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by

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Mechanical Properties and Examination of Cracking in TMI-2 Pressure Vessel Lower Head Material^{*}

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Abstract

Mechanical tests have been conducted on material from 15 samples removed from the lower head of the Three Mile Island Unit 2 nuclear reactor pressure vessel. Measured properties include tensile properties and hardness profiles at room temperature, tensile and creep properties at temperatures of 600 to 1200°C, and Charpy V-notch impact properties at -20 to +300°C. These data, which were used in the subsequent analyses of the margin-to-failure of the lower head during the accident, are presented here. In addition, the results of metallographic and scanning electron microscope examinations of cladding cracking in three of the lower head samples are discussed.

Analyse des Propriétés Mécaniques et Examen Microscopique de Fissures dans le Matériau constituant la Partie Inférieure de la Chambre de Pression du Réacteur TMI-2*

Résumé

Des test mécaniques ont été réalisés sur quinze éprouvettes extraites de la partie inférieure de la chambre de pression du réacteur nucléaire Three Mile Island. Les propriétés mesurées comprennent: les propriétés en traction et le profil de dureté à température ambiante, les propriétés en traction et la réactivité entre 600 et 1200°C et enfin les résultats du test de Charpy sur éprouvettes, taillées en V, sollicitées entre –20 et 300°C. Toutes ces données, utilisées pour l'analyse de la rupture accidentelle de la partie inférieure de la chambre de pression, sont décrites dans le présent document. Ce dernier comprend également une discussion des examens microscopiques effectués sur trois éprouvettes. Les observations portent sur les fissures engendrées dans l'enveloppe interne de la chambre de pression.

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Introduction

Fifteen prism-shaped samples, each \approx 152-178 mm (6-7 in.) long, 64-89 mm (2.5 -3.5 in.) wide, and 64-76 mm (2-1/2-3 in.) deep, were recovered from the TMI-2 lower head during the first phase of the TMI-2 Vessel Investigation Project (VIP). The samples were cut from the inner surface of the lower head and typically extend through approximately half the lower head thickness. The locations from which the lower head samples were taken are shown in Fig. 1.

Mechanical tests were conducted on test specimens from these samples to determine the tensile, creep, and impact properties of the lower head material under conditions relevant to the accident scenario. The tests were conducted by Argonne National Laboratory (ANL) and participating OECD partner laboratories, and the results are summarized here. These results were used to assess the integrity of the lower head and its margin-to-failure during the accident.

Cladding cracks were observed in three of the lower head samples, namely E-6, G-8, and F-10, during initial examinations conducted at ANL prior to sectioning. Cladding cracks were also detected at location G-6 in the TMI lower head, but no sample was removed from this location. Metallographic and scanning electron microscopy (SEM) examinations were conducted on Samples E-6 and G-8 in some detail, to characterize the nature and extent of the cracking. The results of these examinations are also summarized here. More details on the results of both the mechanical tests and the examination of the lower head cracking may be found in Ref. 1.

Test Procedures

The tensile and stress-rupture tests were conducted on specimens of rectangular cross section designed in accordance with ASTM Standards E8 and E139 and applicable standards of the Deutsches Institut für Normung (DIN).¹ The specimen design used for the impact tests was the conventional Charpy V-notch test specimen (ASTM E23).

Tensile tests were conducted at room temperature for comparison with data in the literature. All other tensile and creep tests were conducted at a minimum temperature of 600°C. It was judged that little or no damage would have occurred to those portions of the lower head where the maximum temperature did not exceed this value and that failure was unlikely at these locations. The maximum temperature of 1200°C for these tests is slightly above the maximum 'Jwer head temperature believed to have been attained during the accident. Impact tests were conducted over a temperature range of -20 to +300°C to define the ductileto-brittle transition temperature for these specimens.



- Locations of Lower Head Samples
- Nozzle Positions

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Nozzles with Associated Cladding Cracks

Figure 1. Map of lower head of TMI-2 pressure vessel

The tensile tests, carried out at ANL and in Belgium, France, and Spain, were conducted in general accordance with ASTM Standards E8 and E8M, and all elevated-temperature tests were conducted in an Ar or He environment. The strain rate for the elastic portion of the loading was $\leq 5 \times 10^{-4} \text{ s}^{-1}$, and the strain rate during plactic loading was $4 \times 10^{-4} \text{ s}^{-1} \pm 1 \times 10^{-4} \text{ s}^{-1}$. The reported yield-strength values were obtained by the 0.2% offset method, except where discontinuous yielding occurred; in these cases, the observed upper yield strength is shown.

The creep tests were carried out at ANL and in Belgium, France, and Spain in general accordance with ASTM Standard E139. The tests were conducted in an Ar or He environment except most of those conducted by the SCK/CEN in Belgium. All but one of the Belgian tests was conducted in vacuum; a single test at 800°C and 30 MPa was conducted in an Ar environment.

The tests were conducted on specimens with various prior thermal histories resulting from the accident. Because the number of specimens from the highest-temperature portion of the lower head was limited, it was necessary in some cases to heat treat low-damage specimens before testing to produce the corresponding microstructure. This heat treatment consisted of heating the specimen to 1000°C, holding it at this temperature for 2 h, and then cooling it to room temperature at ≈10-50°C per min. For specimens to be tested at 1000°C or above, this prior heat treatment was omitted because its effects would be negated by the thermal treatment imposed during testing.

Results and Discussion

Tensile Tests. The results of the tensile tests conducted on the lower head base-metal specimens are presented in Fig. 2. Also plotted in Fig. 2 are average values reported by the Japanese National Research Institute for Metals (NRIM) for five other heats of A533, Grade B steel.² The NRIM data were obtained at a strain rate of 5 x 10⁻⁵ s⁻¹ up to yield and 1.25 x 10⁻³ s⁻¹ for the remainder of the test. The NRIM tensile strength data suggest a strain-aging effect between 100 and 300°C, resulting in a local tensile strength minimum at ≈150°C. Both the tensile and yield strengths of this alloy are strongly temperature-dependent; the room-temperature values are reduced by a factor of more than 2 at 600°C and by a factor of more than 10 at 900°C.

The data for specimens taken from lower head samples E-6 and E-8 are plotted separately in Fig. 2; these data lie significantly above the best-fit curve to the remaining data. It has been determined that both of these samples were heated to maximum temperatures of \approx 1000-1100°C during the accident, followed by a relatively rapidly cooling.³ The resulting hardening produced significant increases in strength at both room temperature and 600°C.

Creep Tests. The stress-vs.-time-to-failure data are plotted in Fig. 3 along with a Manson-Haferd best fit discussed below. Materials with slightly different thermal histories were tested at both 600 and 700°C. At 600°C, tests were conducted on specimens from Sampie K-13, for which the maximum temperature during the accident did not exceed 727°C, as well as on specimens from Sample F-5, for which the maximum temperature was apparently somewhat greater than 727°C over a portion of the sample. No significant difference in time to failure is observed in Fig. 3, probably because the maximum temperature did not



Figure 2. Tensile and yield strengths of TMI-2 lower head material

significantly exceed the transformation temperature of 727°C in F-5, particularly in the bottom half of the sample from which the creep test specimens were taken. Similarly at 700°C, specimens from Sample M-11, for which the maximum temperature may have approached or slightly exceeded 727°C, show no difference in behavior when compared to that of specimens from Sample H-8, for which the maximum temperature remained below 727°C.

Two time/temperature correlations were explored in an attempt to fit the basemetal creep data. The first of these was the Larson-Miller parameter L^4

$$L = T[C + \log_{10}(t_f)],$$

where T is temperature in Kelvin, t_f is time to failure in h, and C is a fitting constant. A least-squares analysis determined that the optimal value of C for the present data base was 12.5, and that stress s was related to the Larson-Miller parameter by the relationship

 $\log_{10}(s) = 4.3406 - 0.00018767 \cdot L_{1}$



Figure 3. Stress-rupture data with Manson-Haferd best fit

where the applied stress s is in MPa. However, the resulting fit to the data was only fair.

The Manson-Haferd time/temperature correlation⁵ was therefore evaluated in an attempt to obtain a better fit to the data. The Manson-Haferd parameter M has the form

$$M = \frac{\log_{10}(t_{\rm f}) - t_{\rm a}}{T - T_{\rm a}}$$

where t_f is time to failure in h, T is test temperature in Kelvin, and t_a and T_a are fitting constants. A least-squares analysis was again carried out, and the optimal values for t_a and T_a were found to be 7.57 and 520, respectively. Log(s) was found to vary with the Manson-Haferd parameter M according to the relationship



Figure 4. Absorbed impact energy vs. test temperature

 $\log_{10}(s) = -0.80467 - 261.41 \cdot M - 5291.25 \cdot M^2$.

A comparison of the resulting best-fit curves with the actual s vs. tf data in Fig. 3 shows a reasonably good fit. However, systematic departures from the actual data are noted in the 700-900°C region. This problem may be associated, in part, with the ferrite-to-austenite phase transformation that occurs over the temperature regime from 727 to \approx 850°C.

Impact Tests. The Charpy V-notch impact data obtained in Italy⁶ on specimens from the lower head are summarized in Fig. 4. The three groups of test specimens in which the maximum temperature did not exceed 727°C show similar behavior, with an upper-shelf energy of ≈170 J and a ductile-to-brittle transition temperature of ≈20°C. However, the data from specimens of Sample F-10, for which the maximum temperature was as high as ≈1050°C, stand in marked contrast. The F-10 material shows a significantly higher ductile-to-brittle transition temperature of ≈70°C, as well as a reduced upper-shelf energy of ≈120 J. These differences reflect the reduced ductility and impact resistance produced in this material by the high temperatures and relatively rapid cooling associated with the accident.



Figure 5. Cross section through principal crack in Sample E-6

Examination of Cracks in Vessel Cladding

Results of the metallographic examinations through the cracks in samples E-6 and G–8 indicated that the cracking originated in and was essentially limited to the Type 308L stainless steel cladding used on the vessel. A cross section through the principal crack in E–6 is shown in Fig. 5. Penetration into the A533 vessel steel was only superficial (~3–mm). This was also the case for the two cracks examined from the G–8 sample, where the maximum penetration was somewhat greater (~6–mm).

The rent nature of the 308L stainless steel within the crack of E–6, which was also typical of the G–8 crack, is evidence of the elevated–temperature ductility of the Type 308L weldment and the hot tearing along interdendritic boundaries that resulted in the cracks. The hot tearing would have been caused by the thermal stresses when this hot–spot area cooled rapidly at \approx 10- 100°C per min.



Figure 6. Ag-Cd inclusions (arrow) in intergranular tears in Sample G-8

The SEM examination of the materials found in the E–6 and G–8 cracks revealed evidence of molten material that was present at, or shortly after, formation of the cracks. The principal constituents of this material, which appeared to be layered on the exposed crack surfaces, were Fe, Cr, and Ni, together with Sn, In, Ag, and Cd in combinations as second phases or discrete metallic particles within the general oxidized matrix. The appearance of the material indicated that it was not a surface oxidation product, but had once been molten and was interacting with the cladding in a solid/liquid reaction. These elements are the essential constituents of the Zircaloy–shrouded, stainless–steel–clad Ag–In–Cd control rods. The extensive gray structure in the root of the crack, however, was principally the Fe oxidation product of A533 vessel steel laced with a solidified Sn–In phase. Fuel particles were found only on top of the oxidized layers or as minor constituents of the layers. The absence ot significant quantities of fuel in the cracks indicates that the massive fuel tlow to the lower head was not the source of the solidified material in the cracks.

Inclusions of Ag-Cd were found in numerous intergranular tears on the surface and well into the cladding (~4-mm), as shown in Fig. 6. It is quite likely that interdendritic penetration of these materials as liquids contributed to the hot

tearing of the cladding. Copper was also found in the cladding, next to the cracks, suggesting the causative agent for the hot tearing.

These observations on the superposition of core materials in the cladding and in cladding cracks suggest that the control-assembly materials were already on the lower head when the massive fuel flow from the core region arrived. Because the control-assembly materials would have reached the lower head as solids, they apparently were remelted by the fuel flow, resulting in intergranular penetration of the cladding by Ag–Cd, which probably contributed to the hot tearing during the subsequent rapid cooling.

Summary and Conclusions

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Mechanical tests and microstructural characterizations have been conducted by Argonne National Laboratory and the OECD partner laboratories on material from 15 locations in the lower head of the pressure vessel of the TMI-2 nuclear reactor. Mechanical tests consisted of tensile tests at room temperature, tensile and creep tests at 600-1200°C, and Charpy impact tests at -20 to +300°C. The specimens were taken from locations where the maximum temperature had not exceeded 727°C during the accident and from locations where the maximum temperature had been as high as 1100°C. Microstructural characterizations were conducted by conventional optical metallography and scanning electron microscopy (SEM). The results of these investigations lead to the following conclusions:

- Results of tensile tests conducted on base-metal specimens for which the maximum temperature during the accident (T_{max}) did not exceed 727°C agree well with literature data for A533B steel and show a dramatic drop in strength at temperatures above 600°C.
- Tensile specimens from samples for which T_{max} exceeded 727°C showed significantly higher strengths at room temperature and 600°C than did specimens for which the temperature did not exceed 727°C.
- Creep tests at 600 and 700°C indicated no significant difference in behavior between base-metal specimens for which T_{max} was ≈727°C and those for which T_{max} was well below this value.
- 4. The stress-rupture data obtained from base-metal specimens could be fitted more accurately with a Manson-Haferd time/temperature parameter than with a Larson-Miller parameter.
- Charpy V-notch impact tests conducted on lower head base-metal material revealed a substantial difference between specimens from Sample F-10 (for which T_{max} was as high as ≈1050°C) and specimens from samples for which

T_{max} was <727°C. The F-10 material showed a significantly higher ductile-tobrittle transition temperature, as well as a reduced upper-shelf energy value.

- Cracks through the stainless steel cladding of Samples E-6 and G-8 appear to have been hot-tearing phenomena, probably assisted by interdendritic penetration of liquid Ag-Cd.
- 7. Materials in the cladding cracks suggest the presence of control-assembly debris on the lower head before arrival of the massive flow.

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