MCC

**Materials Characterization Center
Workshop on the Irradiation
Effects in Nuclear Waste Forms**

Summary Report

January 1981

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory
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MATERIALS CHARACTERIZATION CENTER
WORKSHOP ON THE IRRADIATION
EFFECTS IN NUCLEAR WASTE FORMS

SUMMARY REPORT

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January 1981

Prepared for
the U.S. Department of Energy
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Pacific Northwest Laboratory
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1.0 SUMMARY

The Workshop on Irradiation Effects in Nuclear Waste Forms sponsored by the Materials Characterization Center (MCC) brought together experts in radiation damage in materials and waste-management technology to review the problems associated with irradiation effects on waste-form integrity and to evaluate standard methods for generating data to be included in the Nuclear Waste Materials Handbook. The participants represented Department of Energy laboratories, the National Bureau of Standards, universities, and private industry.

The workshop reached the following conclusions:

- The concept of MCC-6 for evaluating the effects of alpha decay is valid and useful. As a result of the workshop, modifications to the proposed procedure will be incorporated in a revised version of MCC-6.
- The MCC-6 test is not applicable to the evaluation of radiation damage in spent fuel.
- Plutonium-238 is recommended as the dopant for transuranic and defense high-level waste forms. When high doses are required, as in the case of commercial high-level waste forms, ^{244}Cm can be used.
- Among the important property changes caused by irradiation are those that lead to greater leachability, e.g., chemical changes that reduce durability and physical changes that increase surface area. Additionally, radiolysis of the leachant may increase leach rates; research is needed in this area.
- Ionization-induced changes in physical properties can be as important as displacement damage in some materials. A synergism is also likely to exist from the combined effects of ionization and displacement damage.
- The effect of changing the temperature and dose rates on property changes induced by radiation damage needs to be determined.

2.0 INTRODUCTION

The Materials Characterization Center, established by the Department of Energy (DOE) and managed by Pacific Northwest Laboratory (PNL), is responsible for ensuring the availability of reliable data on the properties of materials associated with the management of nuclear wastes. The data are needed for analytical models for safety analyses and licensing and decisions on the selection of materials.

To fulfill this responsibility, the MCC will collect and analyze existing data, perform additional tests where necessary, and develop and document procedures. The resulting data and procedures will be included in a Nuclear Waste Materials Handbook after approval by the Materials Review Board.

Since the procedures published in the Nuclear Waste Materials Handbook will have some of the characteristics of national standards, the scientific and technical communities are included in a review process to ensure that the proposed procedures are generally acceptable. As part of the review process, the MCC organizes workshops at which technical experts from universities, DOE laboratories, and private industry are invited to discuss issues relevant to data requirements and methodology and to critique procedures proposed by MCC. The first workshop on leaching of radioactive waste forms was held in February 1980, followed by a second meeting to examine leaching models. The third workshop, which is the subject of this report, was held July 29 and 30, 1980, and was entitled "Irradiation Effects in Nuclear Waste Forms." Twenty-four technical experts in the fields of irradiation effects in materials and waste-management technology participated. A list of the attendees is given in Appendix A.

2.1 PURPOSE OF THE WORKSHOP

The energetic nuclear radiations to which the waste forms will be subjected can alter the properties and may significantly change the durability of the waste forms over hundreds or even thousands of years. Therefore, laboratory evaluation of the anticipated changes must rely on accelerated testing to generate data that can be applied to predicting the long-term behavior of the waste form. This workshop provided a forum for scientific and technical experts to examine the issues related to irradiation effects and to provide input into plans for developing irradiation-effects tests. At the time of the workshop, the MCC had drafted a procedure evaluating the effects of alpha decay on waste forms. Copies of the proposed Standard Test for the Effects of Alpha-Decay in Nuclear Waste Solids (MCC-6)^(a) were given to the workshop participants for critiquing.

2.2 STRUCTURE OF THE WORKSHOP

The two-day workshop was structured around two goal-oriented discussion groups that met concurrently. Group I was assigned the tasks of critiquing the proposed MCC-6 test, assessing the need for additional tests, and evaluating the data requirements for predicting the long-term effects of radiation on waste forms. Group II was asked to examine the fundamental aspects of radiation damage, including the impact of beta, gamma, and transmutation effects and possible synergistic effects from combined ionization and displacement damage.

2.3 ARRANGEMENT OF THIS REPORT

The objectives of this report are to summarize the findings of the workshop and to document the recommendations relating to the proposed MCC test (MCC-6.) Section 3 summarizes the background information on the sources and effects of radiation in waste forms presented at the workshop. Section 4 consists of the summary reports from the two working groups, and Section 5 is a compilation of written comments from the workshop participants on the drafts

(a) The original title of MCC-6 was "Standard Test for Alpha - Recoil Effects in Nuclear Waste Solids."

of the summary reports and the proposed test. Names and addresses of workshop participants are given in Appendix A, and the workshop agenda is given in Appendix B. Appendix C is a draft of the proposed test that was given to the workshop participants on which to comment. (This draft has subsequently been revised to incorporate the findings of the workshop and to respond to the comments and recommendations of the workshop participants.)

3.0 BACKGROUND

In the United States, the inventory of nuclear wastes is predominantly from the nuclear weapons program--the so-called defense wastes. Defense high-level waste and transuranic (TRU) waste are the two waste categories containing sufficiently high levels of radioactivity for irradiation effects to be of concern.

The defense high-level wastes result from the chemical processing of uranium irradiated for producing weapons-grade plutonium. These wastes, which contain the bulk of the fission products, americium, curium, and the unrecovered plutonium and uranium, exist as liquids, sludges, salt cakes, calcines, and some separated and encapsulated strontium and cesium.

The defense TRU wastes result from plutonium fabrication and recovery in the weapons program and from ^{238}Pu and higher actinide production and fabrication. These wastes are in a variety of forms including sludges, ash, contaminated equipment, and organic materials such as contaminated paper, rags, rubber gloves, and clothing. The TRU wastes are currently being stored in retrievable containers. In the past, a large amount of such wastes were stored in shallow-land burial sites. Little of the TRU waste contains more than minor amounts of fission products and the actinide concentrations are low. Hence, the heat load is small, and the potential for radiation effects in the ultimate waste form is not great.

The only waste being generated by the commercial fuel cycle with potential for radiation-damage effects is spent fuel. The current national policy is not to reprocess spent fuel, and the only commercial high-level waste in existence is that stored at the Western New York Nuclear Service Center in West Valley, New York.

Plans are to place the wastes in geologic isolation after immobilizing them as durable solids. A variety of possible immobilization forms are under development. The stability of the forms in high-radiation fields needs to be characterized and understood to allow prediction of their long-term stability.

3.1 SOURCES OF RADIATION IN WASTE FORMS

The principal sources of radiation in the waste forms arise from the decay of the fission products (beta-gamma radiations)^(a) and actinide elements (alpha particles and recoil nuclei). In addition, damage may be caused by neutrons from alpha-neutron reactions and from fission fragments and by neutrons from spontaneous fission of the actinide elements. This damage can generally be ignored because of the low production rates. Projected numbers of beta, alpha, and neutron events per metric ton of heavy metal for a commercial high-level waste are shown in Figure 1 for storage times up to 10^6 yr.

The principal source of atomic-displacement damage is from elastic collisions between high-energy particles and the atoms of the solid. Calculated displacements/cm³ are given in Table 1 for a commercial high-level waste solid at two different exposures for alpha particles, alpha-recoil nuclei, beta, and gamma radiation. Figure 2 shows the estimated number of displacements at various times after emplacement in a repository. (The assumptions used in Figure 2 are the same as given in Table 1.)

The main sources of ionizing radiation in high-level waste are beta and gamma radiations from the decay of the fission products. Figure 3 shows the projected exposure for a commercial high-level-waste solid fabricated at 150 d, 5 yr, and 10 yr after reactor discharge. The decay of the fission products is essentially complete in a few hundred years.

Alpha-decay exposures of TRU wastes and defense high-level wastes are much lower than commercial wastes. Figure 4 shows expected alpha-decay exposures for the three waste categories. The data for TRU waste are a "worst-case" maximum containing 0.6 wt% Pu. Actual TRU waste is defined as waste having transuranic activities ≥ 10 nCi/g. The bulk of TRU waste has plutonium concentrations of 10^{-3} to 10^{-4} wt% and projected exposures are actually three to four orders of magnitude lower than the worst case shown in Figure 4.

(a) Fission-product decay also produces significant numbers of transmutations that can potentially cause radiation damage.

FIGURE 1. Cumulative Doses for a Commercial High-Level Waste

TABLE 1. Atomic Displacements in Commercial High-Level Wastes(a)

Radiation	Displacements per Decay	Decays per cm ³		Displacements per cm ³	
		100 yr	1000 yr	100 yr	1000 yr
Alpha Particles	140	5.0×10^{18}	1.5×10^{19}	7.0×10^{20}	2.1×10^{21}
Alpha-Recoil Nuclei	1500	5.0×10^{18}	1.5×10^{19}	7.5×10^{21}	2.2×10^{22}
Beta	0.13	2.0×10^{20}	2.3×10^{20}	2.6×10^{19}	3.0×10^{19}
Gamma	--	3.0×10^{11}	3.4×10^{11}	10^{18}	10^{18}

(a) Based on waste from light-water-reactor spent fuel
 - 33,000 MWd/MT
 - cooled 5 yr before reprocessing and waste solidification
 - 170 g solidified waste/MT.

(b) Gamma dose in rads.

FIGURE 2. Cumulative Atomic Displacements in Commercial High-Level-Waste Solid

Beta-gamma doses for defense high-level wastes are approximately 100 times lower than for commercial high-level wastes. Very little of the TRU waste contains any significant beta-gamma activity.

Spent fuel from light-water reactors is the major commercial-waste category. The beta-gamma doses for spent fuel are a factor of four lower than

FIGURE 3. Cumulative Dose from Beta and Gamma Radiation in Commercial High-Level-Waste Solid

FIGURE 4. Comparison of Expected Exposures for the Principal Waste Categories

the representative values in Figure 3, but the alpha-decay doses are greater because the plutonium is not removed.

An important consideration in understanding radiation damage from alpha decay is the distribution of actinides, which have several oxidation states and can be incorporated in different phases depending on the waste form and on the oxygen activity at the preparation temperature. Americium and curium are the dominant contributors to the alpha dose in solidified, commercial high-level waste up to $\sim 3 \times 10^5$ yr. Neptunium then becomes the major source, and at much longer times uranium becomes important. At no time does plutonium account for more than 20% of the total dose. Plutonium is generally the dominant source of alpha-decay damage in TRU waste and in the defense high-level waste at Savannah River Plant. Americium and plutonium are the principle contributors in the defense high-level waste stored at Hanford.

3.2. EFFECTS OF ALPHA DECAY

Alpha-decay results in two projectiles: an alpha particle with a range of $\sim 25 \mu\text{m}$ ^(a) and a recoil nucleus with a range of $\sim 100 \text{ \AA}$. The recoil nucleus interacts with the atoms by elastic collisions, producing atomic displacements along its path. The alpha particle loses energy by electronic interactions causing ionization; but near the end of its range, after dissipating much of its kinetic energy, it undergoes elastic collisions causing atomic displacements.^(b) The number of displacements can be estimated from the Kinchin and Pease model (Kinchin and Pease 1955); the recoil nucleus produces about 2000 displacements/event and the alpha particle about 100–500 displacements/event. Because of the short range of the recoil nucleus, it damages only the phase in which the alpha decays take place. However, the alpha particle can effectively bombard the entire waste solid if the actinide-bearing phase is uniformly distributed and the crystal size is small relative to the alpha-particle range.

(a) The range depends on the energy of the alpha particle and the waste-form composition.

(b) The probability for atomic displacements from alpha particles is low at high energies ($>0.5 \text{ MeV}$).

A list of potential actinide phases in crystalline waste forms (Table 2) indicates the diversity of structures that can be present. (In this listing, CaTiO_3 and $\text{CaZrTi}_2\text{O}_7$ are included as suggested host phases for the actinides.) Because the rare-earth concentrations are large compared to the actinide concentrations, they can be expected to control the actinide behavior. Actinide doping and alpha autoradiography have been used to observe the formation of actinide bearing crystalline phases in glass (Ross et al. 1979). These experiments showed that actinides always formed solid-solution phases with the lanthanides. This is also true for crystalline ceramics and supercalcine waste forms doped with curium.

Property changes in waste forms induced by alpha decay (combined effects of recoil nuclei and alpha particles) are observed by incorporating short-lived actinides (e.g., ^{244}Cm or ^{238}Pu) in the waste form and allowing it to damage by self-radiation. Properties that have been measured include

- density changes (Kelley 1975; Mendel et al. 1976; Mendel et al. 1977; Bibler and Kelley 1978; Ross et al. 1978; Ross and Mendel 1979; Rusin, Gray, and Wald 1979; Weber et al. 1979)
- microstructural changes
 - loss of crystal structure (Weber et al. 1979; Rusin, Gray, and Wald 1979)
 - cracking (Scheffler and Riege 1977; Bibler and Kelley 1978; Weber et al. 1979)
 - bubble formation (Bibler 1978; Hall 1976)
- stored-energy buildup (Roberts, Jenks, and Bopp 1976; Scheffler, Riege, and Hild 1976; Scheffler and Riege 1977; Mendel et al. 1977; Ross et al. 1978; Ross and Mendel 1979; Weber et al. 1979)
- leachability (Mendel et al. 1976; Hall et al. 1976; Mendel et al. 1977; Scheffler and Riege 1977; Bibler and Kelley 1978; Weber et al. 1979)
- radiolytic gas formation (Turcotte 1976; Hall et al. 1976; Bibler and Kelley 1978)

TABLE 2. Actinide/Lanthanide Phases Observed in Nuclear Waste Ceramics

<u>Idealized Phase Composition</u> (a)	<u>Probable Oxidation State</u> (b)	<u>Probable Structure Type</u>
MO_{2-x}	<u>4⁺</u> , 3 ⁺	Cubic
$Ca_2M_8(SiO_4)_6O_2$	<u>3⁺</u> , 4 ⁺	Hexagonal
$Ca_3M_7(SiO_4)_5(PO_4)O_2$	<u>3⁺</u> , 4 ⁺	Hexagonal
M_4O_7	<u>3⁺</u> , 4 ⁺	Cubic
MPO_4	<u>3⁺</u>	Monoclinic
$(Zr,M)O_{2-x}$	<u>4⁺</u> , 3 ⁺	Tetragonal
M_2O_{3+x}	<u>3⁺</u> , 4 ⁺	Cubic
$CaTiO_3$	<u>3⁺</u> , 4 ⁺	Cubic
$CaZrTi_2O_7$	<u>3⁺</u> , 4 ⁺	Cubic or Monoclinic
$NdAlO_3$	<u>3⁺</u>	Cubic
$2CaO \cdot M_2O_3 \cdot 2[(Fe_2O_3)_{1-x} (Al_2O_3)_x]$	<u>3⁺</u>	--
$M_2Te_3O_9$	<u>3⁺</u>	--
$(M)MoO_6$	<u>4⁺</u>	--
MbO_3	<u>3⁺</u>	--

(a) M = rare earths, actinides.

(b) Underlined numbers indicate most probable oxidation state

- mechanical strength (Mendel et al. 1977; Scheffler and Riege 1977; Weber et al. 1979).

Density changes for glass waste forms generally do not exceed 1% at saturation-damage doses. The glasses either expand or contract depending on the glass composition. Radiation-induced density changes tend to follow exponential curves (Figure 5). Similar behavior is seen for glass and glass ceramic of the same chemical composition as well as supercalcline SPC-2 (Figure 6) (Rusin, Gray, and Wald 1979).

Buildup of stored energy also follows similar exponential ingrowth (Figure 7). Stored energy at saturation damage is generally in the range of 80-130 J/g. Saturation damage as manifested by density change and stored energy usually occurs at about 6×10^{18} alpha decays/cm³ for most waste forms.

FIGURE 5. Density Changes in ^{244}Cm -Doped Waste Glasses

FIGURE 6. Density Changes in ^{244}Cm -Doped Glass, Glass Ceramic, and Supercalcine SPC-2

FIGURE 7. Stored Energy in ^{244}Cm -Doped Waste Glasses

Studies reported to date indicate that structural changes induced by alpha decay do not adversely affect leach rates in glass waste forms. Measured changes have all been less than a factor of two. Less information is available on waste forms other than glass, and it is premature to conclude that the effects are unimportant in all waste forms.

Bibler and Kelley (1978) reported that ^{244}Cm - and ^{238}Pu -doped waste glasses exhibited randomly oriented microcracking after exposures $>1.9 \times 10^{17}$ alpha decays/g of glass. The microcracks appeared to have no detectable effect on plutonium or curium leach rates. Multiphase materials may be more susceptible to cracking. A partially devitrified glass containing large crystals showed microcracking at an exposure of less than 0.8×10^{18} alpha decays/cm³ (Weber et al. 1979). The microcracking was attributed to volumetric expansion of the apatite phase ($>3\%$) as a result of radiation damage. The same material containing smaller crystals and a vitreous specimen of the same material showed no microcracking.

Experimental data on the effect of alpha decay on the crystal structure of ceramic waste forms are sparse. However, results from studies of the

crystal-lattice behavior of pure actinide and actinide-doped phases help in understanding radiation damage in crystalline waste forms. Data from some of these studies, summarized in Figure 8, show that the change in lattice parameter, a , as a function of dose generally follows an exponential relationship of the form:

$$\frac{\Delta a}{a_0} = A[1 - \exp(-k\lambda t)]$$

where λt is the absorbed dose and the constants A and k represent the saturation values and the rate constants for self-annealing. The fluorite-type actinide oxides (Figure 8) all saturate at low lattice expansions compared to the silicates and perovskite-type structures. Zircon, as well as apatite-, perovskite-, and pyrochlore-type structures all become X-ray amorphous at exposures in the range of $10\text{--}60 \times 10^{18}$ alpha decays/cm³ in contrast to fluorite-type dioxides for which amorphization has not been observed.

Experimental data reported by Weber (Chick et al. 1980) showing the effects of alpha bombardment on some materials with cubic structures are given in Figure 9. An important feature of these curves is that the lattice expansions for UO₂ and CeO₂ are much larger than those for PuO₂, AmO₂, and CmO₂ damaged by alpha decay (Figure 8). It has also been reported by Weber (accepted for publication in J. Nucl. Mater.) that fission-particle-induced lattice expansions are much smaller than those from alpha-decay damage, which in turn are less than those from external alpha-bombardment damage. These results are significant when considering radiation damage in the non-actinide-containing phases of crystalline waste forms.

There is little data available on damage ingrowth as a function of temperature, but considerable information on loss of crystal structure is available and was reviewed by Naguib and Kelley (1975). A list of the compounds studied is given in Table 3, along with the data on alpha-decay damage from Figure 8 (after conversion of the doses to a planar flux). The ionic materials tend to retain their crystallinity while the others become amorphous.

In summary, alpha decay in nuclear waste forms is probably the principal contributor to structural damage. Because of the small grain sizes and actinide partitioning in crystalline waste forms, the effect of alpha-particle

FIGURE 8. Lattice Expansion on Exposure to Internal Alpha Decay for Various Fluorite-Related Actinide Oxides

FIGURE 9. Lattice Expansions for Cubic Structures Bombarded with Alpha Particles Emitted from a $^{238}\text{PuO}_2$ Source

TABLE 3. Structural Stability Under Ion Bombardment
($10^{13} - 10^{17}$ ions/cm²)(a)

Materials that Remain Crystalline						Materials that Become Amorphous or Reduce to a Lower Oxide					
1 ⁺	2 ⁺	3 ⁺	4 ⁺	5 ⁺	6 ⁺	1 ⁺	2 ⁺	3 ⁺	4 ⁺	5 ⁺	6 ⁺
Cu ₃ O	BeO	YAl ₂ O ₃	MoO ₂		W ₁₈ O ₄₉	Ag ₂ O	CuO	Al ₂ O ₃	GeO ₂	Nb ₂ O ₃	MoO ₃
	CaO		PuO ₂			NiO		Bi ₂ O ₃	SiO ₂	Ta ₂ O ₅	WO ₃
	CoO		ThO ₂			PdO		Cr ₂ O ₃	SnO ₂	V ₂ O ₅	
	MgO		UO ₂					Fe ₂ O ₃	TeO ₂		U ₃ O ₈
	MnO		ZrO ₂						TiO ₂		
	NbO		AmO ₂						ZrSiO ₄		
	PbO	Am ₂ Hf ₂ O ₇							CmO ₂		
	VO							CmAlO ₃			
	SnO							Cm ₂ O ₃			
	TiO										
	ZnO										
		Fe ₃ O ₄									
Ti ₂ O ₃											
V ₂ O ₃											

(a) From Naguib and Kelley (1975).

and recoil-nucleus damage (i.e., alpha-recoil damage) in actinide-containing phases must be considered, as well as alpha-bombardment damage of all phases. Phases either remain crystalline, with small volume changes (<3%), or become amorphous, often with large volume changes. Ionic compounds appear to fall mainly in the stable category, and probable stability can be judged based on an estimate of ionicity.

With respect to dose dependence on damage level, changes induced by both alpha decay and alpha bombardment follow an exponential function to some saturation value. Neither the rate nor magnitude of ultimate changes can be estimated without experimental work. If commercial nuclear fuels are reprocessed and wastes solidified, the potential for radiation-induced changes needs to be evaluated. Defense waste, on the other hand, may never accumulate doses sufficiently high to produce serious radiation-induced changes.

3.3. EFFECTS OF IONIZING RADIATION

Much of the emphasis on radiation damage has been placed on the effects of alpha decay. However, the dominant radiation during the first 250–300 yr after fabrication of the waste form is beta and gamma radiation from fission-product decay. These radiations produce few direct atomic displacements but expend energy in electronic excitation resulting in ionization.

Ionization effects can be significant, but the impact on waste forms has not been thoroughly evaluated. Ionization reactions can produce structural changes; for example, ionization causes compaction in vitreous silica. The results of electron bombardment of vitreous silica (Primak and Kampwirth 1968) are given in Figure 10. The data show a density change of 0.3% at 3×10^{11} R, which is within the projected exposures for commercial high-level waste shown in Figure 3.

In electron-bombardment experiments at Harwell, England, using borosilicate glass, three glass formulations irradiated to 3×10^{11} R resulted in leach-rate increases of less than a factor of 2 and a density change of 0.2%. In Japan, a similar study showed leach-rate changes of no more than a factor of 3. The doses used approach the life-time beta doses expected for commercial high-level wastes and greatly exceed the life-time doses of defense high-level wastes, yet the effects on the leach rates are not large.

The effects of ionizing radiation can result not only in structural changes within the waste form, but can also lead to changes in the chemistry of the leachant. Both of these factors can affect the chemical durability of a waste form in an aqueous environment. Radiolysis of nitrogen in the presence of water can produce nitric acid (Jones 1959). Water radiolysis may form a variety of reactive free radicals and ions as well as hydrogen peroxide (Burnes and Moore 1976). McVay, Weber, and Pederson (1980) recently reported that significant increases in the leach rate of a complex, simulated waste glass were observed in the presence of gamma radiation. In that study, nitric acid, generated from air radiolysis in the leach vessel, appeared to preferentially attack zinc and lanthanides. When air was excluded to eliminate nitric-acid formation, the leach rates were still greater when in a gamma field, indicating that water-radiolysis products are also an important factor in leaching of waste forms.

FIGURE 10. Dilatation of Suprasil on Electron Bombardment
(Primak and Kampwirth 1968)

3.4 EFFECTS OF TRANSMUTATION

Another potential source of radiation-induced changes is the transmutation of the fission products. The sources of the two principal transmutations are the intermediate-lived fission products, ^{137}Cs and ^{90}Sr :

Transmutations can involve valence changes and major changes in atomic radii. Glass waste forms are likely to tolerate these changes since they already have 10-25 elements present, including substantial concentrations of barium and zirconium. Ceramic waste forms have ordered structures and are less able to accommodate this kind of change than glass.

A study is underway (Weber, Wald, and Gray 1980) to evaluate the effect of transmutation on waste forms, including

- three glasses
- pollucite
- supercalcline SPC-2.

These simulated waste forms were fabricated incorporating excess ^{133}Cs and then were irradiated in the High Flux Isotope Reactor (HFIR). The ^{134}Cs formed from the reaction

is now decaying, and post-irradiation testing is underway. Preliminary results indicate no significant effect of transmutations on waste-form stability. This study is continuing.

4.0 WORKING GROUP REPORTS

This section contains reports of the individual working groups. The reports were prepared by the discussion leaders, who incorporated comments from the working-group participants.

4.1 WORKING GROUP I: PROPOSED TEST FOR ALPHA-DECAY EFFECTS IN NUCLEAR WASTE SOLIDS

The assignment for Working Group I was to critique the proposed MCC test for the Effects of Alpha Decay in Nuclear Waste Solids (MCC-6) and to recommend any changes. The participants were as follows:

Frank Roberts, Discussion Leader, MCC
Bill Weber, Discussion Leader, Pacific Northwest Laboratory
Tony Cheng, National Bureau of Standards
Larry Crisler, TRU Waste Systems Office
Dave Howitt, University of California
Vic Kelsey, ARAI/EG&G
Ed Lewis, Mound Facility
Bill McDonnell, Savannah River Laboratory
Jim Merrill, MCC
Ron Nelson, MCC
Harry Pearlman, Rockwell International
Bob Schuman, Exxon Nuclear Idaho Co., Inc.
Rich Van Konynenberg, Lawrence Livermore Laboratory

4.1.1 Introduction

The working-group discussion began with short presentations by several of the group members on different irradiation and measurement techniques employed in their laboratories. The group then focused on five major areas of discussion:

- the proposed irradiation technique and its limitations
- dose requirements
- irradiation temperature

- the proposed test procedures for property measurements
- the applicability of the proposed test to spent fuel.

Discussion on these topics is summarized below.

4.1.2 Proposed Irradiation Technique

There was a consensus that the actinide-doping technique for irradiating test specimens is an appropriate technique for a comparative test of the effects of alpha decay on the physical and chemical properties of nuclear-waste solids. Such a test does not simulate transmutation effects nor the potential synergistic effects resulting from concurrent alpha, beta, and gamma radiation. Several techniques for studying these effects were proposed and briefly discussed; however, the group recognized that there are insufficient experimental data to justify incorporating these techniques into MCC-6.

There was criticism that MCC-6 emphasized the use of ^{244}Cm as a dopant. In many cases, the use of ^{238}Pu may be advantageous and in some instances may be mandatory, particularly in multiphase systems if the actinides partition into different phases because of their valence states. The use of ^{238}Pu as a dopant in tests of defense waste forms may be preferred because the dominant source of alpha decay in defense wastes is from plutonium. The group recommended that MCC-6 be revised to clearly indicate that the use of either ^{244}Cm or ^{238}Pu is acceptable. The selection of one dopant over another should be based on a thorough understanding of actinide behavior in the waste form under consideration, information on the principal alpha-emitting actinides in the waste, the availability of the dopants, and the dose levels that are to be achieved.

4.1.3 Dose Requirements

Some property changes induced by radiation (e.g., density and stored-energy changes) appear to reach maximum values at doses of $\sim 6 \times 10^{18}$ alpha decays/cm³. MCC-6 originally required that a fluence of this magnitude be achieved in about a 1-yr period, but proposed defense waste solids will not even approach fluences of this level in 10^6 yr, the time period of interest. For this reason, several group members recommended relaxing this dose

requirement in those waste forms specifically intended for defense waste. However, it is recognized that radiation effects can be better understood (at little extra cost) by carrying the studies out to dose levels at which saturation effects are generally observed.

4.1.4 Irradiation Temperature

The temperature at which the specimens are to be maintained during the course of the proposed test was considered to be a key test condition. The proposed use of room temperature is arbitrary; but if only one temperature is to be used, then room temperature is a logical choice, particularly for defense waste forms. The temperature of the defense wastes will approach the ambient repository temperature after a few hundred years, during which time the effects of alpha decay should be negligible. It is only over the time period approaching 10^6 yr that the effects of alpha decay will be important. During most of this time, the defense wastes will be at the ambient repository temperature. Therefore, room temperature should be used but a slightly wider tolerance should be allowed, $23^{\circ} \pm 5^{\circ}\text{C}$, to be within the normally observed temperature variations of a glovebox.

Maximum effects of alpha decay may occur at the elevated temperatures that the wastes will experience during early storage in the repository. The temperature at which any given effect is at a maximum may vary from one waste form to another, and this temperature may be different for different properties. As a result, it is difficult to identify what additional temperatures, if any, should be considered as test conditions or even what criteria should be used for temperature selection. It was suggested that MCC-6 include a disclaimer relative to its applicability at high temperatures until experimental results regarding temperature dependence can be evaluated.

4.1.5 Proposed Test Procedures for Property Measurements

After specimens of a waste form are prepared by actinide doping, it is necessary to measure and evaluate the effects of alpha decay on the properties and the long-term integrity of the waste form. The specific tests to be performed and the properties to be measured were controversial issues. Some group members indicated that their laboratories did not have the facilities to carry out some of the required test procedures (such as X-ray diffraction) and

preferred that MCC-6 be rewritten to allow alternative tests or to at least make most of the tests optional.

Change in chemical durability (leachability) resulting from irradiation damage was recognized as the most important property to be evaluated by MCC-6 and one that must be evaluated by the same test procedure by all laboratories adopting the proposed test. There were, however, differing opinions on how a leach test should be performed. The suggested procedure is to measure leachability by using the proposed MCC-1 Static Leach Test. In this test, a monolithic specimen of the waste form is immersed in a static leach solution for a predetermined period at a constant temperature. The leach tests of the specimens are to be performed at the beginning and at the completion of the irradiation. Some felt that leach tests of such short duration do not simulate the radiolysis occurring at the specimen-solution interface. Instead, they proposed that a continuous leach test in the radiation environment provided by the specimen would be more appropriate to evaluating the synergistic effect of radiation damage in the specimen and radiolysis at the specimen-solution interface. This issue was not resolved.

The usefulness of a mechanical strength test in MCC-6, particularly the proposed diametral-compression test, was questioned. The consensus was that compressive strength is not really an important concern after emplacement in permanent storage and that alpha-decay effects would be negligible prior to final emplacement. If a mechanical strength test is to be performed, a different test may be used such as a microindentation technique, where hardness and fracture toughness are determined by the dimensions of the indent and crack patterns around the indentation, respectively. This method is neither easily applied nor interpreted in the case of ceramics.

In preparing specimens for leach testing, they should be cut with a diamond saw with no surface preparation other than thorough cleaning as prescribed by MCC-1. The leach specimens should not be sectioned with the diamond saw until the leach tests are to be conducted; in this way, only fresh surfaces will be exposed to the leach solution. Specimens subject to nondestructive testing could be used for more than one type of measurement, thereby reducing the total number of specimens needed.

The measurement of thermal-conductivity changes is not included in MCC-6. It was suggested that the effect of a decrease in the thermal conductivity of nuclear-waste solids could be modeled and, if there is a significant effect, measurements of the change in the thermal conductivity should become a procedure within MCC-6.

In summary, a test of leachability is the single most-important procedure to be required, but the issue of the best method of measuring leachability was not resolved. With regard to other test procedures, volumetric changes should be determined and as much microstructural and structural analysis as possible should be performed (e.g., X-ray diffraction, electron microscopy, and alpha-autoradiography) within the capabilities of each laboratory. The group consensus was that stored-energy and mechanical-strength measurements may be unnecessary but might be included as optional tests.

4.1.6 Applicability to Spent Fuel

The consensus of the working group was that the additional radiation effects from alpha decay in spent fuel would be insignificant compared to changes induced by in-reactor radiation. However, there is clear evidence, based on self-damage studies of $^{238}\text{PuO}_2$ and on a study of alpha-bombarded UO_2 , that much higher point-defect concentrations can be introduced by alpha decay than by fission damage. Therefore, studies are needed on the effects of prior fission-induced damage on the ingrowth kinetics of defects from alpha decay.

The working group agreed that there is no practical way to apply MCC-6 to accelerated testing of alpha-decay damage in spent fuel. Other methods for evaluating the effects of alpha decay in spent fuel are to evaluate HFIR fuel, which contains a high concentration of short-lived actinides, to study accelerated effects of alpha decay, and to evaluate data from the Oklo natural reactor.

4.2 WORKING GROUP II: GENERAL RADIATION EFFECTS IN WASTE FORMS

Working Group II was asked to examine the fundamental issues of irradiation effects in nuclear waste forms. This included the magnitudes and relative importance of displacement damage, ionization damage, and effects of transmutation. In addition, the group was to assess the practical and theoretical

significance of radiation-induced property changes. The participants were as follows:

Ray Turcotte, Discussion Leader, MCC
Alan Ardell, University of California, Los Angeles
Lynn Boatner, Oak Ridge National Laboratory
Frank Clinard, Los Alamos Scientific Laboratory
Rodney Ewing, University of New Mexico
Linn Hobbs, Case Western Reserve University
J. Read-Holland, Westinghouse Research and Development Center
Paul Levy, Brookhaven National Laboratory
Pete Macedo, Catholic University
Rich Strickert, Pacific Northwest Laboratory
Lou Vance, Penn State University.

4.2.1 Introduction

Discussions began with short presentations by each group member on some aspect of irradiation effects important in disposal of nuclear wastes. Discussions of these and other issues are summarized in the following sections dealing with

- general stability
- ionization damage
- transmutations
- temperature and dose-rate dependence
- leaching effects
- damage simulation
- other issues.

4.2.2 General Stability of Solid Wastes

The parameters of major concern are property changes that alter chemical stability or that increase surface area. Both of these changes potentially lead to increased dispersion in the environment. In evaluating the structures of crystalline waste solids, diffraction methods alone are inadequate in the absence of other microstructural information, such as that obtained by transmission electron microscopy. Displacement damage, produced by alpha-recoil nuclei in particular, may not lead to simple production of point defects.

Clustering phenomena, such as generation of metallic precipitates, as occurs in NaCl and CaF₂, void formation as in Al₂O₃, or formation of gases may be more important than changes in point-defect concentration measured by lattice parameter studies.

Diffusion-related changes concerning possible migration of atomic species were discussed, including the migration of helium to such internal surfaces as grain boundaries. These effects would clearly be more pronounced at higher temperatures. Observation of such changes require electron-microscopy studies. However, observation of defects by transmission electron microscopy (TEM) is not a trivial matter in oxide systems because of specimen-preparation difficulties and changes that can be introduced by the electron beam itself unless precautions are taken such as use of a specimen cold stage. Few TEMs are available for use with radioactive specimens. In addition, unexpected phases may appear in radiation-damaged systems because of enhanced mobility, nucleation, and solute segregation.

Evaluation of structures for topological constraints could improve understanding of why some structures are stable and others are not. This approach may be more applicable to isolated defects, such as those induced by ionization processes, rather than high defect concentrations from alpha-recoil displacements. (Displacements result from the alpha particle and recoil nucleus created by alpha decay.)

Observation of exponential changes in any property as a function of dose may be useful, but cannot be reliably interpreted without understanding structural changes at least at the submicron level.

4.2.3 Ionization Damage

For some materials, ionization-induced changes could be more important than displacements caused by alpha decay. In particular, silicate-based systems could be susceptible to ionization damage. Some experimental results show large microstructural changes in quartz, mica, and zeolites caused by electron bombardment. Doses in the range of 10^{12} R could lead to ~0.1 displacement per atom (dpa) in materials that are susceptible to ionization damage. In some cases, a relatively low gamma exposure ($\sim 10^7$ R) measurably increases leach rates up to a factor of two. The post-irradiation leach rates return to their

pre-irradiated value after a few days. These results support the premise that purely electronic processes may be important, although changes in leach rates less than a factor of two are generally not considered serious.

Although the proposed actinide-doping studies provide large ionization doses from the alpha particles, studies are needed to evaluate ionization-induced changes independent of alpha-recoil studies. Electron bombardment and high-dose-rate gamma exposures should allow observations up to the 10^{12} R range. Studies of ionization-induced changes may be necessary in interpreting alpha-recoil damage or more complex situations, for example, in work undertaken to evaluate synergistic effects resulting from interactions of alpha events with beta and gamma irradiations.

4.2.4 Transmutation

More experimental data are needed to understand the potential adverse effects of transmutation resulting from the decay of fission products. Studies similar to those being performed at PNL (Chick et al. 1980) involving the Cs $\rightarrow$ Ba transmutation are appropriate. Observations in the 100^o-200^oC range are needed because the bulk of the transmutations, e.g., $^{137}\text{Cs} \rightarrow ^{137}\text{Ba}$ and $^{90}\text{Sr} \rightarrow ^{90}\text{Y} \rightarrow ^{90}\text{Zr}$, occur during the early storage period when the waste form is thermally hot.

4.2.5 Temperature and Dose-Rate Dependence

Discussions of aspects of irradiation-induced property changes emphasized the need for understanding both the temperature dependence and dose-rate dependence, since the two are related. The primary issue for these studies is defining the roles played by point defects and by the formation and growth of aggregates of vacancies, interstitials, or gases. Property changes should be evaluated in terms of the reduced temperature $(T/T_m)^{(a)}$. In systems forming defect aggregates, a temperature of maximum production efficiency will exist depending on the competitive mechanisms for removal of the point defects--diffusion/aggregation versus recombination (annihilation).

(a) The reduced temperature is the ratio of the absolute temperature of the observation to the absolute melting point.

Experimental observations of the dose-rate dependence for radiation damage of any kind is meager in oxide ceramics. The issue was considered to be important because tests done to simulate long-term irradiation employ dose rates many times greater than will be experienced by real wastes. A useful study of the importance of dose rate might be to use ^{238}Pu doping to evaluate changes in zircon, relating the results to previously published information on metamict zircons where the damage occurred over $\sim 10^9$ yr. Such an experiment is planned by W. J. Weber of PNL.

4.2.6 Leaching Effects

Leaching studies of radiation-damaged material are important and solution-radiolysis effects on leach rates require study. Although discussion of leach behavior was limited in order to focus on solid-state radiation effects, several relevant points were made:

- Studies were cited showing that alpha- or gamma-induced radiolysis can produce nitric acid in air-saturated water. A decrease in pH values to as low as pH = 2 resulted in significant leach-rate changes, particularly for normally insoluble species such as the rare-earth elements, and in much-increased actinide solubilities.
- Waste-glass leaching can cause the actinide concentration to increase at the surface. There can be two important consequences: increased radiation damage to either the gel or surface layers and increased radiolytic attack. This suggests that long-duration leaching studies are needed, particularly for actinide-doped materials.

4.2.7 Damage Simulation

Ion-bombardment or fast-neutron irradiation techniques can provide rapid, meaningful information on some aspects of radiation damage. There is a limitation in using this approach to study some polyphase ceramic waste forms, since all of the phases are damaged rather than just the actinide-host phases. Use of fission damage to irradiate waste materials does not appear to be a promising means for simulation of alpha-recoil damage, although the method may provide a convenient mechanism for producing the fully damaged, amorphous form of some structures.

4.2.8 Other Issues

In addition to the above topics, some attention was given to the need for studying

- leachability in gamma fields
- potential synergistic interactions such as strain, radiation-induced defects, and thermal gradients
- radiation-induced changes in thermal conductivity.

5.0 COMMENTS BY PARTICIPANTS

The participants in the workshop reviewed and commented on the drafts of the working-group summary reports and the proposed procedure (MCC-6). Revised drafts that responded to these comments were subsequently submitted to the participants for review. This section presents the formal comments received from the participants. In addition, short comments suggesting changes that were not too extensive were received. In most of these cases, the suggested changes were made but are not noted in this section.

J. Read Holland – Westinghouse Research and Development Center

1. In considering a mechanical-strength test for MCC-6, the suggested micro-indentation technique raises serious questions with respect to interpretation and utility of the data. Because of these problems, microindentation is of questionable merit. A flexure test, such as four point bending, to failure could yield meaningful data on resistance to fracture, fracture stress, and strain to fracture. A disadvantage would be the necessity to cut bend bars from the specimen discs.
2. Damage Simulation – Dual ion bombardment, using simultaneous irradiations of He ions and medium to heavy ions, has proven to be a useful technique for simulating 14 MeV neutron radiation damage. Alpha damage has not been studied using dual ion bombardment, but this simulation technique should be readily adaptable to simulation of alpha damage in nuclear waste forms. The technique lends itself to separating He and dose-rate effects and other fundamental issues related to understanding the damage processes. In addition, the method could be used to quickly determine the magnitude of radiation-induced changes in properties such as chemical durability because the simulated damage can be accumulated in a matter of hours as opposed to months for Cm^{244} or Pu^{238} doping. The ion-bombardment method should be considered a potential method for complementing the planned doping technique; it is not a viable substitute. The inherent characteristics of a limited ion range in solids and homogeneous distribution of ion-induced damage in all phases of a multiphase solid should be taken

into account in planning ion-bombardment experiments on surrogate waste forms. While these limitations are real, they do not preclude meaningful use of ion bombardment in studying radiation-damage effects in many waste forms.

Harry Pearlman - Rockwell International, Energy Systems Group

Proposed Irradiation Technique

It is expected that the effects of transmutation on waste-form stability would be greater in a crystalline than a glass composition because of the long-range (as well as short-range) order associated with the crystalline state. For example, the valence change accompanying the transmutation of ^{90}Sr to ^{90}Zr would probably be more easily accommodated in glass than in a crystalline solid. There, the effects on the lattice of the changes in the cation size and field could influence properties of an entire phase (or phases). Overall integrity could be affected, as well as density and, especially, leachability. It is true that the half-lives involved are relatively short, so these transmutations occur while the waste form is still thermally hot. However, it is not obvious how any annealing that might occur would affect results of the fundamental change in chemistry.

The above considerations apply to the class of waste forms of which tailored ceramics and Synroc are examples. The particular effects that are of concern do not appear to be simulated well by doping with a short half-life actinide. This is especially so with the tailored ceramics formulation of Savannah River Plant waste, where essentially all of the actinides are expected to be concentrated in a uraninite phase. The majority of the fission products, where most of the transmutations occur, is expected to concentrate in other phases. A different technique has been proposed to simulate the transmutation effects in this waste form. Samples prepared with lithium-6 and/or boron-10 are to be irradiated in a nuclear reactor, where thermal neutron nuclear reactions effect transmutations. Some alpha-particle generation also results; and fast neutrons produce lattice displacements.

Robert P. Schuman - EG&G Idaho, Inc.

Comments on Proposed Standard Test, MCC-6

Standard Test for Alpha-Decay Effects in Nuclear Waste Solids

I feel the test is a good start. I have a number of comments but most are questions, not recommendations for alterations to the test. We have not yet made radiation-stability tests.

Dopants

I feel that 87.7-year ^{238}Pu [$t_{1/2}$ for S.F. (spontaneous fission) 4.8×10^{10} year] will usually be the best dopant if only one is needed. One problem will be the limited solubility of PuO_2 in many glasses. The advantage of ^{238}Pu is the much lower neutron production per alpha decay compared to 18.1-year ^{244}Cm ($t_{1/2}$ S.F. 1.35×10^7 year). Alpha emitters will also give neutrons due to α , n reactions. The size of the batches that are prepared for the test and the size of the samples should be kept as small as possible to minimize radiation dose and contamination potential.

For our ICPP waste forms in which ^{238}Pu is responsible for most of the alpha activity, it will be difficult to add enough ^{238}Pu or ^{244}Cm to give saturation radiation damage in one year. We may need to use 163-day ^{242}Cm . A possible way to produce the ^{242}Cm -loaded samples would be to prepare ^{241}Am -loaded samples and then irradiate the samples in a reactor:

Such a doping method would add the neutron irradiation damage to the alpha damage but would decrease the hazard of working with such a short-lived dopant.

Dose Requirements

For general interest, I would like to see the samples irradiated to saturation, but I do not feel it should be required. Our ICPP waste forms will be far from saturation after all alpha decay has occurred.

Irradiation Temperature

As soon as the maximum expected temperature in the repository is known, e.g., 70°C for our waste in WIPP, a test should also be run at that temperature, and if the temperature range is large, at an intermediate temperature.

Helium Loss

Would a test to measure He release from a sample be worthwhile?

Leach Test

For the purpose of evaluating the effect of radiolysis on leaching, I feel it would be useful to start one sample leaching just after preparation, and continue the leach for at least a year. Since the samples are highly alpha active, the leaching of the dopant could be followed by removing small aliquots at intervals.

I wonder how much of the apparent leaching will be due to alpha recoils and knock-ons that are stopped in the leachant. One factor that needs to be evaluated is the effect of oxidation potential, E_h , on radiolysis. Basalt and granite repositories will most likely have reducing ground waters. The oxidation potential will be very critical for the leaching of UO_2 in spent fuel and ceramics and also for Np leaching. Experiments at Argonne indicate Pu can be radiolytically oxidized to Pu(VI) or perhaps Pu(V).

I also wonder whether in the future we might want to include a test in which alpha-spiked samples are irradiated in a high gamma field. The ATR here has a gamma facility with a decaying field from spent fuel elements. We may want to try leaching samples there (field $>10^6$ R/hr). Canister corrosion should also be tested in a radiation field and in waters with the proper oxidation potential.

David G. Howitt – University of California, Davis

Regarding the Application of Electron Microscopy to the MCC-6 Test
Procedure

While it is particularly difficult to define an evaluation procedure for transmission electron microscopy as it might apply to the candidate materials for the containment of nuclear waste, it is undoubtedly true that a wealth of information could be gained from the application of this technique.

The restrictions imposed by radiation damage to the application of electron microscopy to ceramic materials are significant, and it is true that analytical electron microscopy requires particularly high electron doses. Nevertheless, experiments in our laboratory, with simulated nuclear waste glasses at least, show that these effects can be tolerated within the framework of commercially available instruments.

However, the capability to examine the types of specimens that the MCC-6 test procedure entails is not readily available. While I would not suggest that individual investigators need to develop extensive programs in electron microscopy before adopting the MCC-6 procedure, an analytical electron microscope dedicated to radioactive specimens could prove to be an invaluable asset to them.

Paul W. Levy – Brookhaven National Laboratory

Comments on Working Group Reports

Section 4.1.3

This section points out that the proposed test will employ a dose rate very much higher than will occur in practice. However, it does not point out the possibility that the accelerated test may reduce or even eliminate the damage that would affect the waste-form stability in actual practice. Until eliminated by suitable studies, one should proceed on the basis that the damage effects may increase, on a unit dose basis, as the dose rate decreases. It is suggested that in this section, or in some other suitable place, something like the following be inserted: Dose-rate effects should be studied sufficiently to

establish the dependence on dose rate and, in particular, demonstrate that troublesome effects will not occur at the dose rates encountered in repositories.

Secondly, this section should also indicate--or warn the reader--that saturation effects as measured by one criterion do not necessarily mean that all damage formation has reached saturation. For example, when rock salt is irradiated, the radiation-induced vacancy concentration reaches saturation before the sodium metal colloid formation begins, and the vacancy concentrations remain at saturation as the colloid formation continues to increase.

Section 4.1.5

As far as I can determine, all of the leach solutions included in the proposed tests do not represent the full range of possible realistic solutions. In any repository, water that comes in contact with the waste form will, almost certainly, have been in contact previously with the repository rock, backfill or barrier materials such as bentonite, the canister material (e.g., mild steel), and possibly other substances such as concrete. Consequently, leach tests should be performed in which a combination of all of these substances should be included in the reaction chamber.

Measurements now under way at AERE Harwell, UK, indicate that the leaching of waste-form glass is greatly reduced when granite is included in the reaction chamber. In fact, it appears that the granite supplies materials that cause a particular coat to be formed on the waste-glass surface.

Section 4.2.3

In its present form, this section overemphasizes the possibility that ionization damage will be important. For example, the sentence "Doses in the range...through atomic rearrangements" could be prudently rewritten to read "Doses in the range of 10^{12} R could conceivably produce 0.1 dpa in any silicate-based systems that were unusually susceptible to ionization damage."

Furthermore, it is not entirely certain that ionization damage has been demonstrated in glass. Effects which have been tentatively attributed to ionization damage could equally well be attributed to devitrification reactions, etc.

Section 4.2.5

We commented on dose rate effects under 3.1.3.

Section 4.2.8

It is my conclusion that the first two items in the section are grossly underemphasized; the third is being investigated in a number of laboratories.

Leachability measurements in gamma-ray fields are important from the point of view of establishing the role of radiation, which affects both the waste form and the leaching solution. For example, it should be established whether radiolysis products produced in the leaching solution play an important role in the glass leaching process.

It has been established that there are radiation-related synergistic effects involving strain, defects, thermal gradients, etc. in many systems. I would like to see: 1) a greatly increased effort to discover if any important synergisms occur in the fast-form systems and 2) increased emphasis on synergisms in the report.

Richard Van Konynenburg - Lawrence Livermore Laboratory

1. The draft MCC-6 specifies a required accumulated dose of 3×10^{18} alpha decays per cm^3 (pages 1 and 5). I believe this should be changed either to read that the required accumulated dose should match that expected in 10^6 years for the type of waste under development (defense or commercial) or that separate specifications should be given for defense and commercial wastes. The reasons are as follows:
 - a. There seems to me to be little point in requiring tests of defense waste to doses far in excess of what they will actually receive. A value closer to $1-5 \times 10^{17}$ alpha decays per cm^3 seems more reasonable.
 - b. The information gained from overtesting defense wastes would not be directly transferable to commercial wastes anyway, since the

composition, particularly for crystalline waste forms, will be different. The mixes will be tailored to each waste and will probably include different crystalline phases.

- c. We have agreed that plutonium-238 is the best choice for defense waste radiation-effects simulation because the plutonium isotopes will be the chief sources of alpha-decay damage in defense wastes. However, the doping level of ^{238}Pu required to achieve 3×10^{18} alpha decays per cm^3 in a reasonable time would likely be so high that it would seriously perturb the composition of crystalline waste forms, rendering the simulation questionable at best.
 - d. I do not view it as a disadvantage if measurable property changes do not occur. After all, our job is to develop satisfactory waste forms, not to create as much damage to them as possible. While it may not be very satisfying to radiation-effects experimenters to observe no effects, this is really our ultimate goal in waste-form development.
- 2. The draft specifies that the required dose should be given in a time of one to two years (pages 1 and 4). I believe this may be unnecessarily restrictive. In our own case, because of set-up times and program deadlines, we may have to deliver the dose in a time somewhat less than a year. How about a specification of 6 months to 2 years? Since we are compressing the time scale by a factor of about 10^6 , another factor of two should not make much difference, and it would be a big help to us.
 - 3. On page 5 the draft states that the specific activity of the actinide used as a dopant must be known to within $\pm 2\%$. I think it would be preferable to specify that the specific activity of the waste-form samples should be measured to within $\pm 5\%$. It does only a little good to know the specific activity of the dopant because material may be lost during sample preparation. Ge(Li) gamma ray spectrometry of the actual samples, combined with the techniques mentioned in the first paragraph on page 5 to examine the

distribution, seems like a better approach to finding out what is actually there. A 2% tolerance seems to be tighter than necessary and may be difficult to achieve with the samples themselves.

With these suggested changes, I believe the draft is excellent and will serve its purpose well. I commend you on a very forthright approach to establishing standard tests.

W. R. McDonell - E. I. duPont deNemours & Co.

Radiation-Induced Phase Changes in Nuclear Waste Forms
(A Preliminary Perspective)

A coherent organization of information on radiation-induced phase changes in nuclear waste forms is emerging from recent reviews (Permar and McDonell 1980, Kneff 1975, Lee 1980, Dayal, Swyler, and Soo 1980, Czyscinski, Swyler, and Klamut 1980) summarizing experimental work on radiation effects in inorganic solids (Das and Mitchell 1974, Hobbs 1979, Naguib and Kelley 1975). Phase changes are potentially important aspects of waste-form behavior, since they can affect volume, internal cracking, leachability, helium agglomeration and release, and other characteristics of the waste form. Phase stability is generally necessary for preservation of properties initially invested in the material.

Waste forms, either crystalline or glass, tend to be inorganic mineral solids characterized by mixed covalent and ionic bonding. Such substances are susceptible to radiation-induced changes in atomic arrangements in ways that can either enhance or destroy crystallinity. Thus, initially crystalline materials may undergo loss of crystallinity to form glass-like structures (metamictization) and initially glassy materials may be devitrified to form crystalline phases under irradiation. The metamictization is generally accompanied by large volume increases, while devitrification decreases volume. Other alterations can also occur, including stoichiometry changes, microfracturing, and constituent solubility changes. These reactions are predominantly produced by heavy particle radiations, such as recoil atoms accompanying alpha decay, but evidence is accumulating that beta-gamma radiation can have analogous effects at some level in damage-susceptible materials.

The susceptibility of the various materials to radiation-induced phase changes varies widely, depending on the type of chemical bonding characteristic of the material. Crystalline materials with relatively high degrees of covalent bonding, such as SiO_2 , are most susceptible to metamictization reactions under irradiation, while ionically bonded materials, such as NaCl , are most resistant. Conversely, the covalently bonded glass materials are most readily retained in amorphous condition, while glasses with ionically bonded modifiers are most readily devitrified. Complex crystalline compounds (Na_2SiO_3) and glasses with mixed covalent and ionic bond structure are expected to exhibit intermediate behavior; partially disordered or multi-crystalline-glassy phase structures may be observed.

Bond-type parameters that correlate with phase stability under irradiation include ionicity and crystallization-to-melting temperature ratio. Ionicity is defined by Pauling's equation $I = 1 - \exp(-0.25 (X_A - X_B)^2)$ where X_A and X_B are electronegativities of atoms A and B in the structure. Of 72 non-metallic solids surveyed in a comprehensive review (Naguib and Kelley 1975), 25 substances which amorphize on heavy ion irradiation had ionicities ≤ 0.47 , 18 substances that are stable in crystalline form had ionicities ≥ 0.59 , while 9 substances with varying behavior had intermediate ionicities. The crystallization-melting temperature ratio, T_c/T_m ($^{\circ}\text{K}$) is a measure of the glass-forming susceptibility of a material. Substances with $T_c/T_m > 0.3$ are generally subject to loss of crystallinity under heavy ion irradiation, while substances with $T_c/T_m < 0.3$ retain crystallinity. These criteria provide bases for selection of materials, either crystalline or glass, expected to resist radiation-induced changes in initial structure. Effects of specific fluxing or waste additions to waste glasses—for example, on susceptibility to irradiation-induced structure and property changes—may be indicated by characterization of the above parameters.

Effects of beta-gamma radiation on the phase stability of waste forms is less well characterized, but analogous trends may occur. Crystalline quartz, for example, has been shown to undergo amorphization under intense electron beams (Das and Mitchell 1974, Hobbs 1979, Dayal, Swyler and Soo 1980) and densification of fused silica under high-energy electron and electromagnetic

radiation indicates some devitrification can occur. Additional microstructural and crystallographic characterizations of beta-gamma irradiation effects in crystalline and glass waste forms are needed.

The complexity of most practical waste forms, either crystalline or glass, suggests that long-term structures produced under either alpha or beta-gamma radiation exposures will be more or less intermediate between the original crystalline or glass form. The structural and property changes that occur as a result of irradiation will depend on the rate and magnitude of exposure, as well as the characteristics of the final equilibrium structures. The characteristics of the equilibrium state and the rate at which it is achieved may differ considerably for beta-gamma and alpha radiation exposures. Since high beta-gamma exposures are incurred relatively early (within 1000 years) after repository storage of the waste form, but alpha exposures build up more slowly over a million years or more, complex changes in critical properties such as volume changes could be manifested during the storage life of the waste form.

6.0 ACKNOWLEDGMENTS

This effort was supported by the Department of Energy, Office of Nuclear Waste Management. The accomplishments of the workshop were attributed to the sincere efforts of the participants in giving of their time and knowledge.

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APPENDIX A

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APPENDIX B

WORKSHOP AGENDA

MATERIALS CHARACTERIZATION CENTER
WORKSHOP ON IRRADIATION EFFECTS IN NUCLEAR WASTE FORMS

Battelle Seattle Research Center
Seattle, Washington
July 29-30, 1980

AGENDA

July 28	8:00 p.m.	Informal reception in Apartment B-3	
July 29	8:15 a.m.	Welcome - Announcements	Orv Hill
	8:25	Characterization of Nuclear Waste Forms and the Role of MCC in Waste Management	Ron Nelson
	9:15	The Fundamental Problem of Radiation Damage in Waste Forms	Ray Turcotte
	10:00	Break	
	10:15	Measurement of Radiation-Induced Changes in Waste Forms	Frank Roberts
	11:00	Introduction of Study Group Topics	Ray Turcotte Frank Roberts
		General Discussion	
	12:00	Lunch	
	1:00 p.m.	Separate Study Group Meetings	
	2:30	Break	
	2:45	Continuation of Study Group Meetings	
	4:30	Reconvene Entire Workshop for Summaries Discussion	Ron Nelson
	5:00	Cocktail hour - Apartment B-3	
	6:00	Dinner	
		Evening - Prepare progress reports of study groups.	

July 30	8:15 a.m.	Reconvene Entire Workshop for Schedule Update	Ron Nelson
	8:30	Working Groups Meet to Continue Discussions and to Review and Revise Reports	
	10:00	Break	
	10:15	Reconvene Entire Workshop Presentation of Group Reports and Discussion	Ron Nelson, Moderator
	12:00	Lunch	
	1:00 p.m.	Discussion of Prepared MCC Procedure and Recommend Changes	Ron Nelson, Moderator
	2:30	Break	
	2:45	General Discussion	Ron Nelson, Moderator
	4:30	Adjourn	

APPENDIX C

STANDARD TEST FOR THE EFFECTS OF ALPHA DECAY IN NUCLEAR WASTE SOLIDS (MCC-6)

(Draft)

Standard Test for Alpha Recoil Effects
in Nuclear Waste Solids (MCC-6) (a)

The following test procedure (Standard Test for Alpha Recoil Effects in Nuclear Waste Solids MCC-6) pertains to the waste forms. It provides a standardized method for irradiating waste forms to simulate the effects of the alpha decay of waste actinides. After approval by MRB, the method will be used to obtain data on property changes induced by internal alpha decay. The properties evaluated included density, leachability, microstructure, crystallinity, stored energy, and mechanical strength.

(a) This was the original title of MCC-6. It has since been changed to "Standard Test for the Effects of Alpha Decay in Nuclear Waste Solids."

STANDARD TEST FOR ALPHA-RECOIL EFFECTS IN NUCLEAR WASTE SOLIDS (MCC-6)

Materials Characterization Center

Pacific Northwest Laboratory

Richland, Washington

March 20, 1980

1.0 SCOPE

This test is to be used to evaluate changes in the properties of solid radioactive wastes from the decay of alpha-emitting nuclides contained in the waste. The properties, which may undergo changes with irradiation, evaluated include:

- density
- leachability
- microstructure
- crystal structure
- stored energy buildup
- mechanical strength.

The conditions for self-irradiation of simulated waste specimens, the property measurements, and the dose levels for the property measurements are specified in the test. The details of the property measurement procedures are not included but can be found in other Material Characterization Center (MCC) standard tests or support methods.

This test uses the actinide doping method in which ^{244}Cm , a relatively short-lived alpha-emitting actinide ($t_{1/2} = 18.1 \text{ y}$), is incorporated in the simulated waste forms. The presence of the actinide dopant must not significantly alter the chemical properties of the solid waste but should result in an alpha fluence of $\sim 6 \times 10^{21}$ alpha decays/liter in a reasonably short time ($\sim 1 \text{ y}$). (It has been observed experimentally, and can be shown theoretically by simple physical models of the damage process, that saturation damage is exceeded at this dose.)

2.0 LIMITATIONS

This test is limited to monolithic inorganic waste forms, although it could be adapted to powdered material, multiple barrier forms (e.g., glass beads in a metal matrix), or organic-based systems. However, this test does not describe the necessary variations for examining these materials nor does it address a subtest for possible radiolytic losses of gaseous components. (This test will be developed by MCC if required.) For example, potential radiolytic decomposition of water and canister pressurization from hydrated waste forms, such as concretes, would be a necessary test for that material but not for high-temperature fired ceramics.

This test provides a framework for the collection of basic information. It does not exclude measurement of other radiation-induced changes or changes arising under conditions different from those specified in the test. It is known, for example, that the magnitudes of radiation-induced changes are temperature dependent. Also, microcracking may occur in some materials if the specimens are self-irradiated in water but not in air. More in-depth study of irradiation effects may require that observations be made under a range of environmental conditions.

Other questions concerning radiation stability of waste forms include the role of ionization damage by high energy gamma rays and beta particles and changes caused by transmutation of some isotopes (e.g., $^{137}\text{Cs} \rightarrow \text{Ba}$). At the time of this writing (1980), research in these areas is in progress. Depending on the results of these studies, formal test procedures may be established if needed.

Finally, note that for this test the properties are measured as a function of the radiation dose and the quantities of interest are the changes of the properties brought about by irradiation rather than absolute quantities.

3.0 BACKGROUND AND RELATED TEST PROCEDURES

3.1 Other Procedures

The test uses other standard tests being established by MCC. These include:

- Static Leach Test (MCC-1)
- High Flow Rate Leach Test (MCC-2)
- Compressive Strength Test (MCC-9)
- X-Ray Diffraction Analysis (MCC-SM-5)
- Scanning Electron Microscopy (MCC-SM-6)
- Optical Microscopy (MCC-SM-7)
- Density Measurement (MCC-SM-8)
- Stored Energy by Differential Scanning Calorimetry (MCC-SM-9).

A closely-related standard test method has been proposed by ISO, an international working group, entitled Standard Method for Testing the Radiation Stability of Vitrified Radioactive Waste (ISO/TC 85/SC 5W/G 5N23). The ISO procedure, reproduced in Appendix A, is not fundamentally different than proposed in the present case, although some of the details differ.

3.2 Theory

Nuclear waste will be converted to stable solids prior to storage or ultimate disposal. Borosilicate glasses and partially or entirely crystalline inorganic oxide compacts are the favored forms for high-level wastes. Consideration is also being given to other forms, such as concretes and cermets. The solidified waste will be subjected to radiation doses of every kind that may measurably alter the physical properties and potentially affects the durability of the solid form. Thus, tests are required to provide assurance that irradiation effects do not significantly reduce the long-term stability of the waste form.

The principal sources of radiation in the wastes are alpha decay of the actinide elements and beta decay of the fission products. Alpha decay produces two projectiles: alpha particles with energies of ~4-6 MeV and recoil nuclei with energies of ~90 KeV. The alpha particles dissipate their energy mostly in transient ionization processes, each producing only a few hundred atomic displacements per decay event. The recoil nuclei, on the other hand, interact by elastic collision processes producing several thousand atomic displacements per decay event. Beta decay produces high energy electrons and gamma rays

which interact with the solid by ionization processes and produce very few atomic displacements. Alpha decay is the major cause of radiation-induced changes in the waste solids.

The irradiation technique used for this proposed test is curium doping, i.e., incorporating $^{244}\text{Cm}^{(a)}$ in the simulated waste form and allowing it to self-irradiate to a predetermined dose. ^{244}Cm is used because it is readily available and its half-life permits alpha doses of 6×10^{21} alpha decays/liter in about one year while not exceeding 3 wt% curium oxide in the test specimen. Other isotopes that have been considered are ^{242}Cm , ^{238}Pu , and ^{241}Am , but these are not used for this test, primarily because of their half-lives (^{242}Cm because its half-life is inconveniently short, and ^{238}Pu and ^{241}Am because their relatively long half-lives would require unacceptably long decay times to arrive at an acceptable dose). In some cases, however, use of these other isotopes may be required. An example is the use of ^{238}Pu as a +4 valence dopant in crystal structures not accepting CM^{+3} (the stable valence at high temperatures). Other possible irradiation techniques include ion bombardment (ion implantation) and neutron irradiation. Ion bombardment produces only near-surface effects, which greatly increases the difficulty in interpreting the observed changes. Neutron irradiation, either thermal neutron irradiation of the waste form containing ^{235}U producing fission fragments or fast neutron irradiation producing knock-ons, is not used for this test because the alpha-doping technique more nearly resembles the irradiation that will be experienced in actual waste forms.

The alpha irradiation effects tend to saturate at alpha fluences of less than 6×10^{21} alpha decays/liter for the case of borosilicate glasses (Mendel et al. 1977; Ross et al. 1978; Roberts, Jenks and Bopp 1976; Scheffler, Riege and Hild 1976). Saturation damage also appears to occur at nearly the same dose in compacted supercalcine (Rusin, Gray and Wald 1979).

Because the range of the recoil nuclei is less than 100 Å, recoil nuclei damage in polyphase systems will occur primarily in the phases containing the

(a) ^{244}Cm has a half-life of 18.1 years and decays by alpha emission producing alpha particles with energies of 5.81 MeV (77%) and 5.77 MeV (23%).

actinide elements. Alpha particles have a much longer range and can cause damage in adjacent crystals or matrix materials. The evaluation of the test results must take this fact into consideration. In actual cases here the actinides partition into different phases, e.g., plutonium in one phase and americium in another, simulation with curium may not be entirely valid. For this reason, it is essential to know in which phases each of the actinide elements reside.

The information on the dose rate dependence of alpha radiation-induced changes is very limited, but tests indicate the dependence is not large (Mendel et al. 1977).

3.3 Criteria

No criteria are established for defining the allowable level of radiation-induced changes in solidified wastes.

4.0 SPECIFICATIONS

4.1 General

The composition of the curium-doped simulated waste should be as identical as possible to the waste form being tested. The added curium should replace an equal amount of trivalent rare earth on a molar basis, if possible. In some cases, it may be acceptable to simply add the curium without compensating the rare earth content. Care should be used to assure that the ^{244}Cm is uniformly distributed in the specimen. Alpha autoradiography, optical microscopy, x-ray diffraction analysis, and electron microscopy can be used to verify that the curium is uniformly distributed in the specimen or in the crystalline phase expected.

The specific alpha activity of the specimens should be $2.3 \pm 0.4 \times 10^{14}$ alpha decays/s/liter. This will give an integrated dose of $>6 \times 10^{21}$ alpha decays/liter in one year. The specific activity of the curium used as a dopant must be known within $\pm 2\%$, either from radiochemical analysis by the user or from a certified analysis by the supplier.

The preparation of the specimens should duplicate the processing conditions of the waste form being tested. Adherence to processing temperatures

and times, annealing temperatures and times, cooling rates, curing conditions, etc., encountered in the process are particularly important.

The specimens should be stored during the irradiation period at a controlled temperature of $23^{\circ}\text{C} \pm 3^{\circ}\text{C}$ ($296\text{K} \pm 3\text{K}$). A temperature monitor should be installed in storage areas, and the specimens should be protected against mechanical damage and humidity extremes during storage. During irradiation, sections cut from the original preparation should be butted against one another to provide homogeneous alpha particle and recoil-nuclei fluxes. Without this provision, the doses at the surfaces would be only half that in the interior of the sample.

B. Specimen Fabrication

Glass, ceramic, and concrete specimens are prepared by mixing the component oxides and adding the required curium as a nitric acid solution. The details of curium addition may vary for technical or practical reasons for the different waste forms.

The specimen can be cast in any convenient form that permits sections to be removed for the individual measurements, such as right circular cylinders or rectangular bars. The numbers and sizes of samples needed for the measurements are shown in Figure 1, using a 15-mm diameter right circular cylinder as an example. (For some simulated wastes, two or more cylinders may be more practical).

The stored energy measurement requires a 40–60 mesh powdered sample, obtained by crushing a section and sieving.

5.0 TEST PROCEDURES

5.1 Density

The density of glasses, glass ceramics, and other nonporous forms are to be measured in accordance with Support Method MCC-SM-8.

The density should be measured within 48 h after preparation of the specimen and again at one month intervals until an integrated dose of 6×10^{21} alpha decays/liter has been attained.

FIGURE 1. Diagram of Samples Required for Tests

5.2 Static Leach Test

The leachability should be measured in accordance with Static Leach Test MCC-1. In this test, a monolithic sample of the waste form is immersed in a stationary leach solution for a predetermined length of time at a constant temperature and the leach rate estimated from the amount of specific elements released to the leachate.

A series of tests are to be made beginning within 48 h after preparing the specimen and again after a dose of 6×10^{21} alpha decays/liter has been attained. Each series will consist of three tests differing only in duration using a separate sample for each test. The duration of the tests should be 1, 3, and 7 days. The temperature of the test should be 363 K. Leach rate data shall be reported for cesium, strontium, and other elements based on chemical analysis of the leachate and for curium based on total alpha counting of the leachate.

5.3 High Flow Rate Leach Test

The leachability is also examined by the High Flow Rate Leach Test MCC-2. In this test, a monolithic sample of the waste form is immersed in distilled water in a continuous-flow soxhlet-type apparatus for a fixed period and at a constant temperature. The leach rate is determined by elemental analysis of the leachate and from the weight loss of the preweighed sample after testing. Two series of tests are to be made. The first is to be started within 48 h after preparing the specimen, and the second when a dose of 6×10^{21} alpha decays/liter has been attained. Each series shall consist of three tests, differing only in duration, using a separate sample for each test. The duration of the three tests should be 1, 3, and 7 days.^(a) The temperature of the test should be 363K.

5.4 X-Ray Diffraction

A series of x-ray diffraction traces should be prepared beginning within 48 h after preparing the specimens and subsequently at about 30-day intervals

(a) These durations are tentative, awaiting the completion of High Flow Rate Leach Test MCC-2.

until a dose of 6×10^{21} alpha decays/liter has been attained. Changes induced by the radiation can generally be observed through shifts and decreased intensities of the diffracted x-ray peaks that are characteristic of the crystalline phases present in the simulated waste. The technique for preparing and analyzing the traces, described in X-Ray Diffraction Analysis Support Method MCC-SM-5, should be used.

5.5 Microscopy

A series of optical micrographs, at a magnification of 100–400X, and scanning electron micrographs, at a magnification of 200–20,000X, should be taken of a fixed area of the specimen. The series should start within 48 h after preparing the specimen and again at approximately 30-day intervals until a dose of 6×10^{21} alpha decays/liter has been attained. Scanning Electron Microscopy Support Method MCC-SM-6 and Optical Microscopy Support Method MCC-SM-7 should be followed.

5.6 Mechanical Strength Test (Optional)

Determine the mechanical strength of the specimen using test MCC-9 within 48 h after preparing the specimen and again after a dose of 6×10^{21} alpha decays/liter has been attained. (At the time of this writing, a diametral compression test MCC-9, based on ASTM Standard 694-64T, is being developed.)

5.7 Stored Energy Measurement (Optional)

Stored energy release is determined by differential scanning calorimetry using Support Method MCC-SM 9. The initial measurement should be performed within one week after preparation of the specimen and at one-month intervals thereafter until a dose of 6×10^{21} alpha decays/liter has been attained.

The stored energy release is determined by heating a weighed sample in the differential scanning calorimeter at a constant rate from 50°C to 600°C and recording the differential temperature between the sample and a reference material.^(a) The calorimeter is then cooled and the temperature range again

(a) The reference material may be a sample of the simulated waste form without curium or a thermally-inert material such as Al_2O_3 .

scanned to provide a baseline. The area between the first scan and the baseline, which is proportional to the amount of energy release, is used to calculate the stored energy released.

6.0 REPORTS

A standard format for reporting the results similar to that described here should be used, including a detailed description of the simulated waste form:

- the type and composition of the solidified material
- the details of the preparation, which include formation temperature; pressure and times; cooling rates, if applicable; annealing conditions; and any special treatments
- the specific activity of the ^{244}Cm in the simulated waste and the date corresponding to that specific activity
- alpha radiography and other data defining the adequacy of the ^{244}Cm doping relative to the expected mode of incorporation.

6.1 Results on the Individual Tests:

- density
- leachability
- x-ray diffraction
- microstructure
- mechanical strength
- stored energy.

An outline of the report is given in the following sections. An example of a report with actual data is given in Appendix C.

6.1.1 Section 1: Description of Simulated Waste For Preparation

- 1.1 Waste Form Identification Number
- 1.2 Type of Material
- 1.3 Composition of Waste Form
- 1.4 Composition of Waste

1.5 Alpha Activity of the Prepared Specimen

- calculated specific activity
- measured specific activity

1.6 Description of Specimen Preparation

This part of the report should include a detailed description of the specimen preparation, including pertinent times, temperatures, pressures, and rates of change of temperatures and pressures, if applicable.

6.1.2 Section 2: Density Measurements

2.1 Description of Samples

- dimensions
- weight

2.2 Method Used for Density Determination

- buoyancy or flotation medium

2.3 Results of Measurements

The results should be presented in a tabular form giving:

- date of measurement
- accumulated alpha dose
- density in kg/m^3
- temperature of test.

The results should also be presented graphically, showing a plot of density change versus accumulated alpha dose. The density change is calculated from:

$$\text{change} = \frac{100 (\rho_i - \rho_0)}{\rho_0}$$

where

ρ_0 = measured density at zero dose, and

ρ_i = measured density at the time of measurement.

6.1.3 Section 3: Results of Leach Tests

The results of this static leach test and the high flow rate leach test should be presented in accordance with the format recommended in MCC-1 and MCC-2. A summary of the test results in a tabular form showing the leach

rates in $\text{kg/m}^2/\text{day}$, the duration of the test, and the accumulated alpha dose at the start of each test is recommended.

6.1.4 Section 4: X-Ray Diffraction Results

The relative intensities and the angles (2θ) of all significant reflections are to be tabulated. The crystal phases are to be identified, if possible. The intensity ratio (I/I_0) of one of the major reflections of each phase (identified or unidentified) should be plotted as a function of the accumulated alpha dose.^(a)

6.1.5 Section 5: Microstructure

The section of the report should present the alpha autoradiography and optical and scanning electron microscopy results. Replicas of the micrographs taken in the same area of the specimen at low doses and at saturation level doses should be reproduced here. A summary of the observed changes resulting from the irradiation (microcracking, void formation, evidence of segregation of curium, etc.) should be included.

6.1.6 Section 6: Mechanical Strength Test Results

The mechanical strength test is undergoing development, and the reporting requirements have not been defined.

6.1.7 Section 7: Stored-Energy Results

The results of the stored-energy tests should be presented both in tabular format and graphically, showing the stored energy release in J/kg as a function of accumulated alpha dose.

6.1.8 Section 8: Discussion

8.1 Discussion of Results

A brief summary of the significance of the results should be given. Reference can be made to reports or other literature references containing further discussion of this data, particularly correlations to other data.

(a) The intensity ratio is the relative intensity of the line at the time of the measurement/the relative intensity at zero dose.

8.2 Observations, problems, or other comments concerning the tests can be presented here.

8.3 A listing of the responsible persons and the laboratory reporting the data should be given here.

6.1.9 Section 9: Error Evaluation

9.1 Summary

This section will summarize error estimates for the particular data being reported. Ordinarily, these will simply be reported as relative standard deviations in percent, with discussion when needed.

9.2 Data Availability

A more general discussion of errors and a current summary of error-related data is available from MCC.

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