

## Summary

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This meeting of specialists continues a pattern of approximately triennial meetings on the subject of radiation embrittlement of reactor pressure vessel steels. As with the most recent prior meeting, the theme continues to be the understanding of radiation embrittlement of steels so as to better assure the structural integrity of the primary containment vessels of power reactors. The sponsoring organization within the International Atomic Energy Agency (IAEA), the International Working Group on Reliability of Reactor Pressure Components, has assured the continuity through periodic sponsorship of an appropriate meeting of specialists. The last most recent meeting was held in October 1981 in Vienna. The proceedings of that meeting were published by the American Society for Testing and Materials (ASTM) as Special Technical Publication (STP) 819. The contents of the present meeting reflect changes in the nuclear power business occurring since 1981; that is, it reflects two divergent trends, strong pressure from anti-nuclear groups coupled with escalating capital requirements opposed by a growing worldwide dependence on nuclear power for electricity production. The former fosters close regulatory control and a growing urgency to understand and overcome the potential negatives of radiation embrittlement. These two influences were evident in the contents of the meeting's formal papers and related discussion.

Forty-five specialists from 18 countries presented 21<sup>1</sup> formal and two informal papers in the two and one-half day meeting wherein detailed discussion was encouraged and became a critical part of the meeting's results. Besides a special final session for discussion to summarize and make recommendations, there were four technical sessions: I. Overview of National Programs, II. Surveillance and other Radiation Embrittlement Studies, III. Pressure Vessel Integrity and Regulatory Considerations, and IV. Mechanisms of Irradiation Embrittlement. The four formal sessions are reviewed briefly as a prelude to a more detailed statement of status and a summary of recommendations.

Session I was highlighted by national overviews from Czechoslovakia, the Federal Republic of Germany (West Germany), the United Kingdom, and the United States in which presentations collectively included: changes to regulatory

<sup>1</sup>For reasons of clearance limitations or others, several papers that were presented were not included in the proceedings. Nevertheless, this summary reflects all papers presented and related discussions as well.

guides or rules, delineation of research directions and needs, reactor design alternatives, and the means for dealing with transients. The revision to a crucial U. S. regulatory guide was based upon a new and growing base of data from the surveillance programs in U. S. reactors, but resulted in slightly more conservative data trends that seemed appropriate based upon data generated by the IAEA Coordinated Research Program and that of the German National Program. Nevertheless, the severe nature of possible transients, especially the so-called over-cooling accident that might lead to pressurized thermal shock in some U. S. reactors, was deemed crucial to decisions on both the regulatory guides and the direction of research studies that strongly emphasize data aimed toward answers in the latter area. Data presented by each author validated the value of controlling steel composition to reduce embrittlement and also to design new plants to reduce life-time exposure and, hence, embrittlement. The positive outcome of such presentations are to encourage the use of improved steels as well as design modifications to reduce the chance of placing welds in high fluence regions of the vessel and to generally reduce vessel exposure to neutron radiation both for current and future systems. Research directions emphasized by each national overview presented included, specifically or implicitly, need for added fundamental studies to better understand the nature of the embrittlement process, need for improved quantitative fracture approaches using small specimens, the need for criteria to produce a better quality of steels in terms of high initial toughness and low sensitivity to neutron radiation, and the need to be able to better model both physical processes involved and the consequences in terms of damage or embrittlement trends as a function of the neutron environment and the vessel steel involved. The latter are most important as they often form the basis for regulatory rules. Recommendations growing out of the whole conference impinge significantly upon the suggested directions of national programs.

Session II contained recent results of surveillance programs and research studies as well as descriptions of specific surveillance programs including a newly modified program of the Babcock and Wilcox Company in the United States and that used in India's Tarapur Reactor (with initial results) and Yugoslavia's Krsko Reactor. Specialists' described ways of obtaining the best possible fracture toughness data from small specimens by (1) extracting small notched tension specimens from remnants of tested Charpy specimens, (2) the use of side-grooved pre-cracked Charpy-sized specimens for  $J_{Ic}$  determination, and (3) the modification to impact test machines to allow measurement of dynamic fracture toughness. A massive systematic Italian program for predicting the reference transition temperature increase trends from irradiated specimens was described. Its basis is the chemical composition and related irradiation-induced changes for the steels studied. The results are expected to yield a better equation for establishing trends for embrittlement for regulatory and operational control purposes. The adequacy of various data bases and approaches to their use was discussed in detail considering the relative value of test reactor and surveillance data. The latter seem to have higher merit for trend limit establishment but a strong recommendation was made

for developing a "standardized" data base. This was judged by the specialists to be a proper, even necessary function of the IAEA as a unique international body having the resources, the charter, and the continuity necessary to data base maintenance.

The papers and discussion of Session III on Pressure Vessel Integrity and Regulatory Considerations rested heavily upon the U. S. Nuclear Regulatory Commission (USNRC) Regulatory Guide 1.99, its latest revision and the related German Rule, Safety Standard KTA 3203, which establishes a severe limit on projected fluence at end of life (nominal reactor life). Because of the relative stages of development of nuclear power in these two countries, the very different approaches were deemed to be feasible in each but not necessarily transferrable between the two. The modeling of an "overcooling accident" or pressurized thermal shock condition leads to the conclusion that radiation embrittlement as well as the existing or potential flaws and local stress conditions are of crucial interest on a plant-by-plant basis for those vessels of "old" or sensitive steel compositions. Underlying the papers presented and the discussion following were several strong recommendations for controlling embrittlement, limiting severe conditions of service, as well as criteria for assessing critically the potential for degradation to the point of possible failure.

Session IV concentrated on two main issues, the effect of annealing of irradiation damage and modeling this effect as well as the mechanisms underlying damage. One study of annealing showed nearly complete recovery at 454°C and, with reirradiation, found reduced sensitivity toward reembrittlement. The latter observation was contrary to some earlier results. Discussion of annealing, especially the modeling of effects on copper-defect centers led to the conclusion that much is to be learned about the phenomena affecting steel embrittlement. A brief report of the practical benefits of annealing the vessel of an operating reactor, the BR-3 at Mol, Belgium, was reported to have been fully successful based on annealing at 343°C (650°F) after the reactor's operation at 260°C (500°F). This operation was described as "wet annealing," that is, it was based on the use of the nuclear system to raise the coolant temperature to 343°C and to hold it there for one week. (This paper was offered only for discussion, not for publication.) Discussion of annealing, especially the USNRC attitude of its non-use in the pressurized thermal shock analysis led to questions of this attitude and a conclusion that annealing deserves further study and technological evaluation as a "last resort" means for reducing radiation embrittlement in operating reactors having vessels that are sensitive to radiation.

In spite of the early focus on annealing in Session IV, the main result of this session was to highlight the merits of advanced modeling and microanalytical techniques such as positron annihilation, analytical electron microscopy, and small-angle neutron scattering as tools of high potential for developing a better understanding of the physical bases for irradiation hardening and embrittlement. The coupling of statistical modeling procedures with data from microscopic techniques and parallel analysis of mechanical or physical properties shows spe-

cial promise for describing and even predicting steel response to irradiation. Correlations, incorporating physical hardening parameters and irradiation temperature produced coherent results from the most extensive data base available to date. Copper hardening centers were identified as the primary cause of radiation embrittlement but with nickel contributing in a way not yet well understood.

Besides the four focused technical sessions, a final half-day session was held to assess the current state of knowledge and to recommend directions for future efforts. The latter included specific recommendations for the IAEA Working Group as well as general and specific recommendations concerning the future research and surveillance programs attributable to individual organizations within member countries or for collective action. These are outlined beginning with the recommendations for collective action and following with topical reviews and recommendations for any and all interested parties.

### **Activities to be Supported by the IAEA**

The principal function of the IAEA is and has been properly to support collection, interchange, and dissemination of information that will aid the peaceful use of nuclear energy in a safe and efficient manner. The specialists continued to view these functions as primary to the IAEA's role in this subject but with the addition of some more direct support of research as well as of data analysis and storage.

The history of IAEA involvement in steel embrittlement by neutron irradiation is long and illustrious going back to sponsorship of the first major international conference on the subject in 1967. For this reason and because of the strong long-term continuity following the auspicious initiative in 1967, the specialists believe strongly in the merits of continuing the adopted role of the IAEA working group. Recommendations of the specialists relate to information dissemination as well as the encouragement and support of knowledge growth in critical areas. Principal recommendations to meet these goals were delineated:

#### *Recommendations*

1. Continue the series of specialists' meetings on a triennial (or more frequent) basis to review progress and research needs in the subject.
2. Assure publication of the results of specialists' meetings with appropriate analysis of results to maintain an international collective and authoritative statement of results and needs. Emphasis should continue to focus on safety-related developments and consequently to serve as a guide to those initially embarking on nuclear power development.
3. Commission special publications, as appropriate, to meet needs of the community for disseminating new research or surveillance data including an update of the volume published by the IAEA in 1975 entitled *Neutron Irradiation of Reactor Pressure Vessel Steels* (TRS 163).

4. Commission consultants' meetings, specialists' meetings, and conferences *as needed* to carry out the mandate of optimizing data dissemination.

5. Promote collaboration with other international bodies to assure optimum data release and minimal duplication of effort. In the development of data bases especially, coordinate with such organizations as the Organizations for Economic Cooperation and Development/Nuclear Energy Association (OECD/NEA), the independent International Association for Structural Mechanics in Reactor Technology (IASMiRT), the International Welding Congress (IWC), and the International Committee on Cyclic Crack Growth (ICCGR). (The latter is cooperating with the IAEA for joint sponsorship of a specialists' meeting in 1985.) These efforts are essential to assure meeting general goals just cited and for retaining the good national institutional cooperation that now exists.

6. In the realm of research encouragement and data acquisition, a program was undertaken by the IAEA in the early 1970s and is encouraged vigorously because of its' continuing high payoff. This effort was called the coordinated research program on Irradiation Embrittlement of Reactor Vessel Steels. It is now in Phase 3. The scope has grown from an effort to establish reproducible results on irradiation of one steel among several investigating institutes to a rather massive new program called Phase 3—"Optimizing Reactor Pressure Vessel Surveillance Programmes and Their Analyses", which is just now getting underway with about 25 different steels and 14 participating countries. Investigators in these countries use different approaches but will contribute to improved fracture criteria, neutron dosimetry and spectrum analysis, better knowledge of composition effects, better fundamental understanding of the physical bases for steel embrittlement and, in general, a better system of understanding of what comprises a good surveillance effort. International comparisons and collaborations will be a common feature of this study with interim reports issued at meetings of principal investigators.

The following is intended to approach the results and recommendations from a topical viewpoint and reflects largely the chairman's (editor's) views of the results and related recommendations. The *key subjects* are clearly radiation embrittlement, surveillance, assessing the fracture potential of vessel steels, and mechanisms of radiation damage. Related topics that are subordinate to these include neutron environmental analysis, thermal annealing to correct damage, the science of radiation damage, and the implications of service "transients" to vessel integrity. However, at the risk of being repetitive and raising the level of emphasis beyond that deserved, these subjects are treated as if they were integral parts of the total presentation and the related discussion. Primary emphasis, however, must reside on the four key elements discussed in the following sections.

### **Radiation Embrittlement**

New information dealt principally with the steel composition effects, comparison of research reactor and surveillance data, the means for measuring such

effects (neutron dosimetry, temperature, and fracture criterion used), the implications to reactor vessel integrity during service transients, and the mechanisms of the radiation damage process. While none of these topics is new, new results and indicators were cited.

### *Recommendations*

1. Looking back, the 1979 meeting dwelt heavily on data suggesting a "rate" or "saturation" effect that later was resolved without evidence of significant effect. The 1981 meeting dealt largely with the theme of independent or synergistic effects of residual or alloying elements, especially copper and nickel. The 1984 meeting introduced more sophistication to the latter without its resolution but with a strong emphasis on statistical and mechanistic studies to address "what levels of what elements" caused significant negative effects. The general consensus appeared to suggest serious damage if the copper level was over about 0.20% with nickel over 0.50%. Significant disagreement surfaced regarding the role of nickel at different levels. The strongest recommendation then came in the form of endorsing the Coordinated Research Programmes (CRP) directions to study a series of laboratory steel compositions having contents using both varying nickel and copper and using both conventional mechanical properties along with fundamental detectors for assessing the nature of the atomic level interactions. This recommendation is now built into the CRP Phase 3 both as to approach and to substantive content.

2. Phase 3 of the CRP program that was being finalized formally concurrently with the Specialists' meeting is not defined in detail but it should suffice to verify that carefully formulated laboratory and commercial-scale heats of varying steel compositions will permit elaboration on synergism or lack thereof between effects of copper, nickel and phosphorous.

3. An almost universal call of the specialists was for more directed scientific programs to assess the mechanisms of neutron interaction with the elements in typical reactor pressure vessel steels. This does not mean that no such results were available but the scope of selected variability within a given type of steel was inadequate to make most effective use of the tools such as field ion microscopy, analytical electron microscopy in general as well as special techniques such as small angle neutron and X-ray scattering and others now available for "seeing" the defects in irradiated steels.

4. New data showed some effects of temperature. Temperatures slightly lower than those typical of current light-water-reactor operation showed significant effects in terms of higher embrittlement. Full understanding of the role of temperature and time as well as strain aging effects, offsetting or abetting neutron exposure, is poorly understood. And, while not crucial for "standard" operating temperatures, it is part of the unknowns in the complex interactions of flux, temperature, time, strain, and steel types that will ultimately provide the "whole" story of neutron irradiation embrittlement. These should, therefore, be studied

from this view as well as the empirical one in which a specific reactor vessel operates at an "abnormal" temperature, whether lower or higher, than the general population.

5. One statistical study of data bank results in the United States suggests that, for reasons not clear, that data from surveillance of power reactor vessel embrittlement provides a more consistent base for analysis and projection of anticipated changes. This is encouraging in the sense that it suggests capsule integrity, good technique in dosimetry and related environmental analyses, but discouraging in that the most flexible tool for full study—research reactor experiments on an accelerated basis—may not be as useful as had been thought. However, the recommendation is that this analytical comparison be done with a weighting process that gives less credit to older research data where dosimetry might have been less well developed. Clearly, much more needs to be done in this realm. From this grew the recommendation that the IAEA assume and retain the role of data base proprietor not just for the CRP but for all available irradiation embrittlement data.

### **Surveillance**

The program content included surveillance from three perspectives, (a) content of national program, (b) redirection with new knowledge, and (c) data for comparison with the "world" data bank. Results came primarily from (a) and (c); the former involved descriptions from India and Yugoslavia both of which sought to enhance their programs by using small fracture specimens and the latter in connection with papers comparing special research data with that from power reactors. The volume of the latter and the redirection of programs (b) seem poised for very rapid growth.

### *Recommendations*

1. While surveillance programs may be locked into old technologies for a while, there is a clear and highly desirable trend toward modifying programs as was described by the Babcock and Wilcox representative to (a) add new fracture mechanics specimens and (b) to assure that the true "weak link" material is prominently included in key surveillance capsules.

2. All representatives endorsed the notion that surveillance is essential and should be updated as is possible. This means both improved specimens and dosimetry as possible. The goal of Phase 3 of the IAEA CRP will build nicely on this effort as the principal goal in this study is to optimize surveillance. Further, where the power level or core configuration has changed significantly for core management reasons or as a result of an overcooling accident and the related condition called pressurized thermal shock, new capsules may need to be added to assure that one can integrate effects over years that may involve significant core changes.

3. New or revised programs should capitalize on research developments just cited as part of ongoing studies. Of special interest is the best knowledge of chemical composition in order to have the best chance to choose the "weak link" material, to locate the capsule at the peak fluence location, to use the best possible miniature specimen for optimum fracture data, and ultimately to contribute to a better data base in the future.

4. Several papers described procedures for using small or modified specimens (before or after irradiation) for obtaining quantitative fracture mechanics numbers. Each surveillance program monitor should maintain his knowledge of such developments to optimize results from available specimens. For example, modifications of Charpy V-notch specimens to obtain  $J_{Ic}$  data have been described. (This type of correlation will be a major part of Phase 3 of the CRP.)

### Fracture Approaches to Vessel Steels

As just cited (Surveillance) several authors have developed criteria for using small specimens to define fracture potential in the vessel steels. This requires thorough evaluation; this means that the specimen must not be materials or geometry or size dependent. The critical nature of this factor is validated by the fact that this is a crucial part of the plan for Phase 3 of the CRP. The complexity of this task is best described by the fact that one must interpret the structural integrity of a vessel from laboratory tests of a few small specimens, while a direct one-for-one test approach is not possible short of the test of a full-sized irradiated vessel and correlated with fracture mechanics specimens taken from a failed vessel. Barring this possibility, it is crucial to be assured that the fracture analysis is based upon an approach that is clearly conservative (more severe) than can be expected in a real vessel.

### Recommendations

1. The overriding significance of this factor required a dedicated effort from all to arrive at a criterion within surveillance or research studies that meets the limits of conservatism implicit to safety.

2. Consideration of the American Society of Mechanical Engineers, Section III, Appendix G basis, the  $K_{IR}$  curve, continues to raise two key questions. First is there a  $K_{Ia}$  (crack arrest) number that can be defined in a standard manner for use in forming the "ductile" portion of the curve after irradiation? Second, what is the shape of the  $K_{IR}$  curve after irradiation for a sensitive steel? While extensive work was carried out on  $K_{Ia}$ , the need still exists for a standard approach. On the second point, large section tests of irradiated specimens and of scale vessels suggests the  $K_{IR}$  curve to be conservative relative to projected vessel behavior. Recommendations are still to use this approach but to be as careful as possible in use of small specimens to pinpoint  $RT_{NDT}$  on the  $K_{IR}$  curve while noting results for assessing the shape of the  $K_{IR}$  curve beyond the  $RT_{NDT}$  point.



3. In the use of quantitative fracture criteria,  $T$  or tearing modulus or a tearing instability factor was not used but rather a  $J$ - $R$  curve and  $J_{Ic}$  from small specimens seemed to be more universally used than ever before. It was recommended that refinements to define the better criteria should be watched carefully and applied as possible but with an effort always to seek a tie to the universally accepted Charpy V-notch specimen.

4. The several papers describing small specimens for  $K_{Ic}$  or  $J_{Ic}$  determination from criteria formerly applied to the Charpy cross-sectional limits suggested advances in the procedures based on side-grooving, especially ones that create the degree of constraint normally found only in much larger specimens. However, it is recommended that these new techniques be studied in Phase 3 of the CRP effort and that more than one organization cross check the approach on a given steel both before and after irradiation.

5. Because of comprehensive overview papers that all addressed the vessel's structural integrity from the view of fracture mechanics, especially as these may be affected by an overcooling accident (producing pressurized thermal shock) in some systems, great importance was and must continue to be placed on fracture analysis in future studies. This implies, as well, close collaboration with specialists in nondestructive testing and stress analysis in order that the three main factors be simultaneously considered. These are: (1) the vessel's irradiated toughness condition, (2) operating or transient stress, and (3) any existing flaw in a critical location of the vessel. These considerations were made more sensitive by the revelation that flaws of significant size relative to vessel wall thickness have been found in the *outside* wall of a U.S. vessel. This condition is made more serious because of the fact that  $K_{Ia}$  or crack arrest considerations were optimistically based on the presence of a flaw on the inside vessel wall. Recommendations were strongly pressed for obtaining the fullest knowledge possible of fracture toughness of a vessel at any point through its thickness and that more conservative considerations of projected flaw sizes and location (as well as the local stresses) be taken. Further, the significance of "low shelf toughness" *must* be quantitatively assessed by an internationally accepted "standard" criterion. Many early reactors are subject to severe analysis on this point. Such analyses can only become more critical with longer operating periods.

6. Analyses for the overcooling accidents that were presented lead once again to the need to minimize radiation and its embrittling effects by all means at the disposal of specialists and for radiation damage specialists to study ever more carefully the opportunities for changing the vessels' condition in service by all available means. (See Summary in *ASTM STP 819* for a review of this topic based on results available at the sister meeting in 1981.)

7. All of the preceding recommendations have crucial implications to criteria development by regulators for assuring structural integrity. There can be no letup in the study by specialists and these studies must integrate the technologies applying to vessel integrity assurance. For example, the question of pressurized

thermal shock (PTS) resulting from an overcooling accident must be related to fracture potential analyses for each reactor vessel, especially those wherein significant radiation embrittlement is possible because of "sensitive" steels or high projected neutron fluence levels during the anticipated life of the vessel. Plant specific analyses are essential to assess all factors that impinge on PTS and related potential for vessel fracture. Other accident or transient conditions that may heighten fracture potential must be assessed in conjunction with embrittlement probabilities.

### **Mechanisms of Radiation Damage**

The early studies and observations of radiation damage of steels were largely empirical in nature and, because of the potentially serious nature of embrittlement observed, studies concentrated on engineering implications in order to minimize the negative consequences of such effects in nuclear plants. Later, programs were undertaken to understand the phenomena involved. These highlighted the role of the residual elements, copper and phosphorus, then later other elements such as nickel, vanadium, and so forth. Empirical results pinpointed copper and nickel as serious actors in this drama and several studies sought clarification of their negative roles. Several papers at the meeting directed further attention to the complexity of this subject while validating the need for better knowledge in this area.

### *Recommendations*

1. Refined tools for microanalytical evaluation of irradiated steels offer opportunities that should be accepted and fully utilized. Techniques such as field ion microscopy, analytical transmission, and scanning electron microscopy, as well as small-angle neutron and X-ray scattering for assessing physical and chemical features of steels after irradiation, plus techniques that simulate neutron bombardment offer hope for further clarification of the embrittlement phenomena and were encouraged by meeting participants.
2. The ongoing CRP sponsored by the IAEA Working Group on Reliability of Reactor Pressure Components should focus attention on fundamental studies. Full use of the laboratory steels offered by Japan should permit better knowledge of the role of the cited elements (copper, phosphorus, and nickel) on the process of embrittlement. (It should be noted that nine of fourteen national participants in the Phase 3 CRP will be studying groups of specially formulated laboratory heats of steels containing differing levels of the elements considered most deleterious to properties of irradiated steels.)
3. Besides the composition oriented studies, others must emphasize the nature of the neutron-atom collision processes in order to complement our knowledge of the role of individual elements in the irradiated steels. This relates to the need for the best possible understanding of neutron interactions with the complex steel

matrix as a function of energy spectrum. The outcome should be two-fold: (a) a better understanding of neutron-produced defects, their stability, and mobility; and (b) better bases for describing the damaging potential of neutrons by energy level (or damage based dosimetry).

4. As knowledge is gained about both the microchemical and microphysical phenomena involved in the embrittlement process, it is desirable, even necessary, that such knowledge be applied to support criteria for specifying improved radiation resistant steels; for clarifying potential failure patterns, such as the through-wall crack arrest potential; and for minimizing radiation embrittlement effects through design modifications (core or vessel) or through thermal annealing.

5. Strong agreement was voiced by most meeting participants for a high priority effort in fundamental studies of steel embrittlement based on the feeling that only full knowledge would permit users to take actions necessary to minimize the negatives of this insidious process and to integrate steps for precluding serious consequences from occurring. Thus, good knowledge becomes the tool for avoiding the problem in new reactors and for ameliorating such effects in existing plants.

### General Conclusion

The optimism inherent to all phases of the meeting, especially the discussion and recommendations session, bodes well for the future of our hope for understanding radiation embrittlement of steels. Further, it reflects a global view of active positivism for nuclear power that this volume reflects both implicitly and explicitly. The recommendations cited are believed to be a distillation of substantive opinions on the topic. Strong support for research and development in the subject undergirds confidence in nuclear power. Further, an overwhelming impression was gained that the IAEA provides the best international outlet for information on the specific topic of irradiation embrittlement of reactor pressure vessel steels and, accordingly, that the sponsoring International Working Group must continue to serve this function and to support the knowledge base through the newly undertaken Phase 3 Coordinated Research Program on "Optimizing Reactor Pressure Vessel Surveillance Programmes and Their Analyses."

*Lendell E. Steele*

Head, Thermostructural Materials Branch, Material Science and Technology Division, Naval Research Laboratory, Washington, DC; specialist meeting chairman and editor.