
Peer Review of the Three Mile Island Unit 2 Vessel Investigation Project Metallurgical Examinations

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Abstract

Fifteen samples recovered from the lower head of the Three Mile Island (TMI) Unit 2 nuclear reactor pressure vessel were subjected to detailed metallurgical examinations by the Idaho National Engineering Laboratory (INEL), with supporting work carried out by Argonne National Laboratory (ANL) and several of the European participants. These examinations determined that a portion of the lower head, a so-called elliptical "hot spot" measuring $\approx 0.8 \times 1$ m, reached temperatures as high as 1100°C during the accident and cooled from these temperatures at $\approx 10\text{-}100^{\circ}\text{C}/\text{min}$. The remainder of the lower head was found to have remained below the ferrite-to-austenite transformation temperature of 727°C during the accident.

Because of the significance of these results and their importance to the overall analysis of the TMI accident, a panel of three outside peer reviewers, Dr. Robert W. Bohl, Mr. Richard G. Gaydos, and Mr. George F. Vander Voort, was formed to conduct an independent review of the metallurgical analyses. After a thorough review of the previous analyses and examination of photo-micrographs and actual lower head specimens, the panel determined that the conclusions resulting from the INEL study were fundamentally correct. In particular, the panel reaffirmed that four lower head samples attained temperatures as high as 1100°C , and perhaps as high as $1150\text{-}1200^{\circ}\text{C}$ in one case, during the accident. They concluded that these samples subsequently cooled at a rate of $\approx 50\text{-}125^{\circ}\text{C}/\text{min}$ in the temperature range of $600\text{-}400^{\circ}\text{C}$, in good agreement with the original analysis. The reviewers also agreed that the remainder of the lower head samples had not exceeded the ferrite-to-austenite transformation temperature during the accident and suggested several refinements and alternative procedures that could have been employed in the original analysis.

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Executive Summary

The peer review panel (R. W. Bohl, R. Gaydos and G. F. Vander Voort) was requested, under Contract B95554, to review the metallurgical reports dealing with the estimation of the maximum temperature of the lower pressure vessel head of the Three Mile Island (TMI) Unit 2 nuclear reactor as a result of the incident of 28 March 1979. To accomplish this goal, the panel members were supplied with a number of reports prepared by members of the staffs of the Idaho National Engineering Laboratory (INEL) and the Argonne National Laboratory (ANL), plus reports by various cooperative nuclear laboratories in other countries. Panel members also visited ANL on 2-3 June 1993 to discuss the reports, view original photo-micrographs, and examine the microstructures of specimens taken from the TMI-2 lower head as well as that of control specimens from an unused reactor head subjected to various thermal exposures. The panel also met with the Nuclear Regulatory Commission (NRC) in Rockville, MD on 26 August 1993 to present its findings.

Each panel member prepared a report of his findings. These reports are presented here. Each member's review of the work was based on his own personal experiences and background. While the nature of their findings and conclusions vary somewhat on minor details, the panel members are in agreement as to the overall conclusions. These conclusions are listed below:

- There was no evidence of melting or incipient melting in any lower head boat specimen.
- Only four specimens (E-6, E-8, F-10 and G-8), all from the same general region of the lower head, reached temperatures in excess of the critical temperatures for ASTM A533 Gr. B which were in the range of 715-720 and 830-850°C, respectively, for the Ac_1 and Ac_3 , the lower and upper critical temperatures. Heating above these temperatures produces a fully austenitic (γ) microstructure that will transform upon cooling back to ambient temperature.
- For Specimens E-6, E-8, F-10 and G-8, temperatures remained $>Ac_3$, i.e., $>\sim 850^\circ\text{C}$, to the full depth of the boat specimens, ~ 40 -67 mm, depending upon the specimen.
- The conclusions of G. E. Korth about the maximum temperature attained in each of these four specimens (i.e., G-8 and F-10 reached ~ 1040 - 1060°C for 30 min and E-6 and E-8 reached ~ 1075 - 1100°C for 30 min) appear to be reasonably accurate, probably within $\pm 50^\circ\text{C}$.
- It is possible that Specimen E-8 may have attained a temperature as high as 1150 - 1200°C , but certainly no higher.

- Definition of the maximum temperatures experienced by specimens E-6 and G-8 is less precise than that for E-8 and F-10 because of the surface metal removed in order to decontaminate E-6 and G-8.
- For the four specimens that were heated enough to form austenite (E-6, E-8, F-10, and G-8), the cooling rate through the γ transformation region was fast enough to form a relatively hard bainitic microstructure. This structure forms at cooling rates between ≈ 50 and $125^\circ\text{C}/\text{min}$ in the temperature range of 600 - 400°C .
- Gross tears were observed in the cladding from the E-6 and G-8 locations, i.e., within the region that experienced the highest temperatures. In this region, the temperature was high enough to partially dissolve and spheroidize the delta (δ) ferrite in the T308 stainless steel cladding. The cracks appear to be hot tears, which form as a consequence of the inability of the nearly fully austenitic structure to accommodate shrinkage stresses upon cooling to ambient temperature.
- The remaining eleven specimens attained temperatures, in the A533 Gr. B head, of $<A_{c1}$, i.e., $<\approx 715^\circ\text{C}$.

Robert W. Bohl

Richard Gaydos

George F. Vander Voort

1 Background

The Three Mile Island (TMI) Vessel Investigation Project is an international program sponsored jointly by the U.S. Nuclear Regulatory Commission (NRC) and the Organisation for Economic Co-operation and Development/Nuclear Energy Agency (OECD/NEA). Participants in this program include the U.S., Japan, Belgium, Germany, Finland, France, Italy, Spain, Sweden, Switzerland, and the United Kingdom.¹

During the first phase of this project, 15 samples were removed from the lower head of the TMI Unit 2 nuclear reactor. These prism-shaped samples, each extending approximately half-way through the lower head thickness, were subjected to initial examinations at Argonne National Laboratory (ANL) and then decontaminated and sectioned into metallographic and mechanical test specimens. Detailed examinations of the metallographic specimens were performed at the Idaho National Engineering Laboratory (INEL), with supporting work carried out by ANL and several of the European participants.²⁻⁴

These metallographic examinations revealed that a portion of the lower head, a so-called elliptical "hot spot" that measured $\approx 0.8 \times 1$ m, reached temperatures as high as 1100°C during the accident and cooled from these temperatures at ≈ 10 - $100^{\circ}\text{C}/\text{min}$. These rather unexpected results are of great importance to the overall analysis of the TMI accident and, in particular, to the calculation of the margin-to-failure for the lower head. Accordingly, the NRC directed that ANL secure the services of three outside peer reviewers to conduct an independent review and critique of these examinations and the conclusions reached. This review has been completed and the findings of the reviewers, Dr. Robert W. Bohl, Mr. Richard G. Gaydos, and Mr. George F. Vander Voort, are summarized here.

2. Detailed Discussion by R. W. Bohl

2.1 Introduction

The Metallurgy Peer Review Panel was charged with evaluating the results reported by various laboratories which examined specimens taken from the lower head of the TMI-2 reactor vessel. These investigations were designed to determine temperatures reached by the vessel during the core meltdown in order to assess the severity of the threat to the vessel integrity caused by the deposition of core debris on the lower head.

2.2 Procedure

The panel was provided with reports from the various cooperating laboratories, and met at ANL on June 2-3, 1993 to discuss the reports, view original data, and examine metallographic samples taken from the vessel, as well as those from the Midland reactor (archival material), which had been subjected to a range of thermal cycles.

Conclusions regarding the thermal history of the vessel as a result of the accident were based on modifications to the properties (particularly hardness) and the microstructure of the as-fabricated vessel by the thermal excursion experienced.

The vessel was fabricated from 136 mm-thick A533B ferritic steel plate, quenched and tempered at $\sim 660^{\circ}\text{C}$, upon which 5 mm of 308L stainless steel was weld-cladded to the inside surface. After welding, the vessel was stress-relief annealed 50 h at $\sim 600^{\circ}\text{C}$. There is some discrepancy in the carbon content reported for the samples. A533B specifications call for 0.25% carbon maximum. Analyses reported range from 0.22% (ladle analysis) to 0.28%. This range is beyond the tolerance required for chemical analysis and may be rationalized on the basis of the method of analysis (not reported) or the sampling technique used to accommodate the chemically banded plate. Hardness values are sensitive to carbon content, and might affect the hardness levels measured, but not affect the change in hardness observed in the gradient. The variation in carbon composition would not materially affect the temperatures at which microstructural changes took place.

The fabrication of the vessel produced a microstructural gradient with the following features:

1. Columnar weld metal containing about 5% δ -ferrite in an interdendritic network.
2. A "dark band," at the WM-BM interface, which was very hard and formed when the highly alloyed stainless steel was mixed with the carbon-bearing steel plate. The nature of this band was not determined, nor was its identification necessary to establish the temperature gradients attained.

3. A heat-affected base metal zone where the heat of welding raised the base metal into the austenite range, producing a large austenite grain size to a depth of about 10 mm below the junction.
4. The base metal heated to temperatures below the lower critical transformation temperature, and consisting of the original tempered bainite structure, i.e., small precipitated carbides.

As molten core material deposited on the lower head, the temperature increased and modified the as-fabricated microstructure. The following changes (in order of occurrence with decreasing temperature) could occur and identify the temperatures reached during the meltdown:

1. Remelting of the weld clad: 1400°C. Since no melting of the clad was observed, this temperature was not reached.
2. Agglomeration and partial solution of δ -ferrite in the weld metal: 1100-1150°C.
3. Grain growth of austenite in the base metal: 1050°C.
4. Solution of the dark band: 850-900°C.
5. Transformation of the base metal to fine-grained austenite: 850°C
6. Decomposition of δ -ferrite to sigma in the weld metal: 700°C. Since the weld metal was clearly heated above this temperature range in the critical hot zone, this change is of little significance.
7. Initial formation of austenite in the base metal: 727°C.
8. Coalescence of the tempered bainitic carbides in the base metal heated below the lower critical temperature. Since the as-fabricated vessel was stress relieved 50 h at 600°C, any additional coarsening caused by brief heating between this temperature and 727°C would likely be imperceptible.

The microhardness surveys provided valuable insight into the effect of the thermal excursion on the steel vessel. As the ferritic steel was heated over the critical temperature, the effects of the fabrication history, particularly the stress-relief anneal, were eliminated and the structure and hardness were controlled by the attained austenite grain size and the cooling rate. Abrupt changes in both structure and hardness were noted in the samples taken from the "hot zone," i.e., E-6, E-8, F-10, and G-8. Since no other region of the lower head reached these temperatures, attention was concentrated on samples taken from this sensitive region.

The availability and use of the Midland archive vessel to simulate various time-temperature-cooling rate histories was a very valuable aspect of the metallurgy

analysis. The validity of conclusions based on observations of the TMI-2 vessel samples was strongly confirmed by the ability to simulate these features with the Midland vessel steel.

2.3 Comments

If one were to design this study with the benefit of hindsight, there are some features which may have been modified: metallographic studies might have included the use of selective etchants to better reveal the austenite grain size in the heat-affected regions; more use of quantitative metallography might have shed more light on regions of grain-size gradients; exclusive use of microhardness testing could better define hardness gradients; and taking some samples from the outer surface of the lower head (perhaps this was not feasible) could better define temperatures throughout the vessel wall. Temperature gradients through the 50-mm-deep samples suggest that, even in the hottest zone, the outside wall did not exceed 700°C; these gradients appear to be $\sim 3^{\circ}\text{C}/\text{mm}$. Members of the panel agree, however, that such refinements would not affect the final conclusions of the Metallurgy Program to any significant degree.

2.4 Conclusions

There are a great number of uncertainties necessary to accurately model the threat to the integrity of the vessel during this accident. Fortunately microstructural features are based on material properties that are not subject to assumptions; e.g., the steel either exceeded the critical temperature or it did not, as revealed by the microstructure. In my opinion, the Metallurgy Program was well-conceived and carried out, and I concur with the conclusions reached. The close agreement among the laboratories that independently carried out parallel studies constitutes strong support for the interpretations reported. While details of heat transfer, molten core deposition rates, cooling scenarios, etc., may be obscure, the results of these studies are sound and valid.

3. Detailed Discussion by R. G. Gaydos

The objective of the peer consultants was to review the method of examination of the postaccident A533B pressure vessel and the conclusions formed. The objectives of the INEL and ANL Laboratories were to determine, by metallurgical methods, the peak thermal temperatures reached to assess, to the extent possible, the margin to failure.

Numerous reports, which encompassed findings to develop scenarios of the accident, were studied. Nozzle examinations, mechanical tests, and microstructural examinations of the lower head samples were conducted.

The metallurgical sampling, sample preparation, photomicrographs, and lastly, but most important, microscopic examinations of numerous samples taken from the lower head and Midland archive material were compared. Heat-treating cycles were performed with the aid of continuous-cooling curves to evaluate temperatures reached.

The studies were helpful in developing a scenario of events in the lower head during the accident. The delineation of a "hot spot" evolved through studies of microstructural changes from the temperature attained (approaching 1100°C) on or near the clad surface. Similarly, the periphery of the "hot spot" was ascertained by microstructural changes that resulted from exceeding or approaching the lower critical temperature in the A533 Gr. B steel (727°C).

Exact evaluation of phases present was qualitative at best. The nozzles and base metal indicated significant insulating effects by some debris and core material. The extreme temperatures appeared to have occurred some distance above the clad base. The tumult resulting from core material, fuel elements, and water provided a highly variable system. Detailed quantification may have improved the outline of the "hot spot;" however, since microstructural changes involve *time* and *temperature*, exactness in such a highly variable system is nebulous.

This reviewer is in basic agreement with the findings and conclusions of G. E. Korth of INEL and D. R. Diercks and L. A. Neimark of ANL. Lastly, in the category of lessons learned, some differences shall be discussed.

3.1 Chemical Analysis

As will be discussed under microstructure, carbon analysis is important to better understand the diffusion occurring during cladding.

Ladle analysis is the term applied to the chemical analysis representative of the heat. It is common practice to obtain three or more molten samples during teeming operations from the first, middle, and last portions of the heat. These samples are used to represent the chemical analysis of the entire heat.

3.2 Check Analysis Sampling

Each heat or lot must be considered separately. Because of segregation, the location at which samples are taken is important. Simply put, the following, although abbreviated, may be considered standard practice:

1. For thicknesses >2 in. (5.08 cm), samples are taken by drilling the edge of the plate at a point midway between the rolled surface and the mid-thickness.
2. Sample drillings are obtained with a 1/2-in. (1.27-cm) drill.
3. Samples must be uniform and well mixed. Those to pass an ASTM No. 10 (2000- μ m) sieve or too fine to remain on an ASTM No. 30 (590- μ m) sieve are not recommended.
4. Samples can be compacted for spectrographic analysis.
5. Carbon analysis is always determined by wet chemistry. Spectrographic checks for carbon are *not* reliable.
6. For carbon, an allowance of 0.02% is allowable for plate containing up to 0.30 wt. % C. A carbon check greater than 0.24% would be considered rejectable for plate reported as 0.22% C.

Since carbon, silicon, and especially phosphorus are subject to segregation, their influence on diffusion during cladding is important.

Three samples of TMI-2 materials were checked at 0.25% C. Sample 1 was reported at 0.28% C. The vendor reported a heat analysis of 0.22% C and 0.17% Si. Four samples of TMI-2 material were checked for Si at 0.11, 0.16, 0.20, and 0.18 respectively. For up to a 0.30% C heat, a 0.02% Si variation is allowed.

It appears that the method of sample preparation or method of testing for chemical analysis is reflecting the banded (segregated) microstructure in the plate. The P content was 0.008% in all four samples vs. the vendor's heat analysis of 0.014%.

The vendor's and check analysis for P and S meet "firebox quality." This quality is intended for application in pressure vessels when exposed to fire or radiant heat where thermal and mechanical stresses may be encountered.

3.3 Nozzle Evaluation

An area of concern to this reviewer was related to the microstructural aspects where the nozzles were welded to the base. After some discussion, it was related that an error in nozzle sizing during construction resulted in removal of the nozzles and

repair welding. Since regions of investigation were associated with the "hot spot" where nozzles were severely attacked, there was the danger of confusing previous repair welding effects and changes related to the accident. In at least one instance, the writer and D. Diercks agreed that one examined nozzle fell into this category.

Overall, the writer agreed with D. Diercks' concluding scenario regarding the nozzles.

3.4 Repair of Cladding by Welding

The writer emphasized that both AIME and ASTM have produced documents on quality control. In particular, ASTM A647 is a standard specification of special requirements for steel plates for nuclear and other special applications. The specification relates to such items as repair welds, repair of cladding by welding, and nondestructive testing (NDT) of repair welds, etc. The manufacturer would be responsible for any documentation regarding cladding, repair welding, NDT, etc.

It was noted that at least one OECD member^a observed holes at the clad/base metal interface. Flaws of this magnitude should be detectable by NDT.

3.5 Microstructure and Hardness Evaluation

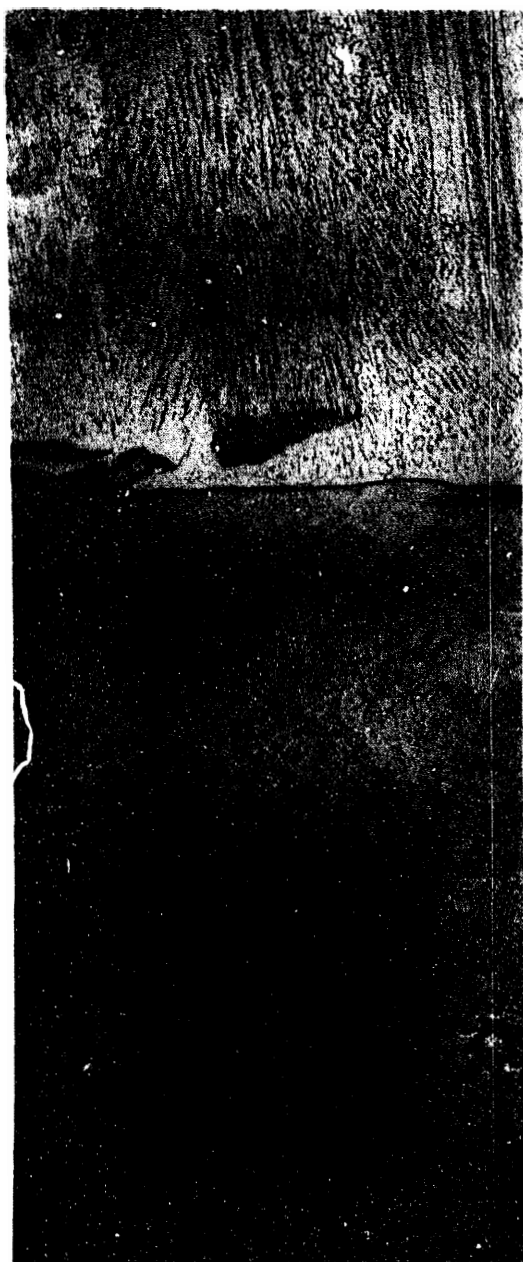
A number of photomicrographs display a "dark feathery" structure in the stainless clad/base metal fusion zone. The archive material and areas of the TMI-2 samples that were not heat affected display the dark structure of varying thickness. The variations in this dark etching carbide precipitate encountered by OECD members, ANL, and INEL investigators are typical of hand applied clad.

All investigators noted that the heat affected samples E-6, G-8, E-8, and F-10 exhibited dissolution of the dark feathery structure. The structure persists in regions not affected. Microstructures comparing such structures (F-10 and K-13) are appended as Fig. 1. After repolishing and re-etching by the review team, the structure was found to have characteristics of bainite.

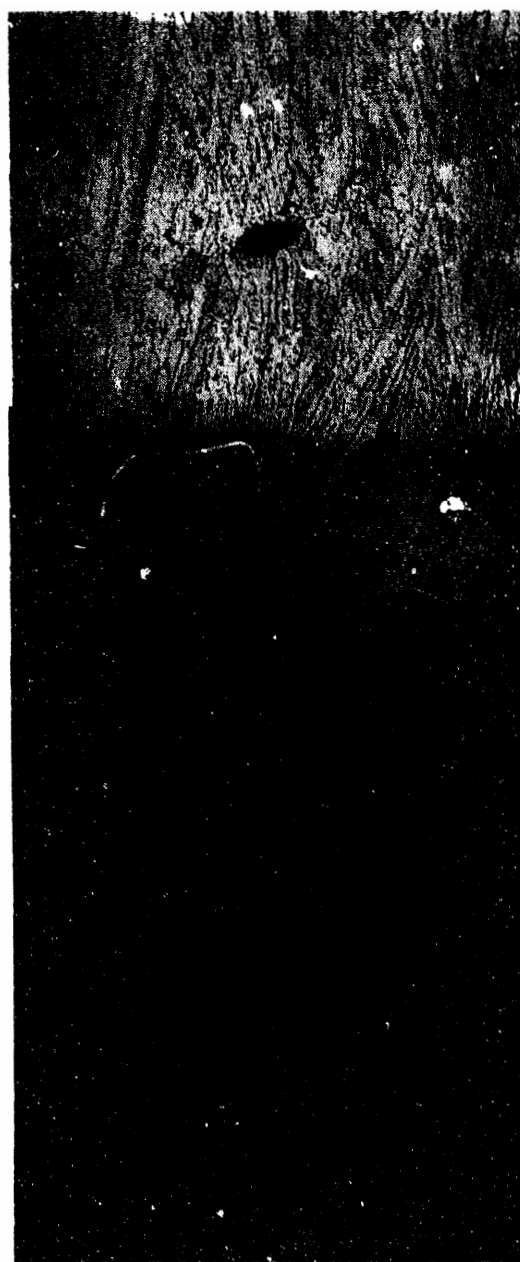
Since the dark structure is the result of carbon diffusion during cladding, the exact identification is not critical in evaluating the extent and magnitude of the heat affected zone (HAZ). Similarly, several laboratories measured the amount of delta ferrite to vary from 4 to 5%. Again, quantification is not critical since it too would be expected to vary.

More importantly, the spheroidization of the δ -ferrite phase could be utilized to establish time and temperature relationships in the HAZ.

^aW. Vandermeulen and W. Hendrix, *Examination Report of the Samples of the TMI-2 RPV Received by SCK/CEN (Belgium)*, SCK/CEN, Mol (March 1992).



K-13, no damage during accident

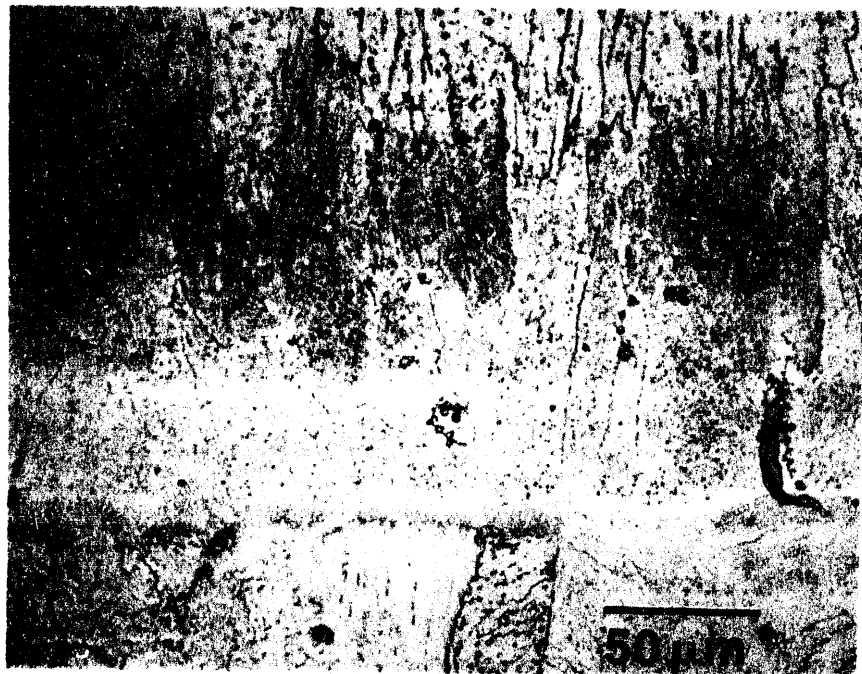


F-10, known thermal damage in accident

Fig. 1. Microstructures at weld clad interface of Samples K-13 (left) and F-10 (right).



(a)



(b)

Fig. 2. Stainless steel/low-alloy steel interface of (a) Sample F-10 (m3) and (b) Sample E-8 (m3), illustrating carbon diffusion band into the stainless steel and the associated cracking.

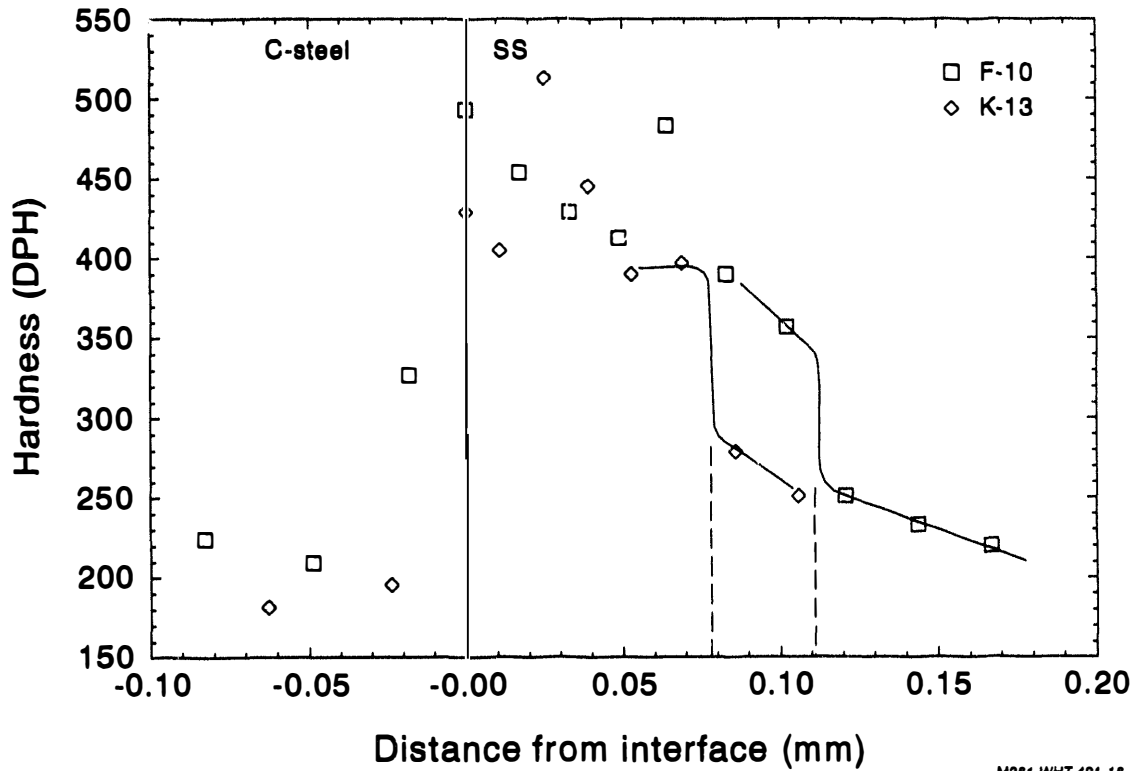


Fig. 3. Microhardness of Samples F-10 and K-13 as a function of distance from stainless steel/low-alloy steel interface (from Ref. 2).

The "hot spot" was also noted by most investigators. Generally the grain-coarsening temperature for fine grain, aluminum-killed low carbon, alloy steel will range from $\approx 1800^{\circ}\text{F}$ (982°C) to 2000°F (1093°C). A value of 1100°C cited by ANL and INEL is in reasonable agreement. Grain growth, being time- and temperature-dependent, would be expected to vary in a variable system.

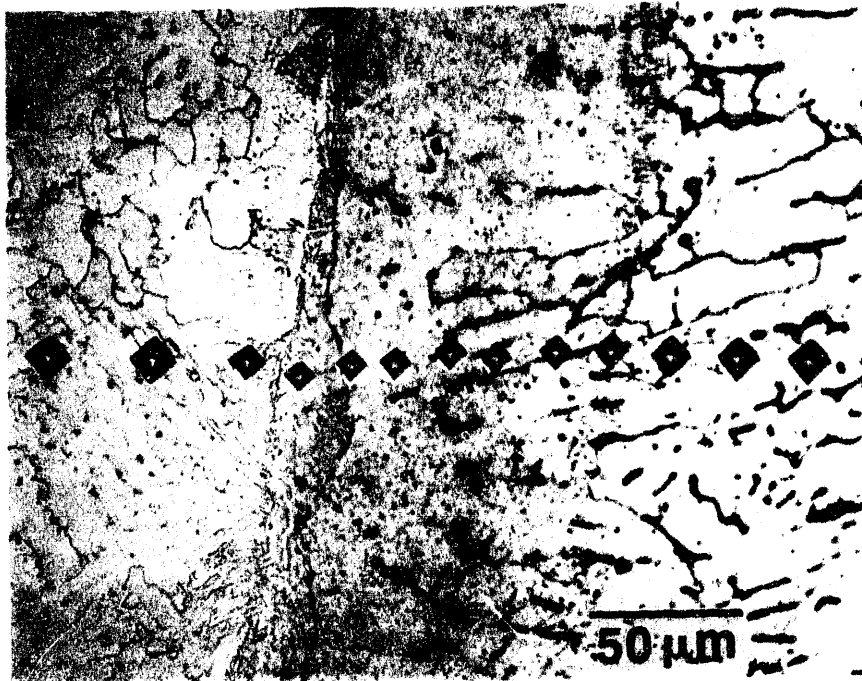
Also, due to the heterogeneity (segregation) common to low-carbon steel plate, the variability of chemistry would be expected to result in additional variations among the investigators.

Cracks were observed which were restricted essentially to the diffusion zone at the clad/base metal interface, as shown in Fig. 2 (taken from Ref. 2). Also appended are a hardness survey of Sample F-10 from the "hot spot" and of unaffected Sample K-13 (Fig. 3), and additional photomicrographs of Samples F-10 and K-13 (Fig. 4).

The hardness surveys clearly display an extension of the hardened zone from heat effects and carbon diffusion resulting from the accident.



(a)



(b)

Fig. 4. Microstructures at stainless steel/low-alloy steel interface in (a) Sample K-13 and (b) Sample F-10.

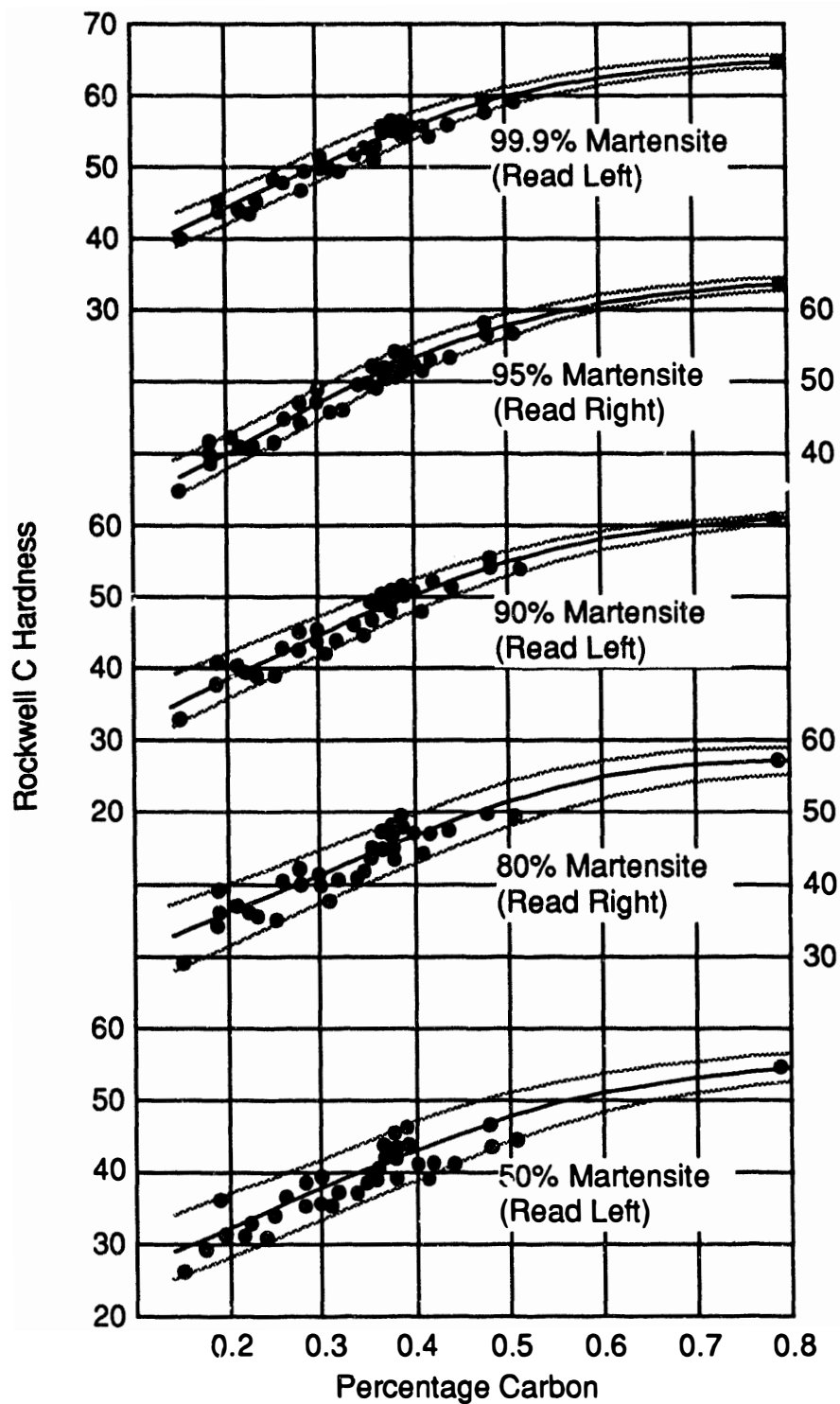


Fig. 5. Hardness of martensitic structures as a function of carbon content (adapted from Ref. 5, used by permission of LTV Steel Corp.).

Microcracks restricted to the hardened zone indicate cooling at a rate sufficient to cause hardening (500 DPH; $\approx R_c$ 50). Microcracks restricted to the hardened region indicate contraction resulting from tempering of the martensitic zone.

A relationship exists between hardness and the carbon content, as shown in Fig. 5.5. Also a relationship exists between the rate temperability of martensite and carbon content; lower carbon martensites are less stable at elevated temperatures.

The carbon content of any given carbon steel is generally lower near the hot-rolled surface, requiring time for diffusion (refer to chemical check analysis). Lastly, increasing the carbon and coarsening the austenitic grain size would shift the M_s of the cooling curves down in temperature and increase the time. Hence, a lower cooling rate would enable the formation of martensite followed by tempering sufficient to cause microcracking.

Considering the mass involved, it is difficult for this reviewer to rationalize cooling rates of $\geq 50^\circ\text{C}/\text{min}$. Certainly turbulence would enhance cooling, and in the presence of water at temperatures in excess of 500°C (932°F), turbulence can be envisioned. The M_s of the carbon-enriched clad high in nickel could be depressed to allow the mass of the plate to be effective in providing a cooling rate, similar to welding, to form martensite. Residual heat flow would temper the martensite to cause localized microcracking.

In summary, the findings of G. E. Korth, D. R. Diercks, and L. A. Neimark appear to be reasonable. Both INEL and ANL personnel are to be commended for designing an approach to investigating the failure in achieving their objective of evaluating the margin to failure.

A constructive critique would be that consultants could have contributed additional expertise to refine the findings at an earlier date. It is realized that budgetary constraints are in effect; however, national laboratories should be allowed to acquire state-of-art capabilities. However, this reviewer does not feel that additional investigation or refinement is warranted. In view of the degree of core loss, the vessel containment appeared to have withstood the accident to a greater degree than may have been anticipated.

4. Detailed Discussion by G. F. Vander Voort

The panel (R. W. Bohl, R. Gaydos, and G. F. Vander Voort) was requested to review the metallographic studies conducted to determine the maximum temperature reached in the lower pressure vessel head of TMI-2 as a result of the incident of 28 March 1979. The review was focused on the report prepared by G.E. Korth² but also included other relevant studies conducted at ANL and by nuclear laboratories in a number of other countries.

4.1 Korth's Approach

The conclusions as to the peak lower-head temperature were based upon qualitative comparisons of the microstructure of specimens from the lower head to that of specimens from a reactor never placed in service that were subjected to various reheating temperatures and times. Estimates of the temperature and time that each TMI-2 specimen experienced were based on the following microstructural observations:

- Start of $\alpha \rightarrow \gamma$ transformation at Ac_1 temperature (727°C quoted).
- Dissolution of dark feathery band at the cladding/base metal interface between 800 and 925°C, depending on time at temperature.
- Formation of small equiaxed grains in the A533 Gr. B steel HAZ adjacent to the interface between 850 and 900°C. These small grains disappear, as a result of grain growth, between 1025 and 1100°C.
- δ -ferrite islands in the cladding spheroidize in the range of 975-1000°C (100 min) or 1100-1125°C (10 min).

4.2 Reaustenitization of A533 Gr. B Head Material

Figure 5 in Diercks⁶ is a CCT diagram of an equivalent grade of German steel. According to this diagram, the lower critical temperature (Ac_1) is 715°C (at a heating rate of 0.4°C/min) and the upper critical temperature is 850°C. A CCT diagram for ASTM A533 Gr. B is also presented in a paper by Kunitake et al.⁷, reproduced here as Fig. 6, which shows that the lower and upper critical temperatures are 720 and 830°C, respectively. Although neither CCT diagram in Diercks (Figs. 4 and 5 in Ref. 6) gives hardnesses for the various cooling rates, they are given in Kunitake's CCT diagram, shown as Fig. 6. Although the Korth report does not state the composition of the lower head, the composition is given in Ref. 6; however the latter document lists erroneous carbon check analyses (see Table 1). The Lukens data reported in Ref. 6 indicated a carbon content of 0.22 wt.%. The maximum hardness of martensite in a quenched 0.22% C steel would be 45 HRC (446 HV, converted).

Hardness testing (Rockwell B converted to Vickers) was used for the initial screening of specimens and was very effective in showing that four were quite different

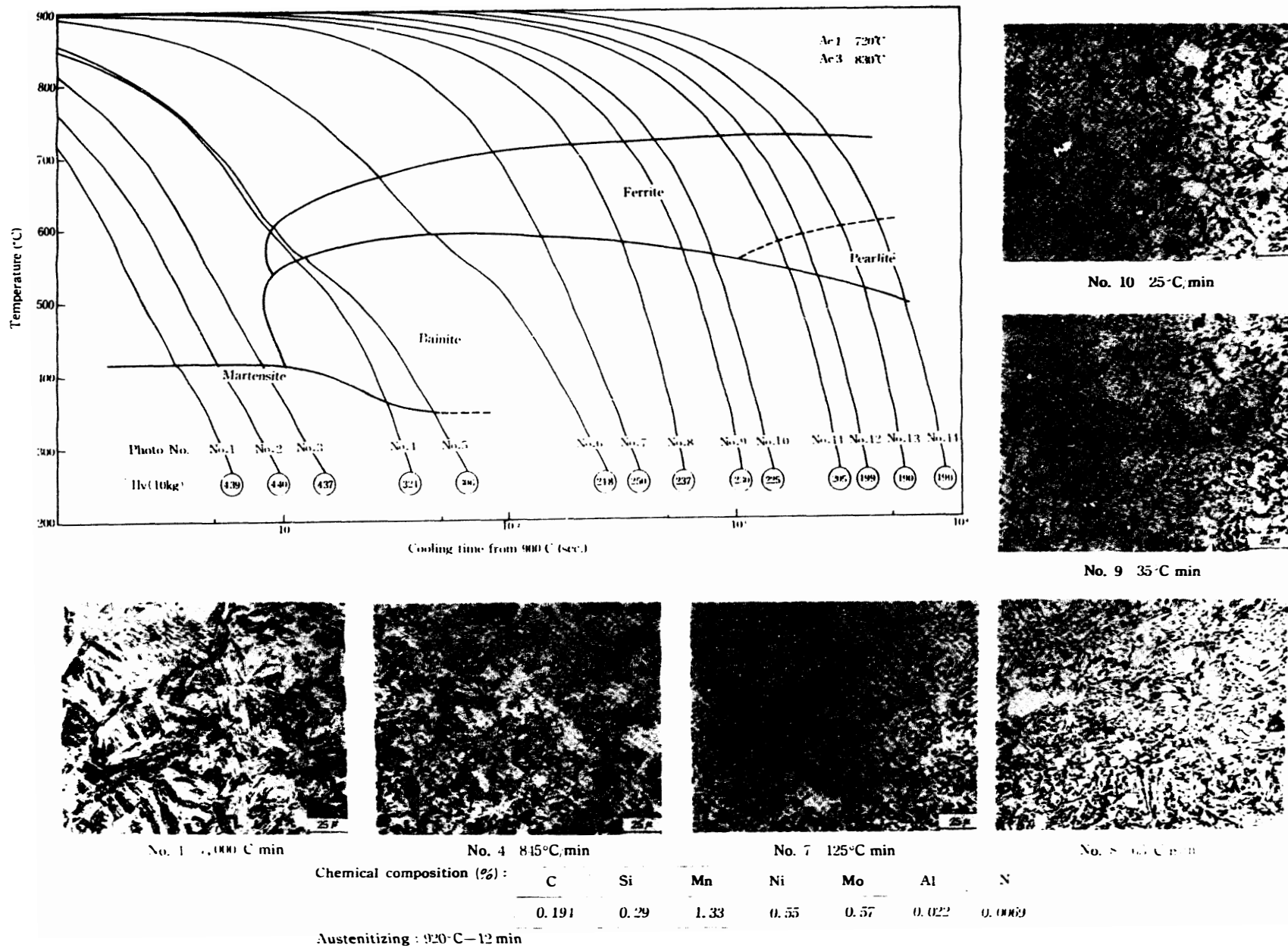


Fig. 6. Continuous-cooling-transformation (CCT) diagram for A533 Grade B steel (from ref. 7, used by permission of Nippon Steel Corp.).

Table 1. Carbon and δ -ferrite data

Source	Specimen	Carbon Content (wt.%)	δ -Ferrite Content (vol.%)
Lukens ^a	-	0.22	-
Diercks ^a	Analysis 1	0.28	-
Diercks ^a	Analysis 2	0.25	-
Diercks ^a	Analysis 3	0.25	-
Diercks ^a	Analysis 4	0.25	-
Spain ^b	M-11 (m6)	-	4.5
Spain ^b	K-7 (m5)	-	4.5
Spain ^b	E-8 (m6)	-	1.4
Germany ^c	F-5 (m6)	0.22	4.0
Germany ^c	E-8 (m9)	0.21	1.4
Germany ^c	K-13 (m5)	0.22	5.0
Germany ^c	L-9 (m9)	0.22	4.5
Germany ^c	M-8 (m4)	0.22	4.0
Germany ^c	M-11 (m5)	0.22	5.0
Germany ^c	H-4 (m5)	0.22	5.0
Germany ^c	H-5 (m5)	0.22	5.0
Germany ^c	K-7 (m6)	0.22	4.5

^aFrom Ref. 6, Table 16.^bFrom Ref. 13.^cFrom Ref. 14.

from the rest (which were similar in hardness to the non-exposed archive material). Hardness testing of the archive material revealed a low hardness in the cladding which increased at the interface and reached a peak value 2-4 mm below the interface. The hardness then decreased, reaching a rather constant lower hardness at ~ 10 mm below the interface down to ~ 40 mm. Four specimens, G-8, F-10, E-6, and E-8, exhibited different results. For E-8 and F10, the peak hardness was much higher and was obtained at ~ 6 mm below the interface (this area could not be tested on the E-6 and G-8 specimens). Testing at greater depths revealed little or no loss in hardness, unlike the archive specimens and the other TMI-2 specimens. A533 Gr. B head material in these four specimens was reaustenitized during the incident, that is, the temperature exceeded the upper critical temperature. The cooling rate was fast enough to form bainite, rather than ferrite and pearlite, but not fast enough to form martensite (which would have been harder, ~ 440 HV). The microstructural appearance was consistent with bainite also. This work showed that the steel in these four specimens was reaustenitized to a depth of at least 40 mm below the cladding, i.e., the temperature exceeded 830-850°C to a depth of more than 40 mm.

The hardness tests were very useful and the results thereof were valid. However, it would have been better to do all of the tests with a Vickers indenter and a load of 10 kgf rather than with the Rockwell B tester and converting the data to Vickers. The upper limit of the Rockwell B scale is 100 HRB, about 240 HV. Because some of the hardness measurements exceeded these limits, use of the HRB scale should have been avoided. Further, there are no approved conversion scales for HRB to HV for hardnesses greater than 100 HRB and there are no approved conversion scales between HRB and HV for austenitic stainless steels.

Figure 6, for an A533 Gr. B steel with 0.19% C, which would have a maximum martensitic hardness of 44 HRC (434 HV, converted), shows that the experimentally developed hardnesses for cooling rates that produced only martensite were 437-440 HV (10 kgf load). For a cooling rate of 125°C/min, Fig. 6 shows a hardness of 250 HV (Curve 7). A cooling rate of 65°C/min (Curve 8, Fig. 6) gave a hardness of 237 HV. A cooling rate of 35°C/min (Curve 9, Fig. 5) gave a hardness of 230 HV. These structures were all bainitic.

According to Fig. 3 in Korth,² Samples G-8, F-10, E-8 and E-6 were all reaustenitized during the event and transformed subsequently to a bainitic structure with a hardness of 250-275 HV from a depth of ~5 to 40 mm below the interface. According to Fig. 5 of Korth,² these hardnesses would be produced by a cooling rate from the γ region of ~50°C/min for austenitizing temperatures from 800 to 1100°C, or for slower cooling rates for peak temperatures of 1000-1100°C. The CCT diagram of Kunitake *et al.*,⁷ shown attached as Fig. 6, was developed using a 920°C austenitizing temperature. These findings suggests that the cooling rates required to produce a 250-275 HV bainitic structure would be somewhat >125°C/min, not in perfect agreement but reasonably close.

4.3 Grain Growth in Heat-Affected Zone

Observations of the prior-austenite grain size (P γ GS) in the specimens was qualitative and appears to have been of minor value in establishing the thermal exposure history of the specimens. The P γ GS in the as-clad and stress-relieved head would vary because of the cladding process. Next to the fusion line, the grains will be quite coarse because the γ -grain coarsening temperature was exceeded. The P γ GS will become finer going away from the fusion line towards the region where the temperature just reached the Ac₃. The stress relief treatment will not alter this grain pattern. However, subsequent reheating above the Ac₁ will alter the pattern. The former coarse-grain region will recrystallize somewhat non-uniformly. The heating during the incident was not isothermal, but gradient. Consequently, when a temperature gradient is placed on the microstructure, with its gradient in grain size, recrystallization will be faster in some regions than in others.

The Korth report² made very little use of variations in the P γ GS of the A533 Gr. B head. Special etchants⁸ should have been used to preferentially delineate the boundaries. Then, a lineal analysis approach, with the line parallel to the interface, could have been used to measure the P γ GS as a function of the distance from the

interface. These measurements can be made with precision, despite the claim by Korth² (p. 19) that they cannot be made with precision. Although the aluminum content of the head was not well defined (the Lukens analysis in Ref. 6 does not list a value), it appears to be in the region of 0.02-0.04 wt.%. This would give a grain-coarsening temperature of 970-1020°C.^{9,10}

4.4 The "Carbon-Diffusion Zone"

Korth² noted the presence of a zone in the cladding, adjacent to the interface in Specimens F-10 and E-8, which he called a "carbon-diffusion zone." "SEM microchemical analysis" proved that the zone was enriched in carbon, apparently from the A533 Gr. B substrate. Korth stated that the hardness of this zone was "up to 500 DPH" and that it contained microcracks, indicating that it was brittle. However, the exact nature of this zone was not defined. It is not martensitic in appearance, and a 0.22% C, as-quenched martensite would not produce 500 HV anyway. Convergent beam electron diffraction of the extracted particles, and perhaps of the matrix using thin foils, is required to properly identify this zone. A complete identification of this zone might shed more light on the thermal history of these specimens. Selective etchants might be quite helpful. It is possible that sigma phase may be present in this zone. The brittleness of the sigma phase,^{11,12} its ability to form from δ -ferrite in austenitic stainless steel welds, and its hardness all make this a possibility.

4.5 Interfacial Dark Etching Zone

Korth² noted that the dark feathery band at the interface was not present in the E-8 and F-10 specimens (the interface was ground away on the E-6 and G-8 specimens during decontamination but examination in the ANL hot cell did not reveal the dark band). It was present on the archive specimens and all TMI-2 specimens believed to have seen temperatures $<A_{c1}$. The exact nature of this zone was not determined. Korth (Ref. 2, p. 19) believed that "it appears to be some sort of carbide." The microhardness of this zone was not stated. As it was quite narrow, the HRB tests probably did not assess its hardness. From the photo-micrographs, it appears that the dark feathery band is a smooth line on the A533 Gr. B side and ragged on the Type 308 stainless steel side. This zone is where dilution has occurred between the two alloys, producing some intermediate composition. Whatever the initial structure was, lath martensite and lower bainite are two possibilities; the structure was dissolved upon heating above the A_{c3} .

4.6 Carbon-Free/Carbide-Free Equiaxed Grains

In his second conclusion (Ref. 2, p. 20), Korth states that carbon diffusion from the alloy steel into the stainless cladding was observed in the accident heat-affected samples (F-10 and E-8) and that this was the mechanism for dissolution of the feathery carbide band and the formation of the cementite-devoid equiaxed grains. However, the nature of the dark feathery band in the weld dilution zone was never established. It was not proved to be from carbides (Why would this zone etch dark if

it were predominantly carbide?). The dark feathery band was more likely lower bainite or lath martensite tempered from the stress relief. The dilution zone would not be rich enough to be fully austenitic but its hardenability would be much greater than that of the A533 Gr. B head material, so martensite or lower bainite could easily form at these low cooling rates after cladding. If they had been carbides, why would a feathery structure persist after the 50 h hold at 610°C, which would surely cause spheroidization?

Furthermore, no evidence of cementite-devoid equiaxed grains is present in any TMI-2 specimen, F-10 and E-8 included. The hardness profiles in Fig. 3 of Korth² are not consistent with carbon- or carbide-free ferrite grains but with upper bainite, as shown in the photo-micrographs of E-8 and F-10 in Appendixes A and C of Ref. 2. In contrast, ferrite grains essentially free of cementite appear to be present below the interface in the Midland archive specimens (Appendix B of Ref. 2) heated at 900 and 1000°C. The 1100°C-1 min specimen has equiaxed, carbon-free ferrite grains, but other grains appear to contain carbides while those held for 10 and 100 min appear to contain carbides. In this respect, the isothermally heated archive specimens do not appear to match the gradiently heated TMI-2 F-10 and E-8 specimens.

4.7 Spheroidization of δ -Ferrite

Korth² noted that reheating to 1000°C for 100 min, or 1100°C for 10 min, dissolved the $M_{23}C_6$ carbide at the γ - δ interface and spheroidized the δ ; he did not note a reduction in the percentage of δ due to spheroidization. The Spanish¹³ and German¹⁴ independent studies both reported 1.4 vol.% δ (by magnetic tests rather than by metallography) in the E-8 specimens (m-6 and m-9 specimens, respectively) compared to 4 to 5 vol.% δ -ferrite in specimens that were heated to $<A_{c1}$. Table 1 lists their data.

The spheroidization of δ -ferrite in austenitic stainless steel welds has been studied¹⁵ for Type 316 (carbon content actually meets 316L) stainless steel. Figure 7 shows how the ferrite content decreases with time and temperature. In this work, f_0 was the initial ferrite content (9%), and f is the ferrite content remaining after exposure to high temperature. For the E-8 specimen, f/f_0 would be in the range of 0.28 to 0.35. If our time at temperature was limited to <60 min, Fig. 7 would indicate that temperatures of 1050-1100°C would be required, assuming that Type 308 responds the same as Type 316L stainless steel. This is in good agreement with Korth's conclusion for Specimen E-8.

Raghunathan et al.¹⁵ also examined the shape change with temperature and time (Fig. 8). The shape factor used, D/L , is the ratio of the average diameter to the length of the δ particles. A D/L of 1 (i.e., 100 in Fig. 8) represents a particle with a perfectly circular cross section. Again, for times <60 min, pronounced spheroidization requires temperatures of 1050-1100°C.

Spruelli et al.¹⁶ have shown that the δ ferrite content in Type 308 weld metal will be reduced at even lower temperatures. For example, aging at 750°C for 2 h reduced

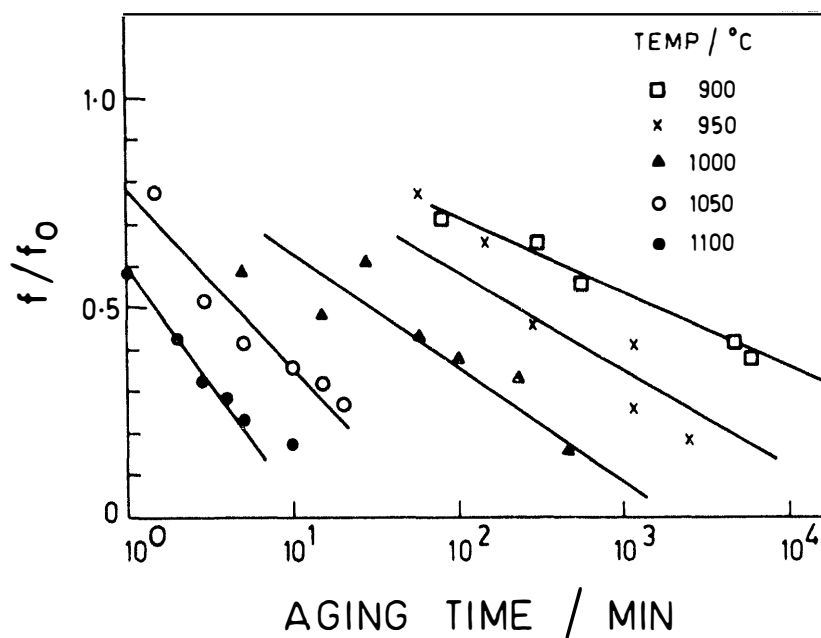


Fig. 7. Variation of fraction of δ -ferrite remaining with the logarithm of the aging time at various temperatures (from Ref. 15, used by permission of ASM International).

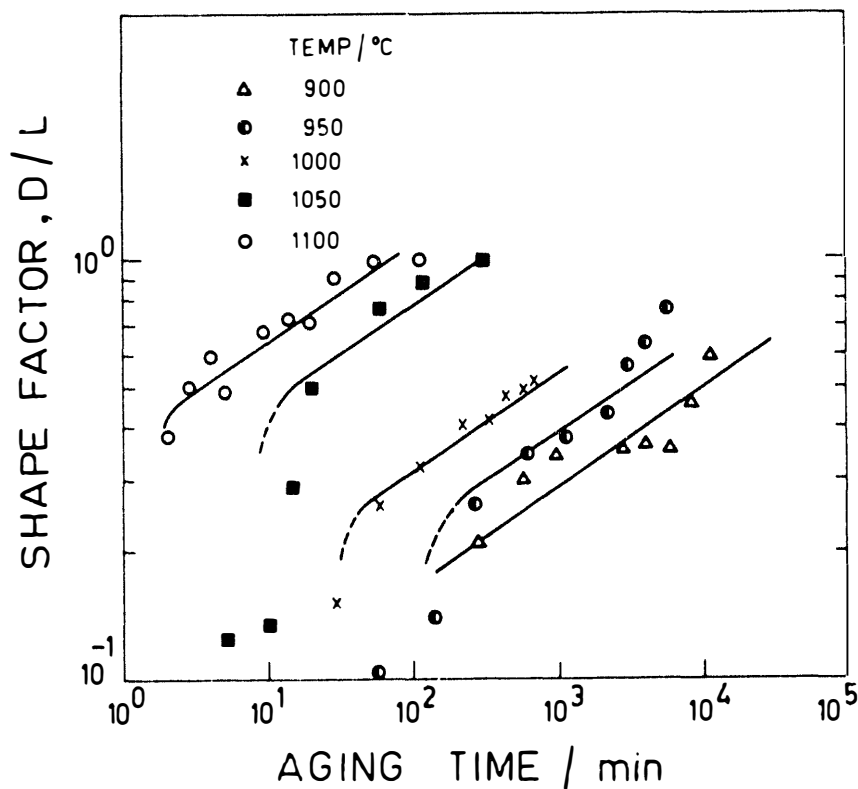


Fig. 8. Variation of the shape factor D/L with aging time at various temperatures (from Ref. 15, used by permission of ASM International).

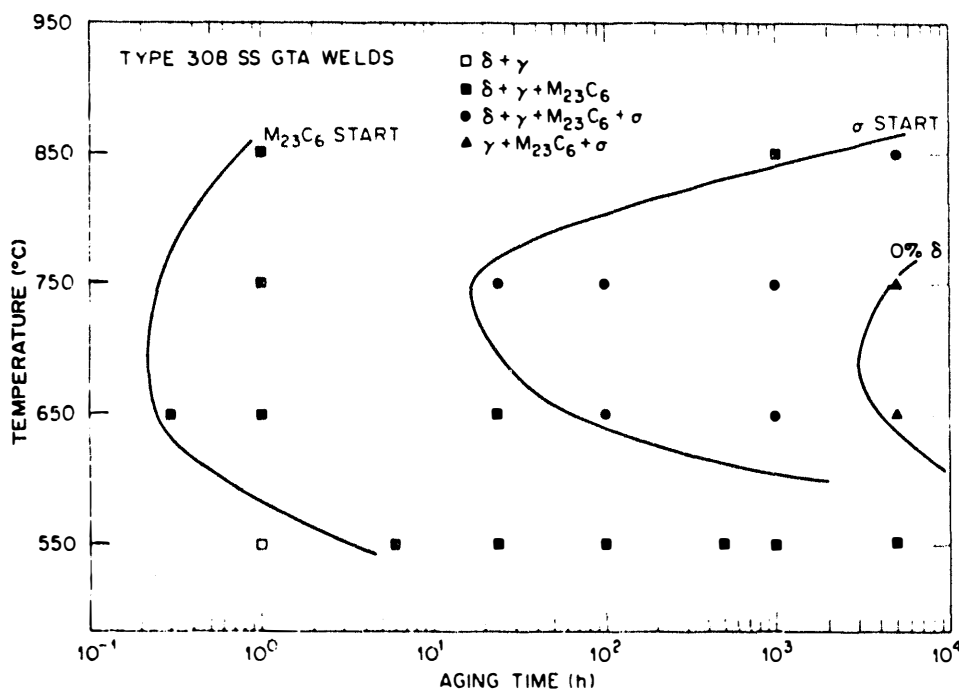


Fig. 9. Time-temperature-precipitation (TTP) diagram for welded and aged Type 308 stainless steel (from Ref. 18, used by permission of the American Welding Society).

the ferrite content by more than 60%. Vitek and David¹⁷ performed electron microprobe analyses of delta ferrite in aged 308 welds. They showed that, with increasing temperature, the Cr content of the δ decreased and the Ni content of the δ increased. Vitek and David¹⁸ also studied the influence of aging temperatures (550-850°C) upon the ferrite content of Type 308 weldments. They found that the rates of ferrite transformation at 650, 750, and 850°C were nearly the same. Aging for 1 h reduced the ferrite level to 60-70% of the initial level. Aging at 550°C had little effect on the ferrite content. Aging at these low temperatures generally produces precipitation of $M_{23}C_6$ carbide. The stress relief operation (50 h at 610°C) causes transformation of some of the as-clad ferrite content and precipitation of carbide at the γ - δ interfaces. Figure 9 shows the TTP curve of Vitek and David for precipitation in Type 308 weldments.¹⁸ This plot shows three carbon curves, one for the start of $M_{23}C_6$ precipitation, one for the start of δ precipitation, and the third for the completion of the δ -ferrite transformation. According to this diagram, δ precipitation is fastest at 750°C, with the reaction starting after 20 h. Time-temperature-precipitation diagrams for ferritic (and also for nonferrous) alloys have recently been collected and published;¹⁹ these diagrams can be very useful aids.

Horton et al.²⁰ showed that $M_{23}C_6$ would precipitate in ~10 minutes at 750-800°C in Type 316 stainless steel. Beckett²¹ showed that δ would form in 1 h at 750°C in a 24.85% Cr-8% Ni austenitic alloy. It is well known that σ forms much more rapidly from δ than from γ . Barcik²² showed that the rate of precipitation of σ from δ ferrite was almost two orders of magnitude faster than from austenite. Weiss

and Stickler²³ studied phase precipitations in wrought, solution-annealed Type 316L austenitic stainless steel. They noted that $M_{23}C_6$ forms most rapidly at grain boundaries, less rapidly at incoherent twin boundaries, and slowest at coherent twin boundaries. Smith and Farrar²⁴ have recently reviewed microstructure-composition-mechanical property relationships in austenitic, 300 series weldments.

4.8 Cladding Cracks

Korth² reported that cracks were observed in the cladding of Specimens E-6 and G-8. Because of difficulties in decontaminating these specimens, the cladding had to be removed and the microstructure was not available for Korth to examine and document. Cladding crack locations were mapped by Diercks (Fig. 30 in Ref. 6) showing that they were in the E-7 and G-6 quadrants, but none were in the G-8 quadrant. Specimen E-6 was oriented diagonally across the E-7 and F-6 quadrants and contained some of the corner of the E-6 quadrant. Specimen F-5 was the nearest one to the G-6 quadrant but its cladding was not cracked. In Fig. 2 of Ref. 2, Korth mapped out the location of the apparent hot spot on the lower head. If we assume that Fig. 30 of Ref. 6 was in error regarding G-6 vs. G-8, we must conclude that the cracking was a result of the high temperatures experienced in this region. It is possible that the cladding cracks formed as a result of the spheroidization and dissolution of the δ in this region, perhaps in a manner analogous to hot cracking in austenitic stainless steel welds that do not contain the critical amount (3-8%) of δ . Unfortunately, the clad surfaces of E-6 and G-8 were removed, so the degree of spheroidization and dissolution of the δ cannot be determined.

4.9 Conclusions

The overall approach of the Korth report² was sound. However, all of the observations were qualitative in nature, nothing was quantified. The overall conclusions appear to be reasonable, although it is difficult to disagree with qualitative data. Had the structures been quantified, then the conclusions could be debated or accepted objectively.

A weakness of the conclusions is that (p. 15, Ref. 2) all three areas (cladding, interface and low-alloy steel) of the standards were compared to those of the TMI-2 specimens. However, the standards were heated isothermally, but the reactor specimens were heated gradiently. Thus, the three areas of the TMI-2 specimens should be compared individually with those same areas of any of the control specimens. We should not expect to see conformance among these three areas of the unknowns with those of any single standard. Also, isothermal heating of Midland specimens at 900 and 1000°C produced carbon- and carbide-free equiaxed ferrite grains below the cladding interface, which were not present in the gradiently heated TMI-2 specimens.

Korth concluded (p. 15 of Ref. 2) that E-8 experienced 1100°C for 10-30 min (1075-1100°C for 30 min, on p. vii and p. 20) while F-10 experienced 1050°C (or 1040-1060°C, p. vii and p. 20) for 30 min. He also concluded that the thermal

excursion on the lower head was ~30 minutes (p. 19, Ref. 2). The basis of this latter conclusion was not given in the report. We assume that it was based on the event history, rather than on observations made by Korth in his study. Examination of the cladding of the control specimens for 1100°C suggests that the degree of δ -ferrite dissolution and spheroidization in E-8 is more typical of the 100-min exposure at 1100°C than of a 10-30 min exposure. It is also possible that this specimen experienced a somewhat higher temperature, if the thermal excursion was limited to 30 min.

The use of special etchants and quantitative microscopy would have added precision to this study, although the final conclusions may not have been that much different. The prior austenite grain boundaries in the A533 Gr. B head could have been preferentially delineated and the grain size could have been measured as a function of depth below the interface. This would be repeated using the standards. The grain size at any depth below the surface as a function of reheat temperature could be plotted and used to determine the temperature profile of the unknowns. Likewise, selective etchants could have been applied to the cladding to see what the structure of the carbon-diffusion zone was. Convergent beam electron diffraction by Auger electron microscopy would also be required. If σ or some carbide type other than $M_{23}C_6$ was detected, then this might lead to other conclusions. The δ phase in the cladding could be quantified and compared with that of the standards to better define the thermal exposure. Microprobe analysis of the δ could be made as well, as was done by Vitek and David.¹⁷ With these types of measurements available, the conclusions as to thermal exposure should be definitive.

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Curriculum Vitae

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Education:

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University of Illinois, College of Engineering	
Instructor, Metallurgical Engineering	1946-1949
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Associate Professor	1956-1959
Professor	1959-1962
Professor, Met. Engr. & Nuclear Engineering	1962-1981
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Honors and Awards

Tau Beta Pi, Sigma Xi, Alpha Sigma Mu (International President, 1970-71), Phi Kappa Phi, Pierce and Everitt Awards (Univ of Illinois), Who's Who in America, A. E. White Award (ASM International)

Professional Societies

American Society for Metals (Fellow), American Nuclear Society, American Society for Engineering Education, American Association for Advancement of Science

Courses Taught

Ferrous Physical Metallurgy, Nonferrous Physical Metallurgy, Phase Diagrams, Mechanical Metallurgy, Metallography, Fabrication of Alloys, Design of Engineering Alloys, Nuclear Power Engineering, Nuclear Metallurgy, Introduction to Metallurgy

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Summer Appointments, Argonne National Laboratory, Battelle Memorial Institute, Ames Laboratory, U.S. Steel, Hanford Laboratory, Caterpillar, Inc.; Adjunct Faculty, ASM International, Consultant, Alloy Engineering & Casting, in addition to numerous industrial, insurance, and legal firms

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Chairman, Corporate Chief Chemists Committee
Member, Corporate Cold Forming Committee
Member, Corporate Free Machining Steel Committee
Member, Task Group on Conditioning and Inspection
Member, Task Group on Tubular Products Quality
Co-author, Republic Alloy Handbook
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Professional Societies:

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Past National Chairman, ASNT Technical Council
Past Member, ASNT National Board of Directors
Past Vice-Chairman, ASNT Education and Qualification Council
Member and past member of numerous ASNT technical committees
ASTM Main Committee Member, E-46 (Quality Provisions in ASTM Standards)
ASTM Main Committee Member, E-4 (Quality Control)
ASTM Main Committee Member, E-7 (Nondestructive Testing)
Chairman, ASTM Sub-committee E-46.03.05 (Penetrant Standard Photographs)
Member and past member of numerous ASTM subcommittees
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Awards:

Alpha Sigma Mu (national honorary metallurgical society)
1982 Distinguished Merit Award, U. of Illinois Dept. of Mining and Metallurgical Eng.
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Publications:

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George received his BS in Metallurgical Engineering from Drexel in 1967—the last class as Drexel Institute of Technology. At Drexel he was twice editor-in-chief of the *Drexel Technical Journal*, including the three years in a row when it was named the number one college technical journal by the Engineering Colleges Magazines Association. He was a member of Blue Key National Honorary Leadership Society and in 1967 he was listed in Who's Who in American Colleges and Universities. Sigma Alpha Mu fraternity named him their outstanding senior in 1967. After graduation, George joined Bethlehem Steel Corporation in Bethlehem, PA. He received an MS degree in Metallurgy and Materials Science from Lehigh University in 1974 studying under Alan Pense, now Provost of Lehigh University. George worked six years in the metallurgy department of the Bethlehem Plant and was then transferred to their Homer Research Laboratories as a research engineer. Nine years were spent in the Metallurgical Service and Investigations group where George was involved in all areas of microscopy, failure analysis work and alloy development. In 1983, George joined Carpenter Technology Corporation where he is supervisor of Materials Characterization in their research center. He is responsible for all areas of microscopy and mechanical testing.

George has 88 publications, nearly all single authored, including the book *Metallography: Principles and Practices* (McGraw-Hill, 1984, 752 pgs.). He has edited, or was a co-editor, of thirteen other books and five special issues of *Materials Characterization*. He is the author of fifteen papers in various editions of the *ASM Metals Handbook* series and he made eleven of the fourteen video tapes in the ASM video course *Principles of Metallography*. He is the author of eight ASTM standards and holds five patents. His micrographs have been on the covers of 32 magazines, newsletters, brochures or calendars. He has won 18 awards for his work in metallography contests including the Jacquet-Lucas Grand Prize. He taught physical metallurgy at the Allentown branch of Pennsylvania State University for nine years and a graduate school course on microscopy at Lehigh University. He has taught over thirty one-week courses for ASM's Metals Engineering Institute (including courses for Aramco in Saudi Arabia, Rockwell International, Ford and United Technologies).

George has been active with the International Metallographic Society, IMS, for nearly 20 years, serving on the board of directors, as membership chairman, secretary, vice president and president (1981-1983). He was general chairman of their 12th annual meeting in 1979. He was series editor of *Microstructural Science* from 1983 to 1989 and he has chaired three of their symposia. He is associate editor of *Materials Characterization* (formerly *Metallography*) published by Elsevier. George received the IMS President's Award in 1987.

George has also been active with the American Society for Testing and Materials, ASTM, where he is a member of committees E-4 on Metallography and E-28 on Mechanical Testing. He has been chairman of E.04.14 on Quantitative Metallography since 1982 where he has developed and written standards for both manual and automated quantitative metallographic measurements. He has served, successively,

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George has been active with the American Society for Metals (ASM International) since 1966. He was a member of the executive committee of the Lehigh Valley Chapter (secretary, 1971-1974). A past member of the MEI and the Academy Committees, he is currently past chairman and member of the Technical Books Committee and a member and secretary of the Publications Council. He has been an MEI instructor since 1977 and a member of the Metallography Committee since 1982. He was made a Fellow of ASM in 1990. In 1993, he received the Bradley Stoughton Award of the Lehigh Valley Chapter.

George is also a member of TMS of AIME, the International Society for Stereology, and the Royal Microscopical Society (UK). He is married and has two children. When he has some spare time, George enjoys watercolor painting, photography, hiking and camping.

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