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Thermal creep properties of Ti-stabilized DIN 1.4970 (15-15Ti) austenitic stainless steel pressurized cladding tubes

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HIGHLIGHTS

- Creep rate and time to rupture of over 1000 tests on different heats are presented.
- Creep rate has been fit with a modified sinh correlation.
- Time to rupture was fit with the Larson-Miller parameter separately for a low and high temperature regime.
- Time to rupture and creep rate were correlated with the Monkmann-Grant relation.
- Low temperature data is compared to 316L data, high temperature data to D9 dynamic recrystallization data.

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ABSTRACT

This paper presents a large database of thermal creep data from pressurized unirradiated DIN 1.4970 Ti-stabilized austenitic stainless steel (i.e. EN 1515CrNiMoTiB or "15-15Ti") cladding tubes from more than 1000 bi-axial creep tests conducted during the fast reactor R&D program of the DeBeNe (Deutschland-Belgium-Netherlands) consortium between the 1960's to the late 1980's. The data comprises creep rate and time-to-rupture between 600 and 750 °C and a large range of stresses. The data spans tests on material from around 70 different heats and 30 different melts. Around one fourth of the data was obtained from cold worked material, the rest was obtained on cold worked + aged (800 °C, 2 h) material. The data are graphically presented in log-log graphs. The creep rate data is fit with a sinh correlation, the time to rupture data is fit with a modified exponential function through the Larson-Miller parameter. Local equivalent parameters to Norton's law are calculated and compared to literature values for these types of steels and related to possible creep mechanisms. Some time to rupture data above 950 °C is compared to literature dynamic recrystallization data. Time to rupture data between 600 and 750 °C is also compared to literature data from 316 steel. Time to rupture was correlated directly to creep rate with the Monkman-Grant relationship at different temperatures.

1. Introduction

Austenitic stainless steels have long been a favored choice for fast reactor cladding material due to good creep resistance, high-temperature mechanical strength and ductility, and established fabrication technology. In the early prototype sodium-cooled fast breeder reactors, AISI 316 was commonly used as fuel cladding material [1]. It was soon discovered, however, that AISI 316 is highly susceptible to void swelling under fast neutron irradiation and from the mid-1960's, various research programs were conducted to improve its swelling resistance. The German-Belgian-Netherlands consortium (DeBeNe) studied initially Nb-stabilized alloys. The development by Sandvik (Sweden) of a Ti-stabilized austenitic steel (12R72HV) with high creep resistance, and indications of limited swelling and He embrittlement under neutron irradiation [2,3], triggered the interest to include also Ti-stabilized cladding materials in the research program. Around 1970, four candidate fuel cladding steels were investigated: Nb-stabilized DIN 1.4988, DIN 1.4981 and DIN 1.4961 and Ti-stabilized DIN 1.4970. From the mid-1970's, it was well-established that the Ti-stabilized DIN 1.4970 had

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a superior swelling resistance compared to the other alloys. Further work was from then on focussing on metallurgical optimization: degree of cold work and annealing times and temperatures to further reduce irradiation swelling [4–9]. In other countries, similar efforts were made to develop swelling-resistant austenitic steels, leading to quite similar results: 15-15Ti/AIM1 in France [10–12], D9 in the US [13] and India [14], and JPCA/PNC316 in Japan [15–17]. A review of the historical developments of the DIN 1.4970 steel as structural material for fuel elements has been published by Bergmann et al., in 2003 [9].

When Ti is added to the steel as stabilizer, it forms fine TiC precipitates and thus effectively binds the free carbon. The absence of dissolved carbon eliminates the formation of chromium carbides and hence reduces the corrosion sensitization of grain boundaries. Under appropriate annealing conditions, fine, nm-sized TiC precipitates form which act as defect recombination centers during irradiation and, together with other minor alloying elements, such as Si, proved to be beneficial against irradiation-induced effects, in particular void swelling [8,11,18,19]. The fine tuning of the alloy composition led to gradual improvements of its behavior under irradiation.

Fuel pins made of austenitic stainless steel have been irradiated to dpa level exceeding 150 dpa without failure [20] but maximum dose for safe operation is probably limited to 120 dpa (embrittlement limit due to excessive swelling) [21].

The excellent resistance to radiation-induced swelling of ferritic/martensitic steels and ferritic oxide dispersion strengthened alloys [22–26] makes them appealing when very higher neutron doses (>120 dpa [27]). Compared to the stabilized austenitic steels, the ferritic/martensitic steels or ODS alloys still face serious challenges for widespread use: limited creep resistance for the former, difficult fabrication and limited experience for the latter. Additionally, they may experience liquid metal embrittlement in certain coolants such as lead-bismuth [28]. It is therefore not surprising that optimized austenitic steels remain the materials of choice for the first cores of several Gen IV reactor projects [27], hence the current renewed interest in the knowledge accumulated on these steels.

For reactor designers, reliable creep correlations derived from actual cladding tubes are essential to define safe operation margins. The experimental data set should furthermore be sufficiently extensive to account for heat-to-heat variations and to cover the inherent scatter encountered in creep experiments. Under the DeBeNe consortium, more than a thousand biaxial creep tests were performed, spanning two decades of work. Yet, the results were in danger of being lost: their records only existed in a few paper archives. The Interatom Data Base is also no longer available. Some data was preserved in an online database maintained by the European Commission [29]. To avoid repeating long-term, costly experiments and to preserve what has been done in the past, it is of great importance for the nuclear community to revive existing datasets while they are still available. It is the purpose of this paper to present and review the existing datasets on creep properties of DIN 1.4970 cladding tubes in the unirradiated condition obtained by the DeBeNe consortium during the development of KNK II, SNR-300 and SNR-2/EFR reactors.

2. Material and methods

2.1. Thermo-mechanical treatments and microstructure

More than 40 heats of the 1.4970 steel have been produced [30] by different manufacturers during the DeBeNe R&D program to manufacture tubes. Other products such as bars and plates were also produced [31] but the present work only focuses on tubes. The steel was generally prepared by melting scrap steel in an induction furnace and remelting it under vacuum (by Vacuum Arc Remelting (VAR) or by Electroslag Remelting (ESR)) to achieve the requested high levels of purity. The chemical composition is listed in Table 1.

The procedure used for processing ingots into billets and hollows was up to the manufacturer as long as homogeneity with respect to chemical composition could be guaranteed and that non-metallic inclusions and TiC primary precipitateswere within specifications. The final step in billet production is a homogenization heat treatment typically at 1200 °C for at least 12 h to bring Ti and C back in solid solution. Hollows from which the tubes are drawn, are made by peel turning and deep boring.

The tubes are obtained through a series of plug-drawing steps, each preceded by a short solution-anneal around 1100 °C for a few minutes in a continuous furnace. The last plug-drawing determine the cold work level defined as the reduction in cross-section. In the ‘aged’ state, a final ageing treatment around 800 °C during 2 h was applied after the last cold-drawing.

Cladding tubes so obtained are fine-grained (grain size < 50 μm), contain $M_{23}C_6$ precipitates at grain boundaries (improving creep resistance by hindering grain boundary sliding [32]) and coarse ‘primary’ TiC precipitates (diameter > 50 nm). During aging, fine ‘secondary’ TiC precipitates (diameter ≤ 20 nm) nucleate at high temperature (>600 °C [33]) on defects introduced during the final cold working [17,34–36]. The fine TiC precipitates are particularly resistant to temperature-driven coarsening and, by pinning dislocation motion, are critical in improving creep properties [17,36–45]. also. It was also found that they similarly nucleated under in-reactor conditions at high temperature (≥~500 °C under irradiation [19]) on cold-worked steel. Swelling resistance of non-aged cold-worked steel was found to be better so the ageing treatment at 800 °C was abandoned in the later stages of the Fast Reactor (FR) development programs. As will be shown later, tubes in the cold worked or cold worked + aged states show similar creep properties, both being significantly better than the solution annealed state [46]. Almost all creep tests performed during the DeBeNe program were performed on tubes with the following two thermo-mechanical (TM) treatments:

- TM1: Solution anneal (~1060–1150 °C, 5–10 min) + cold work 15–25% + aging (800 °C, 2 h)
- TM2: Solution anneal (~1060–1150 °C, 5–10 min) + cold work 15–25%

In the following TM1 tubes will be referred to as *cold worked + aged* (*cw + aged*) and TM2 as *cold worked* (*cw*).

2.2. Dataset content

Close to 1150 tubes have been tested for biaxial creep over the course of the DeBeNe R&D program [30]. Creep tests were performed at different institutions of the DeBeNe consortium, some in parallel with irradiation experiments and others to qualify fuel pin cladding material. Besides the determination of the baseline creep properties and the uncertainty quantification for design purposes, creep tests were often applied to identify abnormal fabrication and served, as such, as an acceptance test including also the qualification of plug welding.

In 1974, based on the results available to that date, *cw + aged* DIN 1.4970 was selected for the first core (MkIa) of the SNR-300 sodium-cooled fast reactor for a target dose of 65 dpa [47]. However, ongoing irradiation experiments with cold worked material showed better resistance to swelling [30,48] by delaying its onset compared to the *aged* state. These results led to the selection of *cw* DIN 1.4970 as the reference state for the second core (MkII).