstitial interactions could be examined individually by measuring damping under various deformation modes. On doping zirconium with praseodymium the oxygen internal friction peak disappears; this may be caused by oxide formation. He showed some very complicated results for which he had little explanation except that the dislocation pinning points were moving with time. Clarification of these results awaits further experimental work.

This aymposium was in large part a success because it brought together the usually-divided communities of power generator designers, Ontario Hydro engineers, AECL research and development workers, and university researchers.

In many of the discussions the initial reactor problems associated with creep were seen as only the beginning of much more severe problems in the future. Although AECL metallurgists had been warning of these problems for some time, only in the last few years had these warnings been heeded. Many of those present were surprised at the frankness with which the designers discussed their problems. While it was not expected that a panacea for zirconium creep would be found by consensus, it was generally agreed that more communication between the various groups, such as this symposium had provided, was necessary. Also the general approach to the situation was the correct one, i.e., in the short term to modify existing empirical formulae to more accurately describe the phenomenon over the widest range of parameters for the designers to use now; for the longer term to continue to advance the more academic approach so that the equations used may have a sounder basis. Overall it seemed that this problem would involve many people at considerable expense for a long period of time.

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