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Evaluation of the Three Mile Island Unit 2 Reactor Building Decontamination Process

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ABSTRACT

Decontamination activities from the cleanup of the Three Mile Island Unit 2 Reactor Building are generating a variety of waste streams. Solid wastes being disposed of in commercial shallow land burial include trash and rubbish, ion-exchange resins (Epicor-II) and strippable coatings.

The radwaste streams arising from cleanup activities currently under way are characterized and classified under the waste classification scheme of 10 CFR Part 61. It appears that much of the Epicor-II ion-exchange resin being disposed of in commerical land burial will be Class B and require stabilization if current radionuclide loading practices continue to be followed. Some of the trash and rubbish from the cleanup of the reactor building so far would be Class B.

Strippable coatings being used at TMI-2 were tested for leachability of radionuclides and chelating agents, thermal stability, radiation stability, stability under immersion and biodegradability. Actual coating samples from reactor building decontamination testing were evaluated for radionuclide leaching and biodegradation. Results indicated that both radionuclide contamination and chelating agents leach from strippable coating waste. It swells and partially dissolves upon immersion in water and organic liquids, biodegrades readily in soil and, upon irradiation, generates gas, principally hydrogen. The coating is thermally stable up to at least 100°C and is not an unacceptable flame and smoke hazard. The strippable coating samples from the TMI-2 reactor building decontamination testing contained significant strontium-90 and cesium-137 contamination and would be Class B under 10 CFR Part 61.

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EVALUATION OF THE THREE MILE ISLAND UNIT 2 REACTOR BUILDING DECONTAMINATION PROCESS

1. INTRODUCTION

It has been more than 3-1/2 years since the March 28, 1979 accident at the Three Mile Island Unit 2 Nuclear Generating Plant which left the reactor core damaged and the reactor building contaminated with fission products. During this time plans[1-7] have been made and effort[8-12] expended to cleanup and recover TMI-2. These efforts are directed toward decontaminating the reactor building, removing the nuclear fuel from the damaged core and decontaminating the reactor coolant system and connected systems. Decontamination of the reactor building will initially focus on those areas needed for defueling. These areas, as shown in Figure 1.1, Include the 305-ft. level, the 347-ft. level (the operating floor) and the polar crane. The reactor coolant system water must also be cleaned up to reduce the concentration of dissolved fission products prior to defueling.

Major milestones[13] in the cleanup and recovery effort have included the following events:

(1) Venting to atmosphere of the 44,000 Curies of the fission product gas krypton-85 which was released in the reactor building during the accident. This was done between June 28 and July 11, 1980.

(2) Removal of dissolved fission product activity from the 565,224-gal of tritiated accident water In the Auxiliary and Fuel-Handling Building (AFHB). The fission product activity was removed by ion exchange (Epicor II*) between November 1979 and December 1980. The highly loaded ion-exchange resins are being transferred to the Department of Energy (DOE) for disposal. The tritiated accident water (<1 $\mu Ci/cm^3$) is being stored for reuse at TMI.

(3) Removal of the dissolved fission product activity from more than 600,000 gal of tritiated accident water which flooded the basement (282-ft elevation) of the reactor building. The fission product activity was sorbed onto zeolite demineralizer beds in the Submerged Demineralizer System (SDS) between September 23, 1981 and March 5, 1982. The tritiated water effluent from the SDS was polished (i.e., the small amount of fission product activity not sorbed onto the zeolites was removed) using Epicor-II ion-exchange resin. The highly loaded zeolites are being transferred to DOE for disposal. Activity levels in the Epicor-II polishing resins were within the Hanford, Washington, commercial land burial site limit (<1 $\mu Ci/g$)

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*Proprietary ion-exchange system of Epicor, Incorporated.

for disposal in the dewatered form and without solidification. The tritiated accident water (<1 $\mu Ci/cm^3$) is being stored for reuse at TMI.

(4) The Gross Decontamination Experiment was performed between October 29, 1981 and March 26, 1982. This effort was directed toward reducing radiation levels on the 305-ft. elevation and above and toward determining the relative decontamination effectiveness of the several techniques selected for testing.

(5) Gross Decontamination of the reactor building including the 305-ft. elevation and above started September 17, 1982. The effort was designed to reduce smearable levels of contamination to the point that workers will be able to remove much of the bulky protective clothing and the respirators.

(6) Removal of fission product activity from the 88,000 gallons of water in the reactor coolant system (RCS) using the SDS has been attempted but has not been completely successful. Strontium-90 from the fuel debris apparently is dissolving into the cleaned water and restoring the strontium-90 activity to near its initial level. This phenomenon may influence the RCS cleanup plans which were not finalized as of this writing.

Major events[12-16] in the cleanup and recovery yet to occur include:

(7) Defueling of the reactor. Plans for removing the fuel from the reactor await detailed inspection of the core. This inspection began with the "Quick Looks"[13,14] into the reactor vessel with a remote TV camera and will continue when the reactor vessel head is removed, which is scheduled for sometime in mid 1983. It is hoped that fuel removal from the core can be completed by late 1985.[15]

(8) Decontamination of the reactor coolant system and connected systems which have been contaminated by fuel debris. The decontamination techniques and procedures to be used for this cleanup will not be decided upon until studies being performed by the Electric Power Research Institute (EPRI) recommend appropriate techniques.[12]

(9) Cleanup of the refueling canal during and after reactor defueling. Transfer of the core debris from the reactor to a packaging facility may result in substantial soluble and particulate contamination of the refueling canal.

(10) Hands-on decontamination of the 305-ft. elevation and above. This is intended to return the working environment in this part of the reactor building to near normal (i.e., pre-accident) conditions. This effort will start sometime after Gross Decontamination, Item 5.

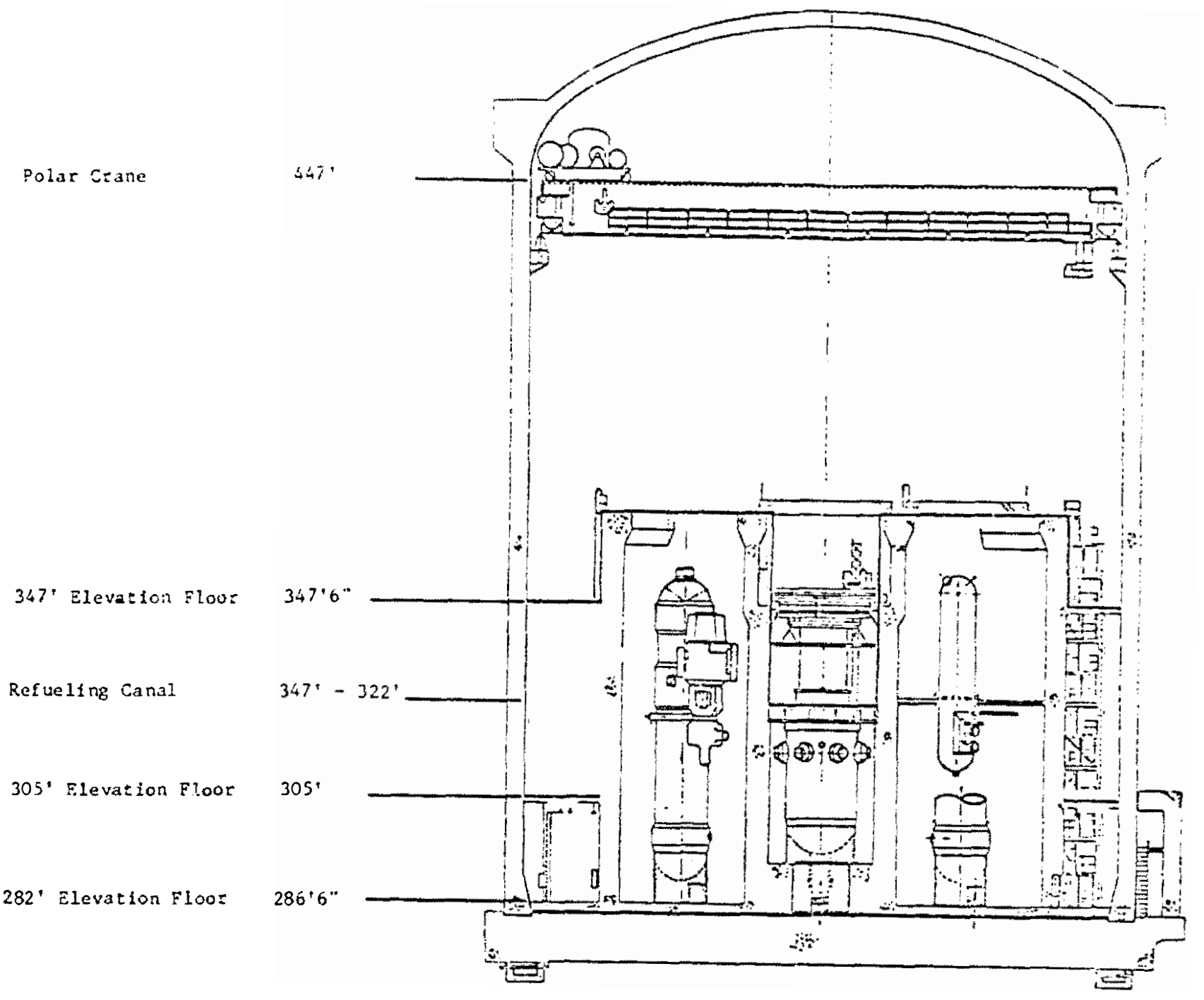

Figure 1.1 Vertical cross-sectional schematic of the TMI-2 reactor building showing the floor levels in comparison to the major components of the nuclear steam supply system.

(11) Decontamination of the 282-ft. evaluation. The 600,000 gallons of highly contaminated water, Item 3, filled this part of the reactor to a depth of about 8 ft. This elevation has much higher radiation fields than the upper portions of the reactor building.

(12) Disposal of the more than 1-million gallons of tritiated accident water, Items 2 and 3. TMI is under specific NRC order[16] not to release any of the accident water. In addition to the considerations of 10 CFR Part 20 with regard to this water, the NRC and the State of Maryland are conducting studies on potential socioeconomic impacts of various alternatives for disposal of the accident water.

Flushing with both low and high pressure water spray has been the principal decontamination method for removal of as much loose debris and smearable and soluble activity as possible. The tritiated accident water (<1 μCi H-3 per mL) is being used for the water flushing decontamination activity. It is routed through floor drains from the upper levels in the reactor building to the basement (282-ft. elevation) from where it is then sent to the SDS for cleanup and return to tank storage for reuse. The Gross Decontamination Experiment (GDE) also tested, in small scale, detergent (Turco 4324 NP-10%) and phosphoric acid (Turco 4512-A 10% Normal) solutions in conjunction with a floor scrubber.[10] These tests were conducted on two 128 ft^2 areas, one for each solution, on the floor of the 347-ft elevation. The liquid wastes, approximately 26 gallons for each solution, were wet vacummed into 55-gal drums and then transferred to storage. The storage container is an 8000-gal tank at TMI in which all liquid wastes are combined for storage. Strippable coatings were also tested during the GDE. These coatings are applied as liquids. After drying to a rubbery film, the coatings are then peeled off for disposal. This results in only solid waste which is then drummed for disposal. Problems associated with the processing of radioactive waste produced by decontamination methods other than water flushing have provided a major inhibition to their use at TMI-2.

The cleanup activities listed above are generating a large quantity of radioactive waste. Some of this waste is unique - such as the damaged core. Some of the waste contains exceptionally large amounts of activity and is not suitable for disposal in commercial land burial. Disposal of these wastes is provided for in the Memorandum of Understanding[17] (MOU) between the NRC and the Department of Energy (DOE). The MOU states that DOE may, on an R and D or reimbursable basis, take posession of the nuclear fuel and those radioactive wastes from TMI-2 which are not suitable for disposal in commercial land burial. However, a large amount of waste will go to commercial land burial.[18,19]

The radionuclides of concern in the cleanup wastes are long-lived fission products - principally Cs-137 and Sr-90, rather than neutron activation products (e.g., Co-60) associated with reactor "crud". Fuel debris from the reactor core will also contain transuranic (TRU) contamination. In wastes from

cleanup of the RCS and possibly the 282-ft elevation, the TRU contamination may be of most concern. The strippable coatings being used in the cleanup contain chelating agents.

Since the cleanup of TMI-2 will continue well into the latter half of the decade, much of the waste generated will fall under the regulation of 10 CFR Part 61, "Licensing Requirements for Land Disposal of Radioactive Waste." The activities of alpha-emitting TRU and the long-lived fission products Cs-137 and Sr-90 are used in 10 CFR Part 61 to classify radwaste for disposal purposes. The waste classification section of the regulation, 10 CFR Part 61.55, is attached as Appendix A. The NRC has contracted Brookhaven National Laboratory (BNL) under FIN A-3162, Task 7, "Evaluation of the Three Mile Island Unit 2 Reactor Building Decontamination Process," for technical assistance in characterizing these decontamination wastes. Evaluation of the strippable coatings being used in the decontamination effort is specifically included in this task. Evaluation of these wastes for compliance with the provisions of 10 CFR Part 61 is a part of this characterization.

The decontamination wastes being generated in the cleanup of the TMI-2 reactor building, as of this writing, are described and their performance under 10 CFR Part 61 is evaluated. Strippable coating properties and uses are presented along with the results of testing for radionuclide and chelating agent leachability, thermal stability, biodegradability, and radiation stability. The activity that strippable coating waste from TMI-2 may contain are estimated from samples of coating from the Gross Decontamination Experiment.

2. WASTE MANAGEMENT FOR TMI-2 REACTOR BUILDING CLEANUP

2.1 Reactor Building Decontamination Planning

Reactor building decontamination planning is based on the alternatives outlined in the Final Programmatic Environmental Impact Statement (PEIS).[3,16] The specific technologies chosen have been based on effectiveness, waste management, and other considerations. The approximately chronological listing in Section 1 of major events in the TMI-2 cleanup also provides a basis for categorizing the various streams of radioactive waste being generated in the reactor building cleanup. Items 1-4 have been completed and item 5 is scheduled for completion no later than the end of January, 1983.*

2.2 Decontamination Wastes

Waste stream characterization for various decontamination and waste processing alternatives has been estimated in some detail in the PEIS.[3] The wastes generated from the activities of Items 2 and 3, Section 1, consisted of SDS zeolites, Epicor-II ion-exchange resins, and general trash. The SDS zeolite waste and the Epicor-II waste were highly radioactive and DOE has agreed to take possession of these under the MOU. An estimate of the Classification that these wastes would have had under 10 CFR Part 61 (draft version) is available.[20] DOE is using some of the Epicor-II wastes, Item 2, and the zeolite wastes, Item 3, in various research programs related to radioactive waste and the rest is being disposed of as "special waste" on a one-time disposal basis.[20,21] The entire reactor core from the defueling operation, Item 7, will be taken by DOE.[17] Wastes contaminated by TRU concentrations in excess of the levels authorized for commercial burial may also be accepted by DOE.[17] Some wastes from (Section 1) Items 8, 9, and 11, may be transferred to DOE under the MOU provision cited in the preceeding sentence. The TMI-2 licensee, General Public Utilities, is disposing of the low level waste arising from the cleanup operations at the Hanford, Washington, commercial radioacitve waste land burial site.[18] The waste being shipped to the Hanford commercial burial site is similar to wastes being routinely generated at other nuclear power facilities.

Characterization of TMI-2 radwaste is accomplished using gamma measurements from which radionuclide inventories are estimated.[22,23] Two categories of radwaste have been defined in the reactor building cleanup so far: (1) Normal Unit-2 radwaste and (2) Makeup and purification (MUP) system radwaste. Normal Unit-2 radwaste is being generated by cleanup activities on the 305-ft elevation and above. The activity in normal Unit 2 radwaste has a specified fractional composition of 0.471 Cs-137 and 0.0234 Sr-90. The remainder consists of the Ba-137m and Y-90 daughters of Cs-137 and Sr-90 plus some Cs-134. The MUP system, which is located on the 281-ft level of the AFHB, consists of ion exchange resin bed demineralizers and heat exchangers.

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*Personal communication between D. Dougherty (BNL) and D. Geifer, Bechtel National Corporation, at TMI, December 14, 1982.

This system is connected directly to the reactor coolant system. It is scheduled to be decontaminated as part of the RCS cleanup. As of this writing no MUP radwaste has been generated. MUP radwaste has been specified to have the following isotopic fractional distribution: 0.176 Cs-137, 0.234 Sr-90, and plus 2.74×10^{-4} TRU. The TRU content limits the fission product activity in a container to avoid exceeding the 10 nCi/g limit imposed by the Hanford burial site. The container activity limits necessary to avoid exceeding the TRU limit of 10 nCi/g are listed in Reference 22. The activity inventory in a waste container is estimated for both categories of radwaste using the following equation,

Container Activity (mCi) = (C.F.) (Average Measured Dose Rate [mrem/h])

in which C.F. is a conversion factor which depends on the container and whether the activity in the waste is normal or MUP. Three types of containers, each with its own C.F., are used at TMI-2 for solid waste disposal. These are compacted and non-compacted 55-gal drums and 98 ft^3 LSA boxes.

Volume reduction capability for solid radwaste at TMI-2 consists of a 30,000 psi compactor for 55-gal drums as of the writing of this report. A 5-gph evaporator is available for liquid wastes but, as of this writing, has never been used.(12) This small evaporator was to be tested on chemical decontamination liquids but there were no plans to place the evaporator in service in the foreseeable future.* There is no solidification facility at TMI-2. A facility for solidifying waste in cement is available at TMI-1, but it is not allowed to be used for TMI-2 wastes. A 30-gph evaporator was considered for installation in 1979, but has not been purchased. Incineration was also a considered option for volume reduction, but an incinerator has not been purchased.(12,24)

Solid waste is packaged in 55-gal drums and 98-cu. ft. L.S.A. boxes for storage, shipment, and disposal. Compactible wastes can be volume reduced by compaction into 55-gal drums. As a general rule, compaction of waste is performed to the maximum extent possible. However, waste that would exceed the LSA classification under compaction and waste whose activity is such that the compactor operators would receive too large a dose are not compacted. That waste suitable for land burial is shipped to Hanford, Washington, commerical burial site for disposal(18) in placarded, exclusive use trucks. Shipments are now averaging about two per month** although they were more frequent when radwaste shipments from TMI-2 started in November 1979. A fully loaded trailer holds 20 LSA boxes (1960 ft^3) or approximately 150, 55-gal drums (1100 ft^3) or a mixture of the two. TMI shipment number Rs-82-028-II, April 29, 1982, may be representative of typical radwaste shipments from

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*Personal communication between D. Dougherty (BNL) and P. Carmel (Bechtel) at TMI, December 17, 1982.

**Personal communication between D. Dougherty (BNL) and T. Moslak (NRC) at TMI, September 8, 1982.

TMI-2.* This shipment contained 6 LSA boxes and 68 drums. Five of these drums labelled "compacted trash" would be Class B under 10 CFR Part 61 for the Sr-90 content or the Sr-90 plus Cs-137 content.

Gross decontamination of the 347-ft elevation was performed with water spray and a mechanical floor scrubber using a non-abrasive pad and water. The contaminated water is cleaned up using the SDS and Epicor-II systems. The activity loading of the zeolites in the SDS from this cleanup is not known. The activity loading of the Epicor-II resins is determined by monitoring activity levels of the influent to and effluent from the Epicor-II system. The loading is limited to <1 μCi/g. The Hanford license** states that resins having activities less than 1 μCi/cm^3 with half-lives greater than 5 years can be disposed of in the dewatered condition. Since dewatered Epicor-II resin has an apparent density of approximately 0.7 g/cm^3, the 1 μCi/g loading at TMI is below that allowed by Hanford (i.e., 1 μCi/cm^3 ÷ 0.7 g/cm^3 = 1.4 μCi/g). Hence, spent resins have been dewatered and shipped to Hanford for disposal.

The Epicor-II system for polishing the SDS effluent has three sections: the pre-filter stage (PF), the first stage (K filter) and the second stage (2K filter). Through 1982 42-PFs, 8-K and 7-2K filters had been used.*** Of these totals 23-PFs, 5-Ks and 6-2Ks were stored at TMI awaiting disposal. The PFs were loaded to approximately 1 μCi/g while the K and 2K filters were loaded to lesser activities. PFs are shipped in shielded casks for disposal, 2K filters do not require shielding for shipment and K filters are shielded or not on a case by case basis. Disposal costs have led to studies of the cost effectiveness of continuing present procedures. An alternative currently being considered is to increase the activity loading on the Epicor-II resins followed by solidification prior to disposal.****

By way of illustration, the 10 CFR 61 classifications that Epicor-II radwaste would have if loaded to 1 μCi/g with activity of Normal Unit-2 and MUP radioisotopic fractional compositions are calculated. The fractional activities of Cs-137 and Sr-90 in Normal Unit-2 Radwaste are 0.471 and 0.0234, respectively The cumulative fraction under 10 CFR Part 61 from Table 2 of Appendix A, using the gravimetric loading limit of 1 μCi/g and the apparent

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*Copies of the shipping papers for this shipment were forwarded to BNL from a request for information on a typical solid waste shipment for commercial land burial.

**The State of Washington, Radioactive Materials License WN-I019-2, Amendment No. 15 in accordance with renewal application dated October 28, 1981, expiration date November 30, 1985.

***Personal communication between D. Dougherty (BNL) and T. Moslak (NRC) at TMI, December 17, 1982.

****Personal communication between D. Dougherty (BNL) and R. Hahn (GPU) at TMI, April 27, 1983.

density of approximately 0.7 g/cm^3 for dewatered Epicor-II resin is

$$\frac{0.471}{1.0\ \mu Ci/cm^3} \times 1\ \mu Ci/g \times 0.7\ g/cm^3 + \frac{0.0234}{0.04\ \mu Ci/cm^3} \times 1\ \mu Ci/g \times 0.7\ g/cm^3 = 0.74$$

Performing the same calculation using the MUP radwaste fractional contents for Sr-90 and Cs-137 leads to a value of 4.2. These numbers mean that if the radionuclide fractional compositions in normal and MUP radwaste are carried through in the SDS effluent to the Epicor-II system, then a 1 μCi/g loading would be Class A for the normal Unit-2 radwaste radionuclide composition and Class B for the MUP fractional distribution. In actuality, any radionuclide composition effluent from the SDS has a larger strontium fraction than the influent. This is because cesium is held up on the SDS demineralizer beds much more than is strontium, as shown by the SDS data presented in Table I and Figure 3, Reference 11. Specifically, the decontamination factors listed in Table I, Reference 11, for processing the reactor building sump water are 140,000 for Cs-137 and 590 for Sr-90. The ratio of Sr-90 to Cs-137 activities in the influent to and effluent from the SDS were 0.04 and 10.2, respectively. It is therefore likely that all of the Epicor-II resins loaded to even a modest fraction of 1 μCi/g will be Class B.

Gross decontamination of the 305-ft elevation is being performed with water spray and a mechanical floor scrubber, as was done on the 347-ft level. Phosphoric acid solution (Turco 4512-A 10%) will also be used on parts of the 305-ft level. The phosphoric acid radwaste solution is to be neutralized and combined with other liquid waste in the holding tank at TMI. Since TMI-2 has no evaporation and solidification facilities for liquid wastes, they are stored. These stored liquid wastes may be solidified by vendors with mobile facilities called in for specific jobs. This was done once as part of a demonstration of the Dow solidification system but there are no specific plans for disposal of the stored liquid wastes as of this writing.

Strippable coatings are being used on some decontaminated sections of the 305- and 347-ft elevations and the polar crane to provide a protective barrier against recontamination. Except for the GDE testing, these coatings have not been used for decontamination on the upper part of the reactor building. These protective layers of coating are scheduled to remain in place until after the reactor head lift which is planned for Spring, 1983. The activity that these coatings contain will depend on both how much contamination they remove from the gross decontaminated surface and how much they receive from contamination of the top surface. The classification of the protective coating waste under 10 CFR Part 61 cannot be reasonably estimated until it is peeled from the surfaces and assayed prior to disposal.

Cleanup of the RCS water, Item 6, is generating highly loaded SDS zeolite and pre-filter radwaste. The pre-filters will also contain TRU activity from suspended fuel debris. These wastes will be taken by DOE.

Items, 7, 8, and 9 are interrelated to the extent that DOE will take the nuclear fuel and may take other TRU contaminated radwaste (in excess of the 10 nCi/g limit of the Hanford commercial burial site). It is uncertain if DOE will accept wastes having TRU activities in excess of 10 nCi/g. It is also uncertain if the 10 CFR Part 61 TRU limit of 100 nCi/g (Table 1, Appendix A) will be applied. Since much of the core has been reduced to rubble, the amount of fuel particulate contamination in radwaste resulting from these cleanup activities may be large. Therefore, SDS pre-filters from these cleanup activities may contain TRU contamination. The SDS zeolites from water cleanup in Items 7, 8, and 9 will be highly loaded with fission products and DOE will take these. Epicor-II polishing resins are loaded so as not to exceed 1 μCi/g in the dewatered form. This allows disposal of these wastes at the Hanford commercial disposal site without solidification. When 10 CFR Part 61 takes effect, this resin waste will require solidification if loaded to near 1 μCi/g, as is the practice as of this writing.

The decontamination procedures to be used in the hands on decontamination of the TMI-2 reactor building have not been determined as of this writing. Therefore, the waste streams that will be generated from this activity have not yet been defined. The Gross Decontamination has involved removing smearable contamination without significant surface penetration. There is some evidence that paint removal by abrasion, grit blasting or similar methods may ultimately be needed for complete decontamination.[8,25] Loss of coolant accident (LOCA) tests have shown that epoxy painted surfaces do not decontaminate readily after a LOCA.[25] Results from the GDE indicate that about 10% of the contamination remains in epoxy painted surfaces after gross decontamination. The activity remaining in floor surfaces averaged approximately 0.5 $\mu Ci/cm^2$ for Cs isotopes after GDE.[8]

There are no firm plans for the decontamination of the 282-ft elevation, Item 11, as of this writing. The wastes resulting from cleanup of this area of the reactor building are anticipated to contain very high levels of fission product activity and may contain significant TRU activity.

Waste management at TMI-2 is evolving as the waste streams from the various cleanup activities are processed. For wastes to be disposed of by commercial burial, efforts have been made to limit activity to LSA limits. Wastes which do not meet the LSA criterion are stored at TMI since, as of this writing, there are no procedures for packaging and shipping wastes which do not meet the LSA criteria.[26] These non-LSA wastes include some solid waste plus the chemical liquid wastes in holding storage at TMI-2. The liquids may be solidified by contractors brought in for these specific jobs, but this has not been planned as of this writing.

3. STRIPPABLE COATINGS - GENERAL

3.1 Introduction

Strippable coatings are film-forming compositions which are applied as liquids and which, after drying, are peeled from the surface. The great advantage of strippable coatings for radioactive decontamination is that only solid waste is produced. They have been found to be useful for removing loose debris and smearable contamination[9,27-33] and for significantly decreasing the level of particulates in the air.[9] The coatings are so useful as a barrier to protect cleaned surfaces from recontamination. If the protective layer is contaminated, it can be removed and replaced or, preferably, a second layer of coating can be applied trapping the contamination betwen two layers of film before stripping. They have also been used to immobilize loose contamination on equipment to allow moving it without spreading contamination.[30] Strippable coatings are most effective on smooth surfaces. Thicker applications, multiple applications, one on top of the other, and/or reinforcement with a matrix, such as cheesecloth, may be required to successfully strip from porous, pitted or corroded surfaces. The coating may not adhere to some surfaces, such as Teflon, or to oily surfaces. Decontamination factors (DF) of 10 are routinely obtained with one application of strippable coating, which compares favorably with DF obtained using liquid decontamination agents.[29]

3.2 Description

Chemically, strippable coatings consist of a film forming agent, usually in colloidal suspension, in a solvent. Other chemicals are added to the mixture to improve the decontamination or physical characteristics. Several strippable coating formulations have been described in the literature. One[31] uses polyvinyl alcohol as the filming agent. Another[28,32] uses prevulcanized rubber latex. A fast drying composition[33] uses a solution of copolymers of polyvinyl chloride and vinyl acetate. Although the filming agent alone is effective in removing loose contamination, the addition of chelating agents such as EDTA and NTA can significantly increase the decontamination effectiveness of these formulations.[28,31,32]

A typical formulation for polyvinyl alcohol based coatings[31] was given as follows: 2-10% polyvinyl alcohol in water, 1% EDTA, 15-20% ethanol, 0.02% sodium carbonate and 1-2% glycerine or ethylene glycol. This formulation consists of only 4-13% solids with the remainder being solvent which evaporates to leave the rubbery strippable film. The ethanol (optional) is included to aid the wetting of the surface. The sodium carbonate raises the pH and helps fix ions such as strontium and barium in the set coating. Glycerin or ethylene glycol acts as a plasticiser to keep the texture of set coating elastic and easily peelable from the surface.

The prevulcanized rubber latex strippable coating[28,32] comes in liquid form as an alkali stabilized emulsion with water. The formulation, which

incorporates an alkaline complexing detergent, is called Detex (for Detergent latex). Fine pumice can be added to Detex to obtain the added benefit of abrasion when the coating is applied with scrubbing.

A fast drying plastic strippable coating of copolymeric polyvinyl chloride (83-85%) and vinyl acetate (15-17%) is claimed to film on contact and to be removable after only 3 minutes drying time. The rapid filming allows this coating to be sprayed onto vertical surfaces without running. However, volatile organic solvents which allow the rapid drying can cause ventilation and flammability problems.

The characteristics that strippable coatings produce only solid waste in relatively small volumes and can be used for both decontamination and for protecting clean surfaces make them useful for radioactive decontamination. At least three vendors* in the United States offer strippable coatings for this use.

*Imperial Professional Coatings Incorporated, New Orleans, LA; Turco Products, Carson, CA; RAD Services, Incorporated, Pittsburgh, PA.

4. STRIPPABLE COATINGS AT TMI-2

4.1 Introduction

The strippable coating being used in the cleanup of the TMI-2 reactor building is Imperial* ALARA 1146 Decon. (Two strippable coating formulations -Imperial 1146 and 1148- were tested in the Gross Decontamination Experiment but only the 1146 coating was recommended for use because of problems encountered in stripping the 1148 coating.) Imperial ALARA 1146 Decon is a waterborne, vinyl composition incorporating chelating agents. A technical data sheet, put out by Imperial, on the properties of the 1146 coating is reproduced in Appendix B. The identities and quantities of chelating materials in 1146 are considered proprietary by Imperial.[34] According to the data sheet, the coating decontaminates a surface both physically and chemically; it absorbs and chemically binds heavy metal isotopes while wet and, upon curing, mechanically locks these absorbed contaminants plus smearable and loose contamination into a polymer matrix. It is also claimed to be useful for protecting clean surfaces, for inhibiting airborne contamination and for shielding against beta emitters. (A typical 25-mil thickness of 1146 coating would provide effective shielding for low energy beta emitters, up to approximately 0.3 MeV.[35] However, this 25-mil thickness would be almost completely ineffective for shielding the 2.2 MeV beta electron from the Y-90 daughter of Sr-90.)

Imperial 1146 strippable coating was tested on the floor of 305-ft elevation of the TMI-2 reactor building as part of the Gross Decontamination Experiment (GDE). This surface[1] consists of a concrete substrate finished with K and L** 7107 epoxy primer and K and L 7475 epoxy paint. (All of the concrete surfaces in the reactor building are finished with K and L 7107 and 7475 epoxy primer and paint; reference 1, Table 4.1.) For testing[10] in the GDE, the 1146 coating was applied to an approximately 500-ft^2 area of the 305-ft. elevation in the northwest quadrant north of the open stairwell. This application followed low pressure water flushing of the area to remove loose debris and water soluble contamination. A description of this test of the 1146 strippable coating including the volume and an estimation of the activity of the radwaste produced is included in Reference 10. (This information is detailed in Section 4.3 of this report along with other experimental results on TMI strippable coatings from the GDE.)

4.2 Testing ALARA 1146 Strippable Coating for Characterization as Radwaste

Tests were performed on ALARA 1146 coating purchased from Imperial Professional Coatings, Incorporated, and on samples of ALARA 1146 from the GDE testing on the 305-ft. elevation floor of the TMI-II reactor building. Characterization of ALARA 1146 is based on test results for leachability of Co,

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*Imperial Professional Coatings, Incorporated, P.O. Box 2977, New Orleans, LA, 70189.

**Keeler and Long, Incorporated, Watertown, CT.

Sr, and Cs radionuclides and chelating agents and on tests for stability toward irradiation, biodegradation, immersion and heat. Scoping test results on the purchased material are presented in Section 4.1 while test results on the samples of the actual TMI-II coating are presented in Section 4.2.

4.2.1 Scoping Tests

4.2.1.1 Sample Preparation

Samples of ALARA 1146 coating were prepared according to directions in the technical data sheet, Appendix B. The shelf life of liquid coating mix is 4 months according to Appendix 8. The liquid coating mix was used within 1 month of its arrival in accordance with vendor recommendations. The liquid material was stirred before and during application and was spread evenly over a surface to cure for at least 2 days (full cure) before stripping. The coating was spread with a groved Teflon sheet instead of being sprayed or applied with a roller as is suggested in the technical data sheet. Spraying and rolling were impractical on the laboratory scale of these tests, and as long as the coating is merely spread and not mechanically scrubbed, the method of application should not matter. The surfaces on which the coating was spread included Lucite sheets and small painted forms* of concrete and steel similar to the epoxy painted surfaces in the TMI-II reactor building. The stripped coating used in all of the scoping tests was 18 $\pm$ 2 mils thick. For the radionuclide leach tests, 40 mL of coating mix was spiked with 0.2 mL of an aqueous solution containing 20 µCi Cs-137, 40 µCi Sr-85 and 40 µCi Co-60. Spiked samples for leach testing were prepared this way rather than by contaminating the surfaces with the spike, allowing it to dry and then applying the coating in order to avoid contamination of the painted samples and to prepare the contaminated coating such that the activity would be held in the coating as firmly as possible. This would then provide radionuclide leach data which should be a measure of the best performance (i.e., lowest leach rates) that could be expected of coating used for actual decontamination. There was no detectable activity remaining on the epoxy painted forms after the coating was stripped indicating that the distribution coefficient between the coating and epoxy painted surfaces is large and in favor of the coating for the radionuclides as tested. The spiked coating mix was stirred for five minutes before being applied to the K and L epoxy painted specimens for cure.

4.2.1.2 Radionuclide Leach Testing for Sr, Cs and Co

Scoping test samples of ALARA 1146 spiked with Cs-137, Sr-85 and Co-60 were prepared as described in Section 4.2.1.1. Half-inch square test specimens - three each from the K and L painted steel and painted concrete

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*Specimen forms furnished by the Keeler and Long Corporation (K and L) included 2x4x1/4-in. carbon steel coupons primed and painted with 6548/7107 epoxy primer and E-1-7475 epoxy enamel and 2x4x2-in. concrete blocks finished with 4129 epoxy concrete curing compound, 6548S epoxy surfacer and E-1-7475 epoxy enamel.

forms - were cut from the spiked coating which had been peeled from the painted surfaces. These specimens were then leached in 32.3 mL deionized water for a leachant volume to specimen surface area ratio of 10.0 cm. Leachants were changed and the activity remaining in each of the test specimens was counted at 1, 2, 3, 4, 8, 9, 11, 15, and 18 days. A GeLi detector was used for gamma counting. Counts were integrated over 5 or 10 minutes, a decay correction applied for Sr-85, and the measured activity compared to that initially present in the sample. Figure 4.1 shows a plot of leached activity at each of the points of measurement stated above. The activity leached was calculated according to Eq. (4.1) for each isotope.

$$\text{Activity Leached (\%)} = \sum_i \frac{C_0 - C_i}{C_0} \times 100 \qquad (4.1)$$

C_0 and C_i are the initial activity and the activity at day i, respectively, measured in a test specimen. The standard deviation of the Sr measurements (six test specimens) is indicated by the error bar attached to each date point. The standard deviation for the Co and Cs data were all less than the width of the symbols and are not shown.

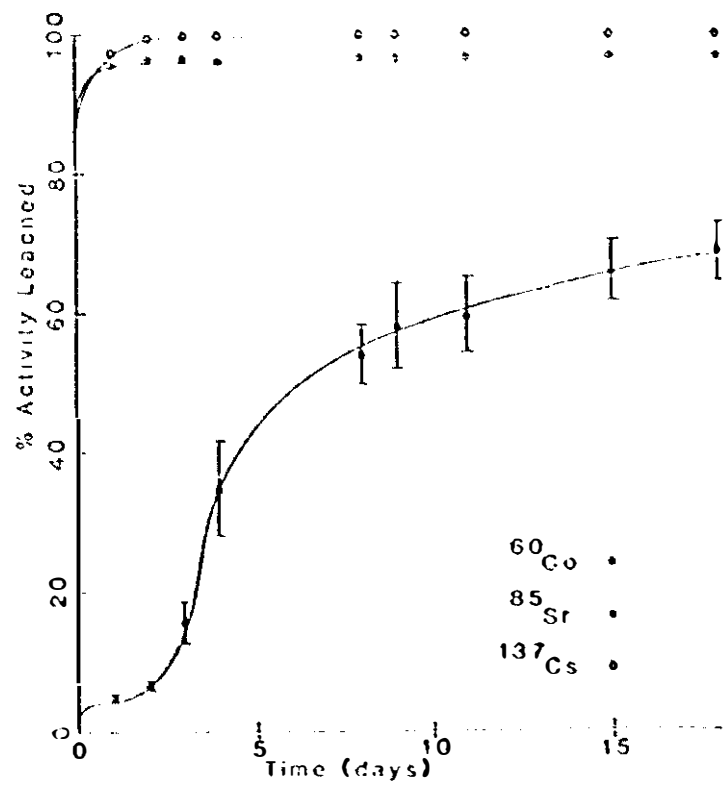

Figure 4.1 Percent activity loss vs time for Imperial 1146 strippable coating leached in deionized water.

The data in Figure 4.1 indicate that Cs and Co leached readily from the coating upon contact with water. All of the Cs activity leached from the coating within four days with over 95% being released in the first leach volume. The behavior of Co paralleled that of Cs except that the coating retained about 4% of the initial Co activity. The behavior of Sr was more complex and varied considerably between test specimens, as indicated by the magnitudes of the error bars. About 5% of the Sr activity leached immediately. However, the curve shows that a longer period of contact with water was needed to release the major part of the Sr activity from the coating. About half of Sr activity was released readily and then the rate of release slowed markedly.

The radionuclide leaching of the coating measures the combined result of at least two effects: (i) the effect of the chelating agents in the coating and, (ii) the properties of the coating material itself. Chelating agents tend to solubilize metal ions and would be expected to enhance leachability whereas radionuclides more strongly bound to the coating material leach less readily. The relative quantity of chelating agent to radionuclide metal ion generally is very large and not a limiting factor.* One may speculate from Figure 4.1 that the Cs and most of the Co may be held in the set coating in the chelated form whereas most of the Sr may be bound preferentially to the coating.

4.2.1.3 Chelate Leach Testing

The ALARA 1146 strippable coating contains chelating agents the identities and quantities of which are considered confidential by Imperial.** According to the definition in 10 CFR Part 61.2, "Chelating Agent" means amine polycarboxylic acids (e.g., EDTA, DTPA), hydroxy-carboxylic acids, and polycarboxylic acids (e.g., citric acid, carbolic acid, and glucinic acid). 10 CFR 20.311b requires that radioactive waste containing more than 0.1% by weight of chelating agents be identified on the shipping manifest and that the weight percentage of chelates in this waste be estimated. As part of an agreement[30] between Imperial Profession Coating Corporation and BNL, Imperial disclosed the identities and quantities of chelating agents in the ALARA 1146 composition to BNL. Based on this information from Imperial, BNL performed standard analyses for the chelating compounds in leachate samples from leaching ALARA 1146 coating in deionized water. The results from these tests on chelates leached from the coating were consistent with the quantities

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*The quantities, in gram atoms (or moles) per μCi of activity, of the radionuclide of concern are: Cs-137 = 8.4×10^{-11}, Sr-90 = 8.1×10^{-11}, Co-60 = 1.5×10^{-11}. Chelating agents generally have molecular weights in the range of about one hundred to several hundred. Assuming, for purposes of conservative calculation, a molecular weight of 1000 and a concentration of 0.1% by weight results in 1×10^{-6} moles of chelate per gram of material. Therefore, for an activity loading on the order of 1 μCi/g in our hypothetical example, there would be more than 10,000 chelate molecules for each radionuclide ion.

**Personal communication between R. Taylor (Imperial) and R. Barletta (BNL), June 7, 1982.

of chelates which were indicated by Imperial to be present in the ALARA 1146 composition. As such, the concentration of chelating agents in the coating wastes does not require identification on the shipping manifest.

4.2.1.4 Immersion Testing

Immersion tests were conducted to determine changes which might be expected when strippable coatings are contacted with five liquids that are believed to bound the range of liquid compositions that may be found in shallow land burial environment. These included a commercially prepared liquid scintillation counting (LSC) cocktail, toluene, xylene, deionized water, and deionized water saturated with xylene and toluene. The LSC cocktail forms a two-phase suspension with water so this mixture was not tested. The results for water and the water saturated with toluene and xylene are the more relevant to conditions expected in a burial environment.

One-inch square coating specimens were weighed and immersed in 50 mL of the liquids. Four samples were exposed to each liquid. Weight gain was measured as a function of time at 1, 2, 6, 8, 9, 13, 17, and 34 days of immersion. Data were calculated as:

$$\text{Weight Gain (\%)} = \frac{W_i - W_o}{W_o} \times 100 \qquad (4.2)$$

where W_i is the weight of the sample at time i and W_o is the weight of the sample before immersion. These data are shown in Figures 4.2 and 4.3.

After 34 days of immersion, the samples were removed from the liquids to air dry in the laboratory. Samples were weighed after 1, 2, 3, 4, and 7 days and percent weight changes calculated with respect to the pre-immersion weights. These data are shown in the plots in Figures 4.4 and 4.5. (The points at day zero in Figures 4.4 and 4.5 are the same as those at day 34 in Figures 4.2 and 4.3, respectively.)

Dimensions of the initially 1-in. x 1-in. samples were measured following the immersion phase and again following the drying phase. The immersion liquids were also evaporated and the residues weighed. These data are listed in Table 4.1 along with computed values for the volume gain based on the observed weight gain and the density of the immersion liquid.

These data indicate that ALARA 1146 coating is affected by exposure to water and the organic liquids tested. The coating swells upon immersion and shrinks upon drying. A part of the coating dissolves upon immersion, as evidenced by the presence of a residue upon evaporation of the immersion liquid. For water and water saturated with toluene and xylene, the residue is a dry powder. For toluene and xylene the residue is a viscous oil. In water and the water solution saturated with toluene and xylene, the sum of the final weight (after drying) and the residue weight is less than the initial weight, as shown in Table 4.1, whereas for toluene and xylene, the final weight plus the residue weight is greater than the initial weight by approximately 10%. This may suggest that toluene and xylene react with some constituent of the coating.

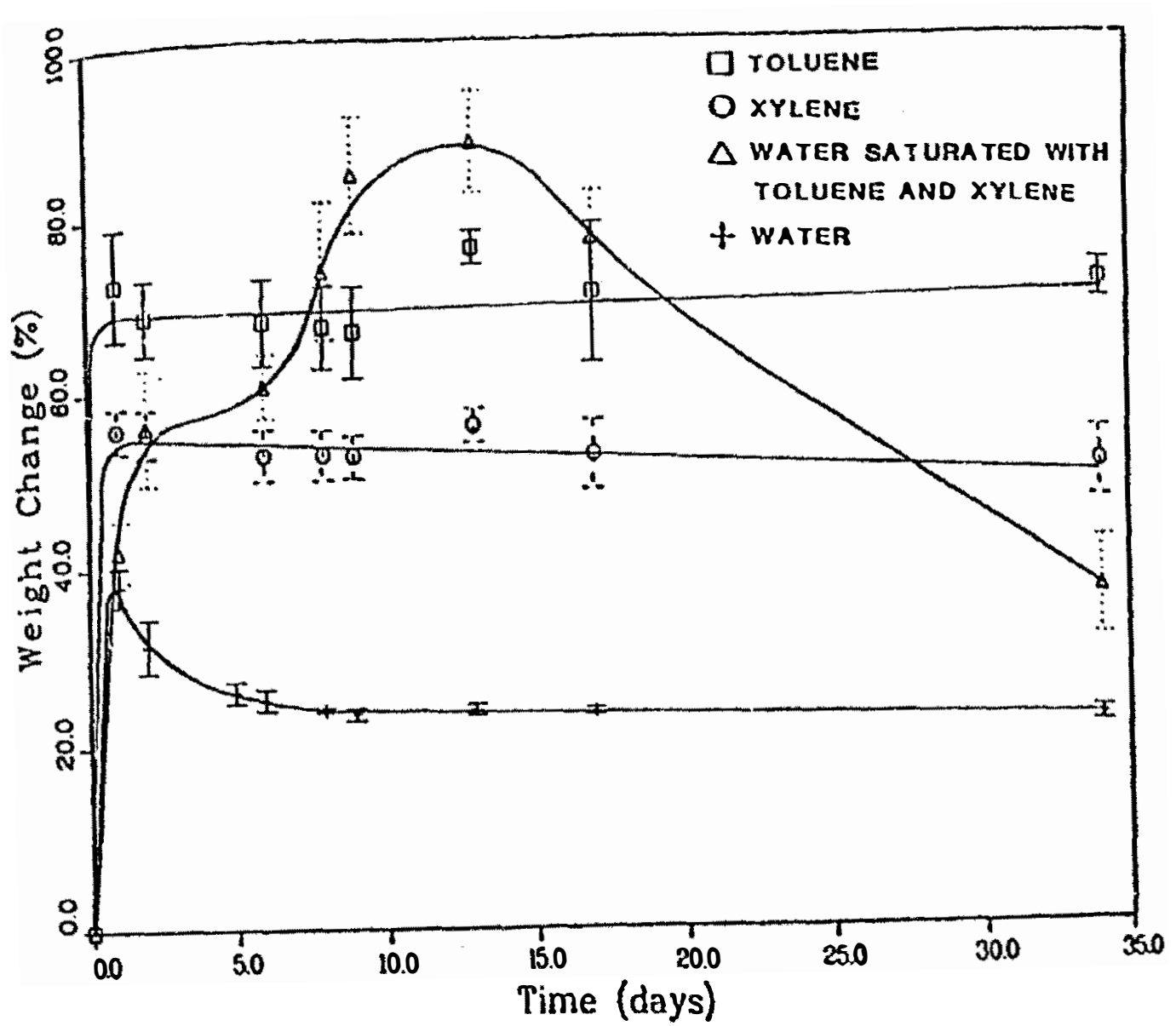

Figure 4.2 Percent weight gain of ALARA 1146 strippable coating upon immersion in toluene, xylene, water, and a solution of water saturated with toluene and xylene.

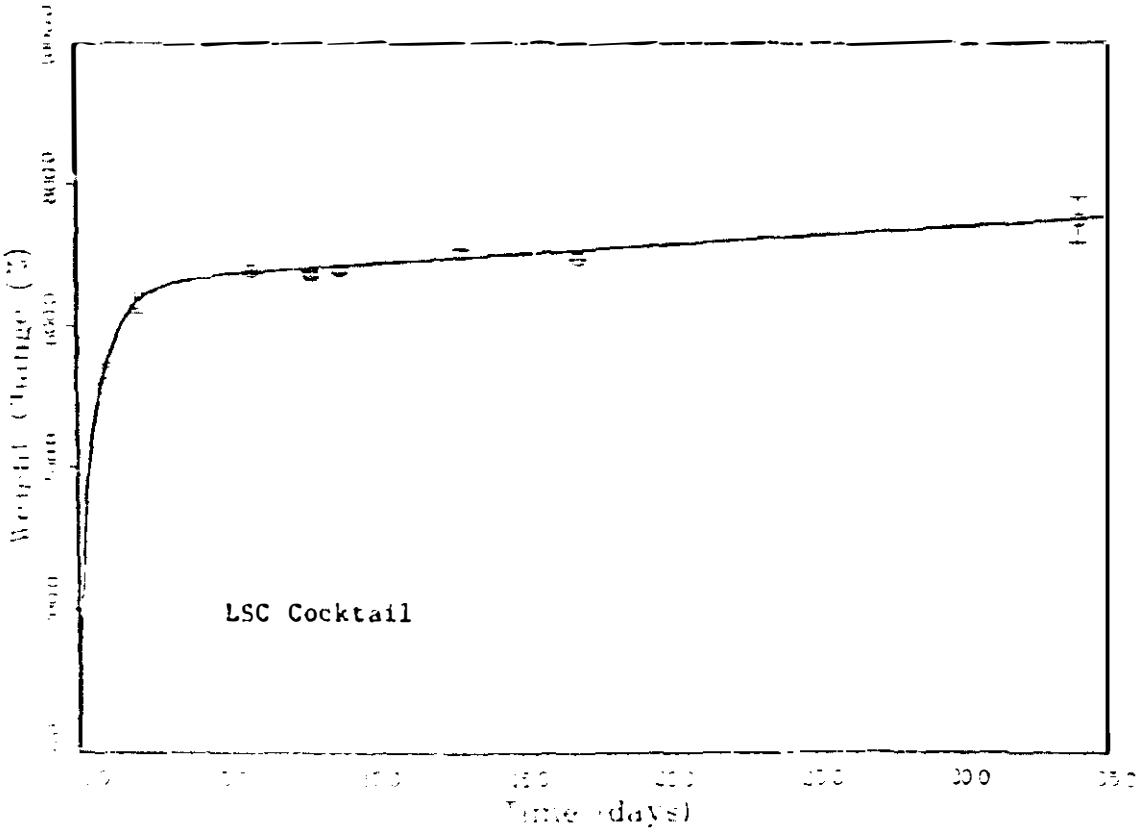

Figure 4.3 Weight gain of ALARA 1146 strippable coating upon immersion in LSC cocktail.

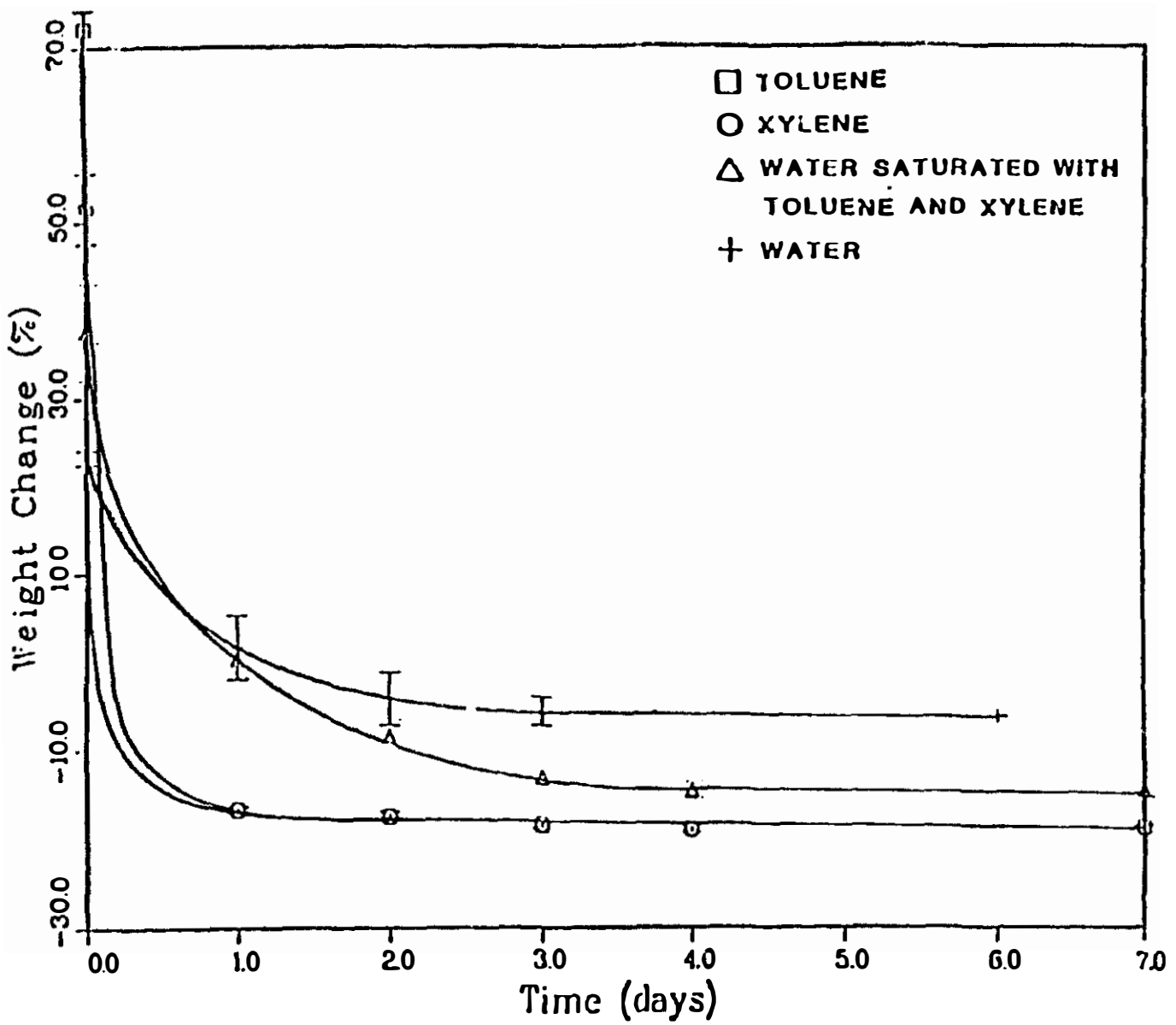

Figure 4.4 Drying curves at room temperature in air for ALARA 1146 stippable coating following 34 days immersion in toluene, xylene, water, and a water solution saturated with toluene and xylene.

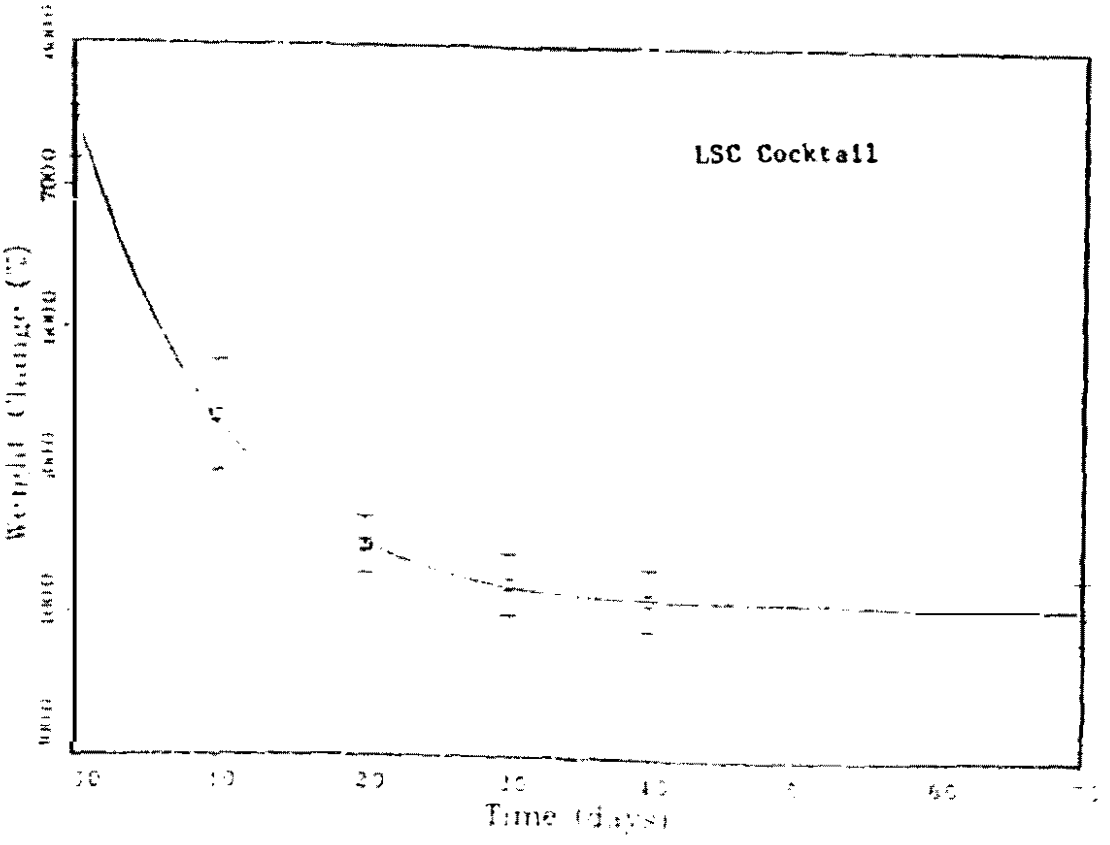

Figure 4.5 Drying curve at room temperature in air for ALARA 1146 strippable coating following 34 days immersion in LSC cocktail.

Table 4.1

Immersion and Drying Data for ALARA 1146 Strippable Coating

Liquid	Immersion Phase: Weight Gain (%) Maximum	Weight Gain (%) Final[a]	Volume Gain (%)[b] Maximum	Volume Gain (%)[b] Final[a]	Test Immersion Dimension (in ± 0.01 in.)	Drying Phase: Weight Loss (%)	Final Dimension (in ± 0.01 in.)	Residue Weight (% of Initial Sample Weight)[c]
Water	36 ± 2	23 ± 1	84	53	1.01	6.2 ± .2	0.88	4.6
Water Saturated with toluene and xylene	88 ± 6	37	159	71	1.08	14.6 ± .2	0.88	5.2
Toluene	76 ± 2	72	146	146	1.20	18.8 ± .2	0.87	28.3
Xylene	57 ± 2	56	113	113	1.15	19 ± .4	0.98	29.2
LSC Cocktail	751 ± 32	534	---	---	1.9	---	not available[d]	---

[a]After 34 days immersion.
[b]Calculated from weight gain data using the formula:

$$\text{Volume Increase (\%)} = \left[\left(\frac{\rho_0}{\rho_i}\right)\left(\frac{W_i}{W_0}\right) - 1\right] \times 100$$

where ρ_0 and W_0 are 1.24 g/cm^3 and the initial sample weight of each 1x1-in. piece of coating, respectively, and ρ_i and W_i are the density of an weight gain in each of the immersion liquids, except LSC cocktail.
[c]The leach liquids of each type were combined and evaporated so the value listed is the average for the four samples in each liquid.
[d]The test samples were hung on wires to dry. These samples had lost so much mechanical strength that they dripped off wires into viscous puddles. The puddled residues could be weighed but their dimensions could not be measured.

4.2.1.5 Biodegradation Scoping Tests

The biodegradability of ALARA 1146 strippable coating was determined two ways. The rate of biodegradation in soils from the Barnwell, SC, and Hanford, WA, shallow land burial sites was quantitatively monitored by measuring the CO_2 produced from microbial respiration. Microbial CO_2 production in soil was monitored using the special flasks and procedure described by Bartha and Pramer.[36] Biodegradation in soil[37-41] is controlled by the inherent biodegradability of the material plus related soil factors including moisture and nutrient mineral content, temperature and microbe types and population. In addition an evaluation of the ability of microbes to utilize the coating as a food source was performed using the ASTM tests G21 (Determining Resistance of Synthetic Polymeric Materials to Fungi) and G22 (Determining Resistance of Plastics to Bacteria). The Branch Technical Position on Waste Form (204.1.5/TCJ/1/5/83, February 14, 1983) specifies the use of G21 and G22 for testing resistance to biodegradation of waste forms which fall under 10 CFR Part 61 Class B or C. In G21 and G22, an ideal environment of moisture, temperature, and nutrient minerals is provided; these tests measure the inherent biodegradability (i.e., ability to be used as a food source) of a material to the microbes specified in the tests.

For biodegradation of Imperial 1146 strippable coating in soil, the 1146 coating was cut into strips approximately 1/2 x 2 inches. Fifteen to twenty of these strips with a total weight of approximately 12 g were used for each sample. Each of these samples was mixed in a special flask with 100 g moist soil (the moisture content was adjusted until the soil was wet but still porous and not muddy) from either the Barnwell or Hanford shallow land burial site. Two sample-in-soil tests were run for both of the burial site soils to provide an estimation of the reproducibility of the results. One sample plus 15 mL water was loaded into a flask without soil and two more flasks containing 100 g of the moist soil from the Barnwell and Hanford sites were used as controls. The difference between the CO_2 produced in the flasks containing soil plus sample and the control containing soil only indicated sample biodegradation (more CO_2 from sample flasks), inertness (no difference in CO_2 production) or toxicity (decreased CO_2 production). A standard potassium hydroxide (KOH) solution, 0.100 M, was used to absorb any CO_2 produced in a flask. When the KOH solution absorbed CO_2, less acid was required for titration of the solution to the phenolphthalein indicator end point. To obtain the amount of CO_2 generated, the KOH solution was removed from the flasks and titrated with 0.0500 M HCl solution. The amount of acid needed to titrate the remaining KOH is related to the CO_2 absorbed by

$$CO_2 \text{ (mg)} = (B-V)NE$$

where B = mL HCl to titrate the soil control, V = mL HCl to titrate the sample in the soil, N = normality of the HCl and E = 22 (the equivalent weight of CO_2). The time interval between titrations is determined by the rate of CO_2 generation so as to not expend more than 2/3 of the KOH in absorbing CO_2.

Results from the biodegradations in Barnwell soil, B1 and B2, and in Hanford soil, H1 and H2, are shown in Figure 4.6 and listed in Table 4.2. Figure 4.6 shows the total quantity of CO_2 generated by each of the four samples with time. Table 4.2 lists data for each including the quantity of CO_2 generated and sample weights before and after the 220 days of biodegradation. The amount of biodegradation that has occurred is estimated from these data. Carbon dioxide generation monitored after the samples were removed from the soils indicated that microbial activity continued at a level significantly above that in the soil controls but was decreasing steadily with time.

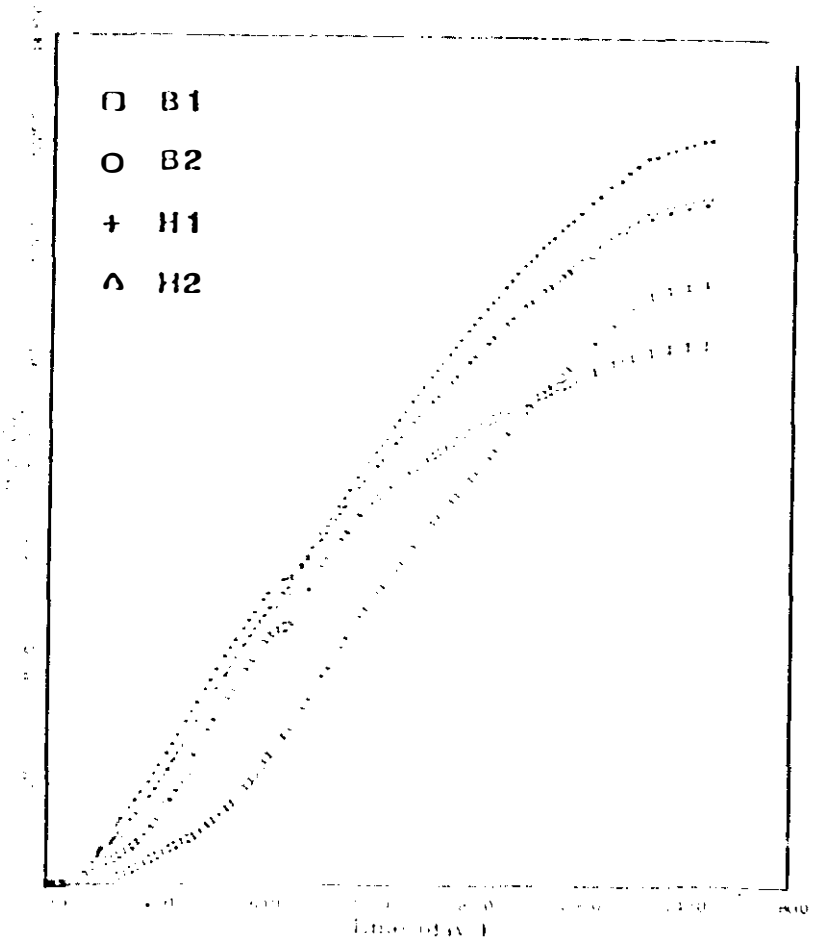

Figure 4.6 Biodegradative CO_2 gas evolution from scoping tests on Imperial 1146 coating in Barnwell and Hanford soils.

Table 4.2

Scoping Test Results on the Biodegradation of ALARA 1146 Strippable Coating in Soils From the Barnwell, SC, and Hanford, WA, Shallow Land Burial Sites

Sample	Initial Weight W_o (g)	Final Weight W_f (g)	Biogenically Generated CO_2 (mg)	Carbon Lost[a] % C	Carbon Lost[a] $\frac{\% C^b}{0.6}$	$\frac{W_o - W_f}{W_o} \times 100^c$
B1	12.11	11.25	875.5	3.77	6.28	7.10
B2	11.97	11.05	961.0	4.19	6.98	7.69
H1	11.24	10.31	1094.5	5.08	8.47	8.27
H2	11.24	10.48	1189.5	5.52	9.20	6.76
Control	11.74	12.18	nil	----	----	-3.75[d]

[a]From the biogenically generated CO_2 data and the measured 52.3% C content of the coating used in these tests.
[b]This is the estimated upper limit of biodegradation based on this CO_2 generation data and published observations, References 39 and 40, that as little as 60% of the available carbon may be evolved as CO_2 during biodegradation.
[c]Percent biodegradation from the initial and final weight following 220 days of biodegradation in soil at 20-24°C.
[d]The sample did not detectably biodegrade in the absence of soil. The weight increase should not be due to retained water since all samples were dried both before and after the tests in the same environment.

Carbon dioxide monitoring provides a lower limit to the amount of biodegradation that has occurred. This is so for two reasons. First, as little as 60% of the biodegradable carbon in a substrate may be evolved as CO_2;[39,40] the remainder is incorporated into additional microbe mass or excreted as metabolic waste other than CO_2. Second, only carbon metabolism is measured by this technique; thus, neither the metabolism of hydrogen and oxygen to produce water nor other transformations (e.g., sulfate or nitrate metabolism) which may occur in the substrate are accounted for in this method. However, CO_2 monitoring does provide a quantitative measure of biodegradation and allows measurement of changes in the rate of biodegradation with time.

Comparison of initial and final sample weights should be a direct measure of the amount of biodegradation that has occurred. Difficulties in separating tightly bonded soil from the coating, the loss of very small amounts of coating with the soil removed from the coating surface and contributions from microbe mass retained on the coating complicated the interpretation of this data. The weight added by retained soil and microbe mass is

offset to some degree by coating removed with soil. A greater quantity of soil was incorporated into the coating than coating was scraped off with the soil (with the probable exception of B2 which is the only sample for which the biodegradation measured by weight change did not lie within the range of probable biodegradation as determined by carbon dioxide monitoring). These factors considered together led us to conclude that the final weights of the samples in soil (i.e., B1, B2, H1 and H2) may be each slightly high, perhaps by as much as, but certainly not more than, a few tenths of a gram.

There are several prominent features in Figure 4.6. Two of these features, the 10-day initial part of the curves with very low rates of CO_2 generation and the "kinks" in the curves from day 84 to approximately day 94, resulted from unplanned occurrences during the tests. The initial region of each of the curves is characterized by a low rate of CO_2 evolution followed by the onset, at day 10, of much higher rates of biodegradation. This was probably caused by cold temperatures (45-57°F daytime, colder at night) due to failure of a steam valve which supplied heat to the lab. From day 10 on, lab temperature was maintained between 68 and 74°F. The region of reduced CO_2 evolution, which is especially noticeable in samples B1 and H1, from day 84 to about day 94 was caused by an accident on day 84 in which some alkaline solution was spilled into some or all of the samples. The microbes recovered from this trauma and the rates of CO_2 generation returned to pre-accident levels within 10 days.

A difference in the way the coating samples were mixed with the soils is probably the cause of relative difference in the B1, B2 and H1, H2 curves. Sample B1 was mixed with soil such that it had approximately 50% more surface area in direct soil contact. Specifically, there was less overlapping of coating strips in B1 than in B2, H1, and H2. B1 initially biodegrades significantly faster than B2 whereas, the rates for H1 and H2 were very similar to each other. It appears that biodegradation of this material proceeds faster in Hanford soil, all other things being equal. The rates of CO_2 generation from all four samples are essentially equal from about day 80 to 120, at which point the rate of CO_2 generation in B1 slows significantly. The reason for this slowing is not clear. It should not have been caused by the accident at day 84 since there was a complete recovery in CO_2 generation and no similar slowing occurred in H1 which was also severely affected by the accident. Although CO_2 generation from all of the samples was gradually slowing, this effect in B1 was much more pronounced.

The samples were removed from the soils on day 220 of the experiment. CO_2 production from these soils minus samples decreased significantly but did not return to background. This may be due to the small quantities of coating adhering to the soil in direct contact with the samples and possibly due to a soluble fraction of the coating, as is indicated by the immersion testing (Section 4.1.4), remaining with the soil. The rate of CO_2 generation in the soils after sample removal gradually decreased toward background with time.

The ASTM tests G21 and G22 were run on coating samples approximately 1 x 1 in. in size. G21 (fungi) showed no growth, i.e., a growth rating of zero on a scale of 0-4 in the test. G22 (bacteria) was positive for bacterial growth on the coating specimen. (There is no quantiative scale in G22, results are reported as growth or as no growth.)

4.2.1.6 Radiation Testing

The stability of ALARA 1146 strippable coating to ionizing radiation was tested using coating specimens sealed into Pyrex tubes both in an air atmosphere and under vacuum. Gamma irradiation was carried out in the Co-60 irradiation facility at BNL. Data included pressure measurements for the tubes both before and after irradiation and mass spectroscopic analysis of the gas in the tubes after irradiation. The coating generated gas when exposed to ionizing radiation, as was expected, since chemically, it is an organic substrate.[42,43] It also was changed physically by the irradiation, becoming much stiffer and stronger with increasing dose. This change could also be anticipated since the polymeric coating would be expected to cross-link upon exposure to ionizing radiation.[43,43]

The measured data from the sealed tube irradiations in air and vacuum are shown in Figure 4.7. Three experimental configurations, as differentiated by the symbols, were used in the 20 cm^3 sealed tubes: squares - 0.5-g coating in air; triangles - 5.0 g coating in air, and circles - 5.0-g coating in vacuum. The vertical bars through the center of each symbol represent the standard deviation of the four measurements at each data point. The 0.5-g sample irradiations were performed first to guide the selection of points for further study. A preliminary gas analysis indicated that the negative slope of the line defined by the square data points up to about 10^8 rad was caused by oxygen depletion. At doses greater than 10^8 rad, radiolytic gas generation became dominant. The line defined by the square data points at 5 x 10^8 rad and 1 x 10^9 rad indicate a G value for gas generation of 1.3.

The quantity of gas generated per gram of coating was less in the sealed tubes containing 5.0 g of coating than in the preliminary tests using 0.5 g coating. This is probably due to the higher pressures, as listed in Table 4.3, produced in tubes with the larger amount of coating. The higher pressures would increase the extent of back reaction (i.e., the coating would react with the radiolysis gas and reduce the net amount of gas produced) and a lower apparent G value. The pressure in the tubes containing 0.5 g of sample irradiated to 1 x 10^9 rad was slightly less than 2 atm.

Table 4.3 lists the gas generation data for doses of 10^8 rad and greater, for the points indicated by circles and triangles in Figure 4.8, i.e., for the 5.0-g samples irradiated in air and under vacuum. The pressure in these tubes at the end of the irradiation is also listed. For reference, the composition of dry normal air at sea level and a gas analysis for an unirradiated 5.0 g sample sealed from air are also listed. The sensitivity of the gas analysis is in the range of 0.05% by volume for detection. The data show that oxygen is scavenged from air over the samples and that hydrogen is the

primary radiolysis product being generated. Carbon dioxide, hydrocarbons including methane, ethane, propane, butane plus others and a small amount of carbon monoxide account for the remainder of the gas produced by radiolysis of the coating. Radiolytic gas generation causes also the coating to blister, as shown in Figure 4.8 for a piece of coating irradiated to 1×10^9 rad.

The gas analysis of the sample which was sealed in a tube but not irradiated was surprising - the coating appears to scavenge oxygen and liberate carbon dioxide, carbon monoxide, and a small amount of hydrogen spontaneously at room temperature. The unirradiated sample remained sealed for 7 weeks prior to analysis. It seems unlikely that biodegradation could be responsible for the oxygen uptake and CO_2 generation in the sealed tube based on the CO_2 monitoring results of Section 4.1.5 on the moistened coating not in soil. Also, there was no moisture in the sealed tube to support biodegradation, and carbon monoxide, which was produced in the sealed tube, is a product of decomposition, not biodegradation.

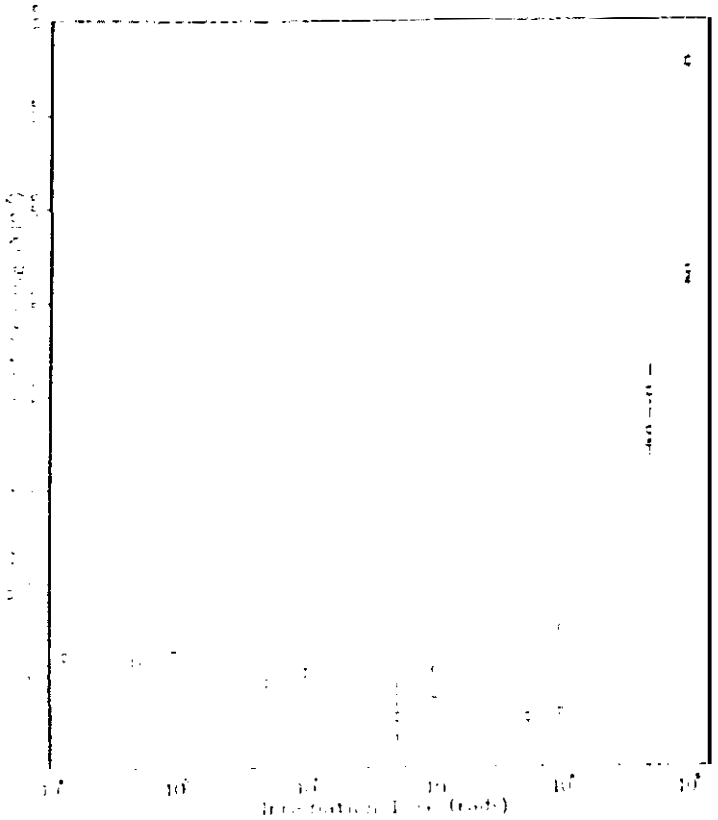

Figure 4.7 Plot of gas generation vs Co-60 gamma dose for ALARA 1146 strippable coating. Three sample configurations were used in 20 cm^3 sealed tubes: squares - 0.5-g coating in air; trianges - 5.0-g coating in air and; circles - 5.0-g coating in vacuum.

Table 4.3

Gas Composition Analysis From Co-60 Gamma Irradiated Tests of ALARA 1146 Strippable Coating in Air and Under Vacuum[a]

Irradiation Dose (rad)	Initial Environment	Pressure (atm)	Gas (% by Volume)											
			N_2	O_2	Ar	CO_2	H_2	CO	CH_4	C_2H_6	C_3H_8	C_4H_{10}	Other Hydrocarbons	Σ
0[b]	----	1.0	78.1	20.9	0.93	0.034								99.964
0[c]	air	0.9	72.5	11.8	0.90	10.8	0.25	3.7						99.95
5×10^8	air	4.8	13.3			12.6	65.4	2.0	2.1	0.7	2.2	1.1	0.6	100.0
1×10^9	air	7.3	6.6	0.2		14.8	70.3	1.8	2.3	1.0	1.8	0.8	0.5	100.1
1×10^8	vacuum	0.7		0.09		12.3	76.8	2.0	1.9	0.6	2.5	2.7	1.2	100.09
1×10^8	vacuum	4.0		0.4		11.7	78.3	2.0	2.1	0.9	2.3	1.7	0.4	99.8
5×10^8	vacuum	6.7		0.2		15.0	75.1	2.6	2.6	1.1	2.0	1.0	0.5	100.1

[a]Five grams of ALARA 1146 coating were irradiated in sealed tubes with a total volume of 20 cm^3. The plenum volume is 16 cm^3 based on the 1.24 g/cm^3 density of the coating as can be determined from specifications in the ALARA 1146 Technical Data sheet, Appendix B.
[b]Normal air at sea level exclusive of water vapor.
[c]Data from a sample tube that was not irradiated. It was sealed at the same time as the tubes for irradiation and the seal was broken for analysis 7 weeks later, when all of the analyses were performed.

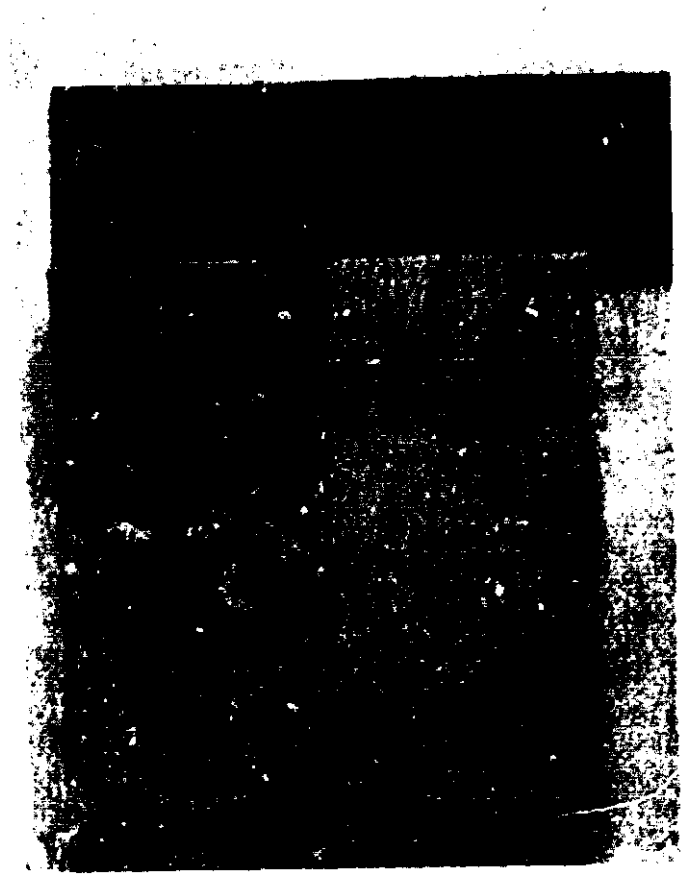

Figure 4.8 Effect of radiolysis on ALARA 1146 coating (left) vs unirradiated specimen (right).

4.2.1.7 Thermal Testing

The thermal resistance of ALARA 1146 strippable coating has been tested according to ASTM E84-77 (Surface Burning Characteristics of Building Materials).[44] This test was performed to document the compliance of the 1146 coating to NRC Draft Regulatory Guide 1.120, "Fire Protection Guidelines for Nuclear Power Plants" (Revision 1, November 1977). ASTM E-84 compares the burning characteristics of a test material to those of asbestos cement board, which is nonflammable, and to red oak wood paneling. Test results are stated numerically on a scale of 0-100 in which the asbestos cement board results define zero and the red oak wood paneling results define 100 on the scale. A 20-25 mil thickness of the 1146 coating had a flame spread rating of 20 and a 30-35 mil thickness was rated at 40. The NRC Draft Regulatory Guide 1.120 limits the flame spread behavior to a value of 50 for use in nuclear plants.

To determine the effect of heat on the coating, without burning, 1 x 1 in. pieces of coating were sealed into glass tubes at atmospheric pressure and then heated for 24 h at 50,100 and 200°C. there were no visible changes

in the coating samples heated to 50 and 100°C whereas the 200°C sample became darker in color. Table 4.4 lists the results of gas analyses on the atmospheres in the tubes. The final pressure in all of the tubes was reduced to a small fraction of the initial pressure of approximately 0.9 atm, which is normal for this procedure.* The disappearance of most of the oxygen from the air over the samples provides additional evidence that the 1146 coating scavenges oxygen from air. Specifically, it was previously observed (c.f. line 2, Table 4.3) that oxygen was disappearing from the air over a piece of 1146 coating in a sealed tube at room temperature. Raising the temperature, as has been done in the thermal tests, would be expected to increase the rate of reaction. Likewise, the generation of carbon dioxide, hydrogen, and carbon monoxide was anticipated plus some pyrolysis at higher temperatures to produce various organic decomposition products. The absence of the nitrogen and argon, as indicated by the low final pressure, as well as the gas analysis was surprising since these gases are inert under the thermal testing conditions used (argon is chemically inert under all conditions). A possible explanation may be that some of the organics generated by thermal decomposition have significant vapor pressure at elevated temperatures but not at room temperature coupled with a leakage of gas from the tube at elevated temperatures and high internal pressures. One could then also expect condensate in the sealed tube after cooling. Although none was observed, no effort was made to determine the possible presence of condensate at the time of the experiment and a colorless, transparent film could have escaped observation. The explanation for the low final pressures in the tubes following the thermal tests is based only on speculation and these results should remain suspect until independently verified by additional experiments.

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*The heat from sealing the break-seal tubes used in these procedures, as well as in the irradiation tests, normally reduces the pressure in the sealed tubes to approximately 90% of ambient pressure.

Table 4.4

Gas Analysis From the Thermal Testing of ALARA 1146 Strippable Coating[a]

Test Temperature (°C)	Final Pressure (atm)	Gas (% by Volume)							
		N_2	O_2	CO_2	H_2O	H_2	CO	Hydrocarbons[b]	% Total
50	0.01	not measured							
100	0.01	37.3	1.4	21.7	36.6	0.9	-	2.0	99.9
200	0.07	11.3	0.5	74.5	-	6.9	4.3	2.8	100.3

[a]Tests were performed on 1 x 1 in. pieces of coating weighing ~0.5 g which were sealed into 20 cm^3 glass tubes at atmospheric pressure and heated to the indicated temperature for 24 h.
[b]The hydrocarbons detected included methane plus higher alkanes and benzene. Alcohols were also detected.

4.2.1.8 Scoping Tests Summary

The results of the scoping tests on ALARA 1146 strippable coating, as presented in Sections 4.1.2 through 4.1.7, are summarized below.

- Radionuclide leach testing in deionized water showed that Cs leaches readily and completely from the coating; Co leaches readily except for a small amount retained by the coating while Sr leaches from the coating much more slowly and with large variations in leach rate between different coating pieces.

- Chelate leach testing indicated that the chelating agents contained in the 1146 coating leach in deionized water. However, the quantity of chelating agent contained in the coating is less than 0.1% by weight.

- Immersion testing showed that the 1146 coating is attacked by water and the several organic liquids tested. It absorbed liquid, as indicated by weight gain and swelling during immersion. Additionally, part of the coating dissolved as indicated by weight loss upon drying and residues remaining upon evaporation of the immersion liquids.

- Biodegradation tests showed that the coating biodegrades readily in moist soil. It appeared to biodegrade faster in Hanford soil than in Barnwell soil. The amount of biodegradation measured by monitoring CO_2 produced from microbial respiration was confirmed by weight loss measurements on the biodegraded coating samples. The biodegradation is probably caused by bacteria and not fungi as indicated by the results of ASTM tests G21 and G22.

- Radiation stability testing showed that the coating generates gas, primarily hydrogen with lesser amounts of carbon dioxide, hydrocarbons, and carbon monoxide, with a G-value for total gas of approximately 1.3. It also consumes oxygen. Radiation improves the physical characteristics of strength and toughness of the coating and may enhance the resistance to chemical and biodegradative attack. In sum, the 1146 coating showed no meaningful radiation damage, except for gas generation, to doses of 10^9 rad. Gas generation becomes significant (i.e., the volume of gas generated is approximately equal to the coating volume) above 10^8 rad.

- Thermal stability testing showed that ALARA 1146 is only moderately flammable and is not greatly affected by heat up to 100°C for short periods of time. It scavanges oxygen from the air even at room temperature. Some pyrolysis, as indicated by the generation of hydrocarbons, occurs at temperatures as low as 100°C.

4.2.2 Testing the Strippable Coating From TMI-2

4.2.2.1 TMI-2 Coating Samples

Samples of strippable coating from the reactor building Gross Decontamination Experiment were sent to BNL for characterization. The TMI coating samples obtained consisted of five pieces individually packaged in numbered plastic bags. A summary of data on the coating pieces received from TMI including the bag number, weight, dimensions, and a gamma-activity measurement at approximately 5 cm is listed in Table 4.5.

The large differences in the measured gamma activities listed in Table 4.5 agree with previous observations[1,8] that the contamination in the reactor building is distributed inhomogeneously. There were no obvious differences between coating specimens from different bags, such as differing amounts of debris incorporated into coating samples with grossly differing activities. In fact, since the surface to which the ALARA 1146 coating was applied had been flushed with water prior to application, very little debris was observed on any of the coating pieces. Due to the inhomogeniety of the activities on the different coating pieces, there is no way of knowing whether this batch is representative of the average activity for the strippable coating radwaste produced in the Gross Decontamination Experiment or whether it may provide an estimate for expected activities on strippable coating radwaste which may be produced in decontaminating other parts of the reactor building.

Table 4.5

Strippable Coating Samples from the TMI-2 Reactor Building
Gross Decontamination Testing
(The bag numbers were on the plastic bags containing
the individual coating samples as received)

Bag No.	Sample Weight (g)	Sample Width (cm)	Sample Length (cm)	Activity at 5 cm (mrem/h)
50	7.77	9.0	17.5	3.5
51	6.95	9.0	19.0	230
52	4.50	9.8	15.2	150
162	5.94	8.5	16.5	250
163	4.13	8.5	11.5	4.0
Total	29.29			

The TMI coatings were tested for radionuclide leaching and for biodegradation. The radionuclide leach tests, in which Cs-134,137 were monitored, were performed to compare results from these decontamination coatings in which the contamination was removed from a surface with the scoping test results in which the radionuclides were added to the liquid coating in an aqueous spike. Following this leach testing, the leachate and the leached coating specimen were sent to a commercial analysis laboratory (EAL Corporation*) for analysis for Sr and Pu content. Biodegradation tests in soil were run on these coatings to clarify the differences in biodegradation that were suggested by the scoping biodegradation tests.

4.2.2.2 Leach Testing and Radionuclide Analysis on TMI-2 Coating

The TMI coating sample from bag 162 (Table 4.5) was leached in deionized water for 43 days and then the leachate and the leached coating residue were sent to EAL Corp., a commercial analysis laboratory, for further radionuclide analysis. Figure 4.9 shows the time intervals at which the leachate was changed and the cumulative Cs-137,134 activity leached from the sample. Each leachate volume was counted for activity in a gamma spectrometer. The activity was quantified using identical volumes with known activities of the specific radionuclides in aqueous solution. No gamma emitters other than Cs-134,137 and the Ba-137m daughter were unambiguously detected in the leachate. No Cs remained in leached coating sample and no other gamma emitters were unambiguously detected in the leached coating sample. Sample 162 had 140 cm^2 surface (one side); the leachant volume used was 70 cm^3 for a volume-to-surface ratio of 0.5 cm. (The use of a ratio of 10 as the IAEA recommends was not practical because of the large volume of leachate that

*EAL Corporation, Richmond, CA, performed radiochemical analyses for Sr-90 and Pu-239,240 on both the leachate and the leached coating residue.

would be produced. Since the total amount of leachate was to be sent out for analysis, the total volume was limited to less than 2 L. After gamma counting, the leachate was acidified to about 2 N with nitric acid and combined with previous specimens in a polyethylene bottle. The acidified leachate volume plus the wash (performed at EAL Corp) from the polyethylene bottle in which the leach coating was shipped after the coating was removed for analysis totaled 1370 mL. The total volume of deionized water leachant was 1050 mL for a cumulative volume-to-surface ratio of 7.5 cm.

Table 4.6 lists the data on the activities of Cs-134,137, Sr-90, and Pu-239,240 from the leachate and residue of the TMI coating sample from bag 162.

The activities listed in Table 4.6 are for the TMI sample with the highest activity, according to the gamma counting results listed in Table 4.5. Assuming that the Cs and Sr activities in the five coating pieces are distributed proportionally according to the totals in Table 4.6 and the gamma counts in Table 4.5, the total activity of the five coating pieces considered as a batch can be calculated.

$$\text{Total Cs-137 activity} = \frac{52\ \mu\text{Ci Cs-137}}{250\ \text{mrem}}\,(3.5 + 230 + 250 + 150 + 4.0)\ \text{mrem}$$

$$\text{Total Cs-137 activity} = 130\ \mu\text{Ci}$$

$$\text{Total Sr-90 activity} = \frac{2.28\ \mu\text{Ci Sr-90}}{250\ \text{mrem}}\,(3.5 + 230 + 150 + 250 + 4.0)\ \text{mrem}$$

$$\text{Total Sr-90 activity} = 5.8\ \mu\text{Ci}$$

These estimated total activities along with the corresponding specific and volumetric activities are listed in Table 4.7.

The limits for Class A radwaste in 10 CFR Part 61, Table 2, are 1 $\mu Ci/cm^3$ Cs-137 and 0.04 $\mu Ci/cm^3$ Sr-90. The estimated volumetric activities in Table 4.7 exceed these limits for both of these radionuclides. If these activities approximate those of the strippable coating, radwaste from GDF, it would be Class B under 10 CFR Part 61 and hence would require stabilization.

The data in Table 4.6 and 4.7 were used to calculate the expected dose that a 55-gal drum of the TMI coating considered as a batch could be expected to receive. This calculation, which follows the procedures presented in Reference 45, is shown in Appendix C. The cumulative dose is estimated to be approximately 2 Mrad in 300 yrs.

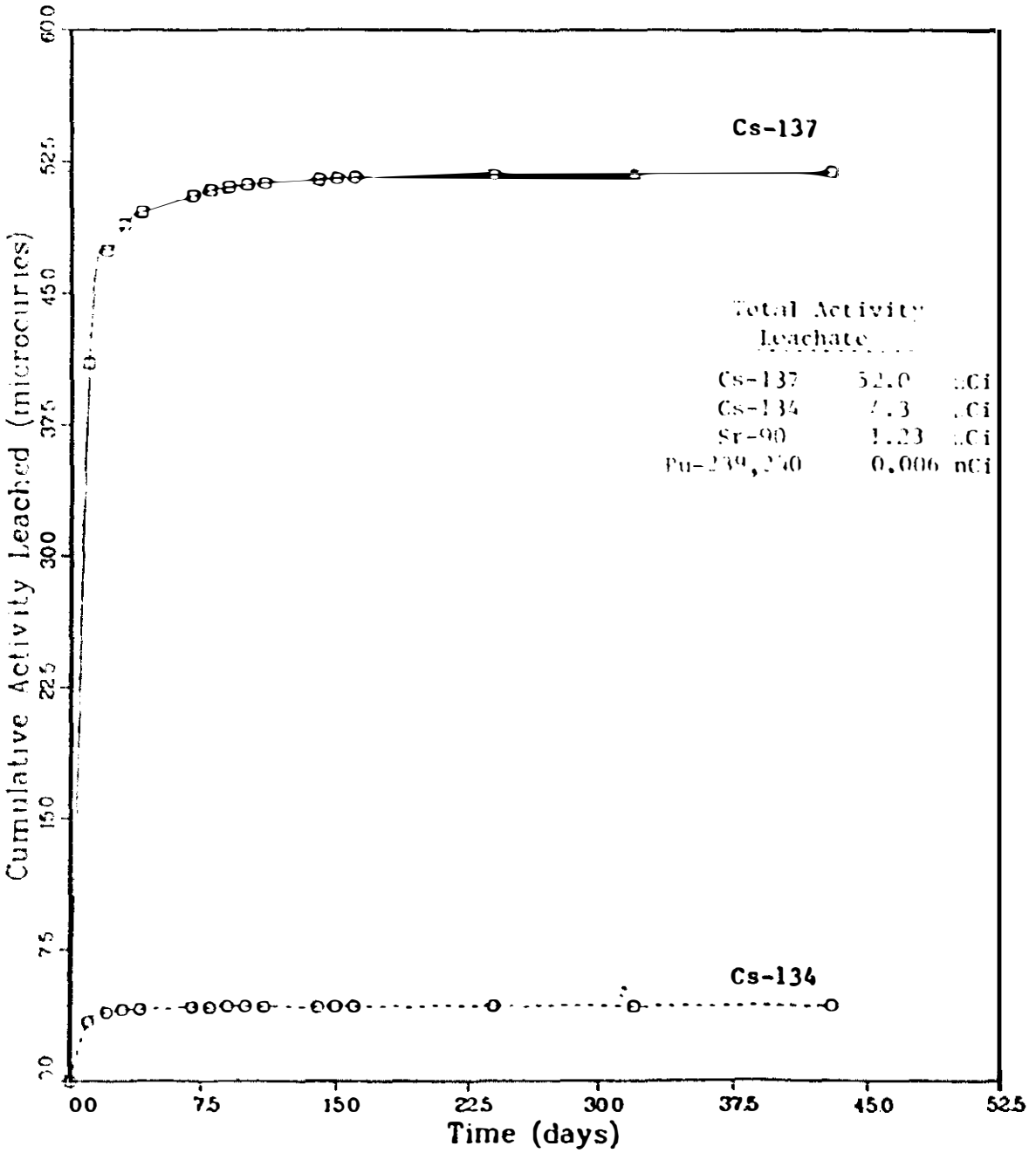

Figure 4.9 Leach test results for a TMI strippable coating sample. Cumulative leached activity for Cs-137 and Cs-134 are plotted vs time. Data points indicate leachate change intervals. The total leached activities for Cs-137, Cs-134, Sr-90 and Pu-239,240 are listed.

Table 4.6

Activity Distribution[a] Between Leachate and Coating for a Sample Coating[b] From the TMI-2 Gross Decontamination Experiment

Radionuclide	Activity in Leachate	Activity Remaining in Leached Coating	Total Activity in Sample	Specific Activity in Sample[c]	Volumetric Activity in Sample[d]
Cs-134	4.3 uCi	nil	4.3 uCi	0.73 uCi/g	0.91 uCi/cm^3
Cs-137	52.0 uCi	nil	52.0 uCi	8.7 uCi/g	10.8 uCi/cm^3
Sr-90	1.23 uCi	1.05 uCi	2.28 uCi	0.38 uCi/g	0.47 uCi/cm^3
Pu-239,240	0.006 nCi	0.02 nCi	0.026 nCi	0.004 nCi/g	0.005 nCi/cm^3

[a]The Cs-134,137 activities were determined by gamma spectroscopy at BNL. The Sr and Pu activities were determined by EAL Corp. Richmond, CA.
[b]The sample came from bag 162 as listed in Table 4.5. The coating is ALARA 1146 Decon, product of Imperial Professional Coatings Corp., New Orleans, LA.
[c]Total activity divided by the sample weight of 5.94 g.
[d]Specific activity in units of uCi/g multiplied by the density of the set coating in g/cm^3. The density used is 1.24 g/cm^3 as calculated from the Physical Properties fact sheet for ALARA 1146.

Table 4.7

The Estimated Activities[a] of Cs-137 and Sr-90 in the Five TMI Coating Pieces Considered as a Batch

Radionuclide	Total Activity (Estimated)	Specific Activity[b]	Volumetric Activity[c]
Cs-137	130 μCi	4.5 μCi/g	5.6 μCi/cm^3
Sr-90	5.8 μCi	0.20 μCi/g	0.25 μCi/cm^3

[a]Estimated assuming a proportional distribution in the five TMI samples listed in Table 4.5 to the measured activities in sample 162 as listed in Table 4.6.
[b]Total activity divided by the batch weight of 29.29 g.
[c]Specific activity in μCi/g times the density (1.24 g/cm^3) of the ALARA 1146 coating as calculated from the Physical Properties fact sheet.

4.2.2.3 Biodegradation Testing of TMI Strippable Coating

Biodegradation of TMI strippable coating samples in backfill soils from the Barnwell and Hanford land burial sites was monitored by CO_2 generation. The experimental procedures using the special flasks[36] were the same as those in the scoping tests of Section 4.1.5. The sample size in these tests was greatly reduced to assure uniform mixing of samples with soil and equal sample surface areas exposed to direct soil contact in each test. Each TMI coating sample consisted of 10 pieces, each 1 cm^2, cut from the specimen in bag 52 (Table 4.5). Samples were mixed with 100 g of moist soil from the Barnwell and Hanford land burial sites and loaded into the special flasks. Soil controls of 100 g of each of the same soils and a sample control moistened with 1-mL water were also prepared. The soil moisture contents were raised to 12.5% for the Hanford soil and to 9.8% for the Barnwell soil from as received values of 8.9% and 4.2%, respectively.[46] (The lower moisture holding ability of the Barnwell soil appeared to be a result of its coarse texture and sandy characteristics.)

Results of the TMI biodegradation tests are listed in Table 4.8 and shown in Figures 4.10-4.12. Figure 4.10 shows the CO_2 generation (mg) vs time from the samples in Barnwell, B, and Hanford, H, soils. Figures 4.11 and 4.12 show the CO_2 generation expressed as percent carbon in the sample. (The value of 52.3% C measured for the purchased 1146 coating used in the scoping tests was also used in these tests. The actual TMI coating samples were not analyzed for total carbon.) The shaded area in these figures represent the range of probable biodegradation based on the data, which is the lower boundary, and the curve produced by dividing each data point value by

0.6, which defines the upper boundary. This probable range is based on observations[39,40] that as little as 60% of the carbon in a material undergoing biodegradation may be evolved as CO_2, the remainder being incorporated into increased microbe mass or excreted as metabolic waste products other than CO_2. Table 4.8 summarizes the data for the biodegradation experiment monitored by CO_2 generation plus data on the weight loss that occurred during the 208 days of the test. Biodegradation measured by weight loss falls in the range of expected biodegradation from the CO_2 measurements.

Table 4.8

Biodegradation Test Results for Strippable Coating Samples From the TMI-2 Reactor Building Gross Decontamination Experiment

Sample	Initial Weight W_o (g)	Final Weight W_f (g)	Biogenically Generated CO_2 (mg)	%C[a]	%C[b] 0.6	$\frac{W_o - W_f}{W_o} \times 100$
B	0.544	0.526	48.62	4.65	7.75	3.31
H	0.554	0.505	94.86	8.92	14.87	8.84
Control	0.554	0.546	None	0	0	1.44

[a]From the biogenically generated CO_2 data and the measured 52.3% C content of the coating used in the scoping tests, Section 4.1.5.
[b]The estimated upper limit of biodegradation based on CO_2 generation and observations, References 39 and 40, that as little as 60% of the available carbon may be evolved as CO_2 during biodegradation.
[c]Percent biodegradation from the initial and final weight following 208 days of biodegradation in soil at 20-24°C.

These results support the results obtained in the scoping biodegradation tests that the ALARA 1146 strippable coating used at TMI biodegrades readily in soil. The rate of biodegradation is initially rapid and decreases with time. The biodegradation of this material proceeds faster in Hanford soil than in Barnwell soil under the conditions of these tests. The differences may be due to the greater moisture holding ability of Hanford soil relative to Barnwell soil, or it may be related to the fact that Hanford soil contains more soluble ions,[46] and therefore, presumably more of the required trace minerals needed for microbe growth, or it may be a combination of these plus other factors. The biodegradability of the ALARA 1146 coating as reflected by these test results shows that, under 10 CFR Part 61, Class B or C strippable coating radwaste would have to be isolated from the burial trench environment to prevent microbe attack.

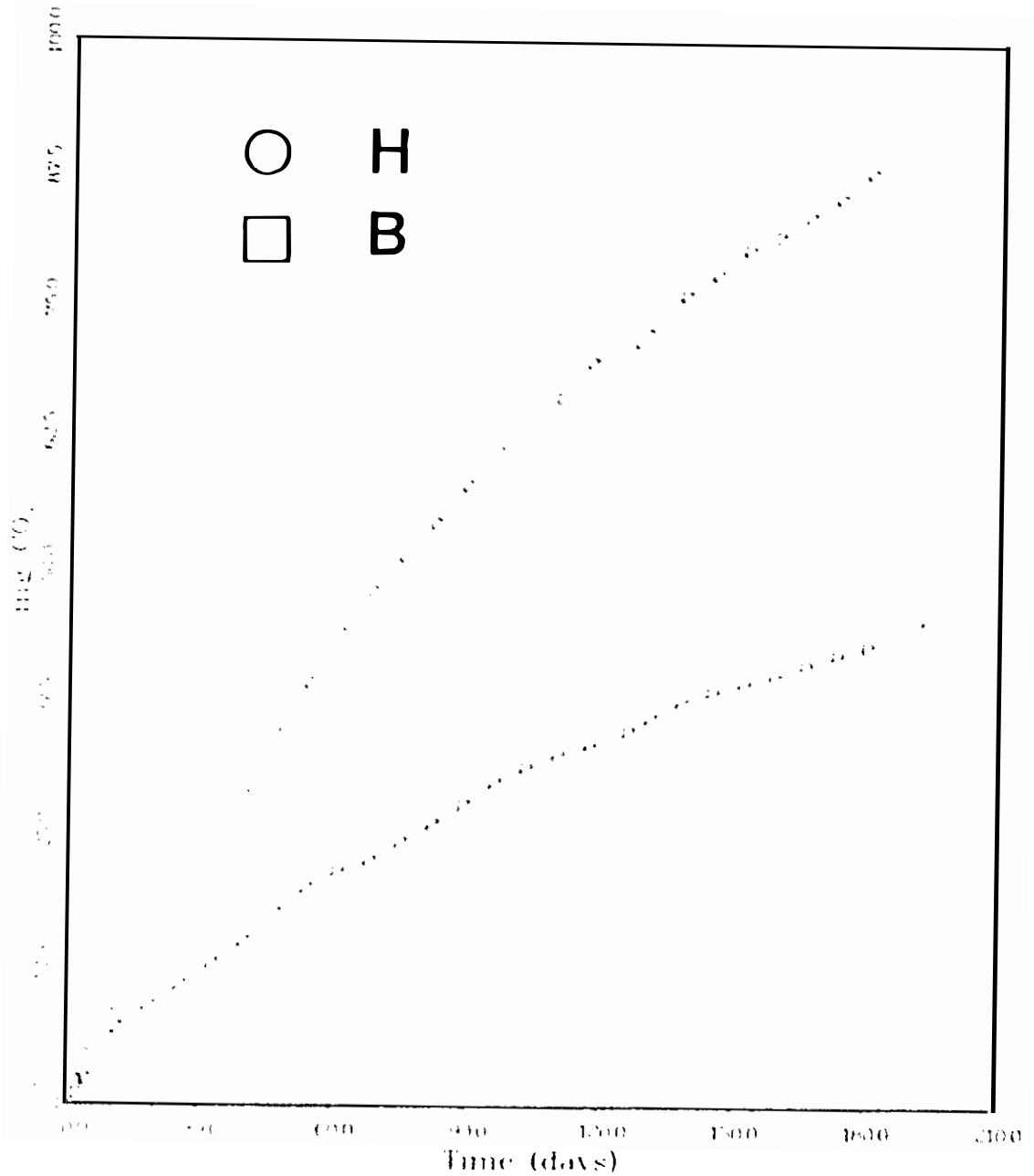

Figure 4.10 Biodegradative CO_2 gas evolution from TMI strippable coating material in Barnwell and Hanford soils.

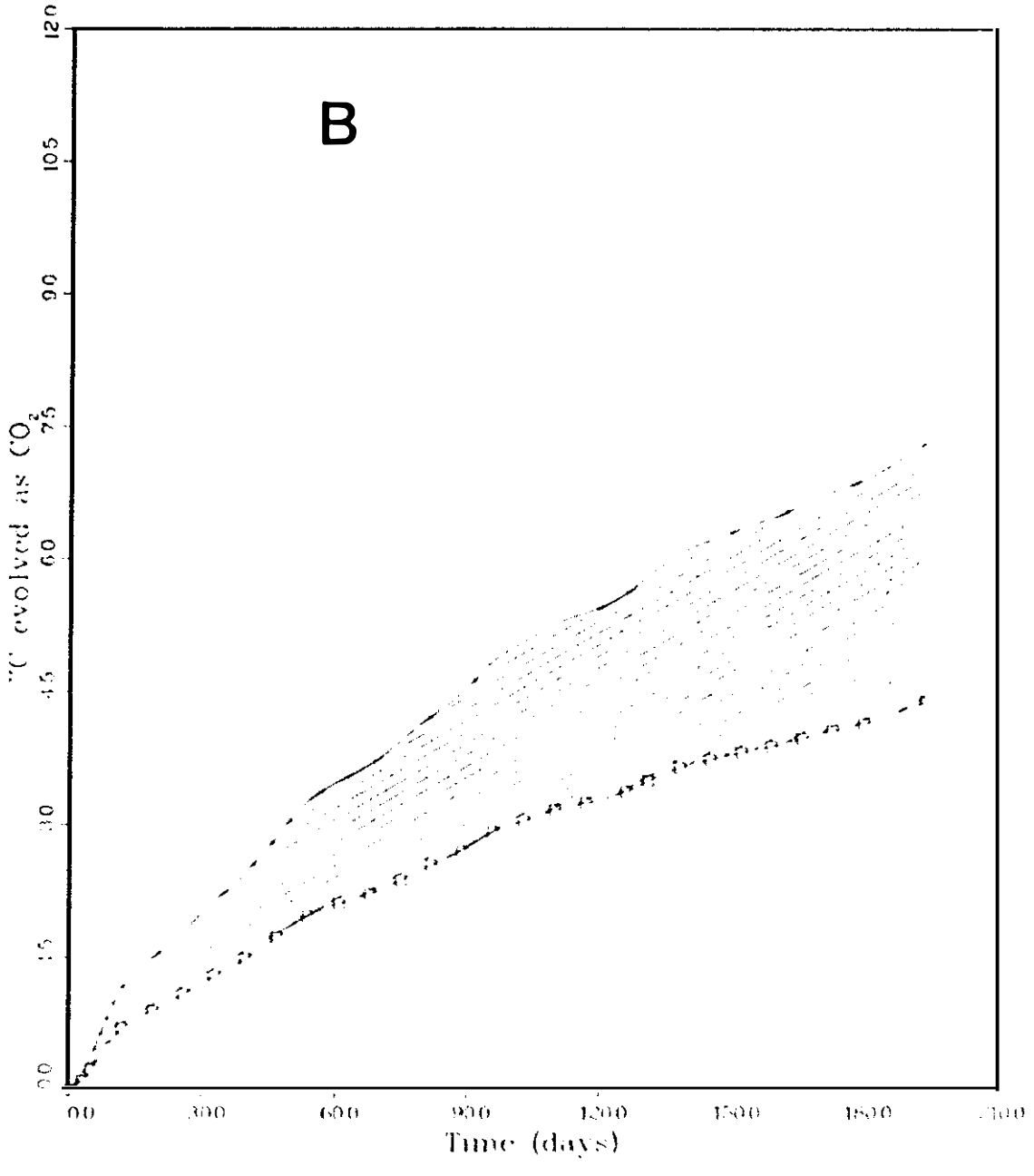

Figure 4.11 Probable range of biodegradation of TMI strippable coating in Barnwell soil.

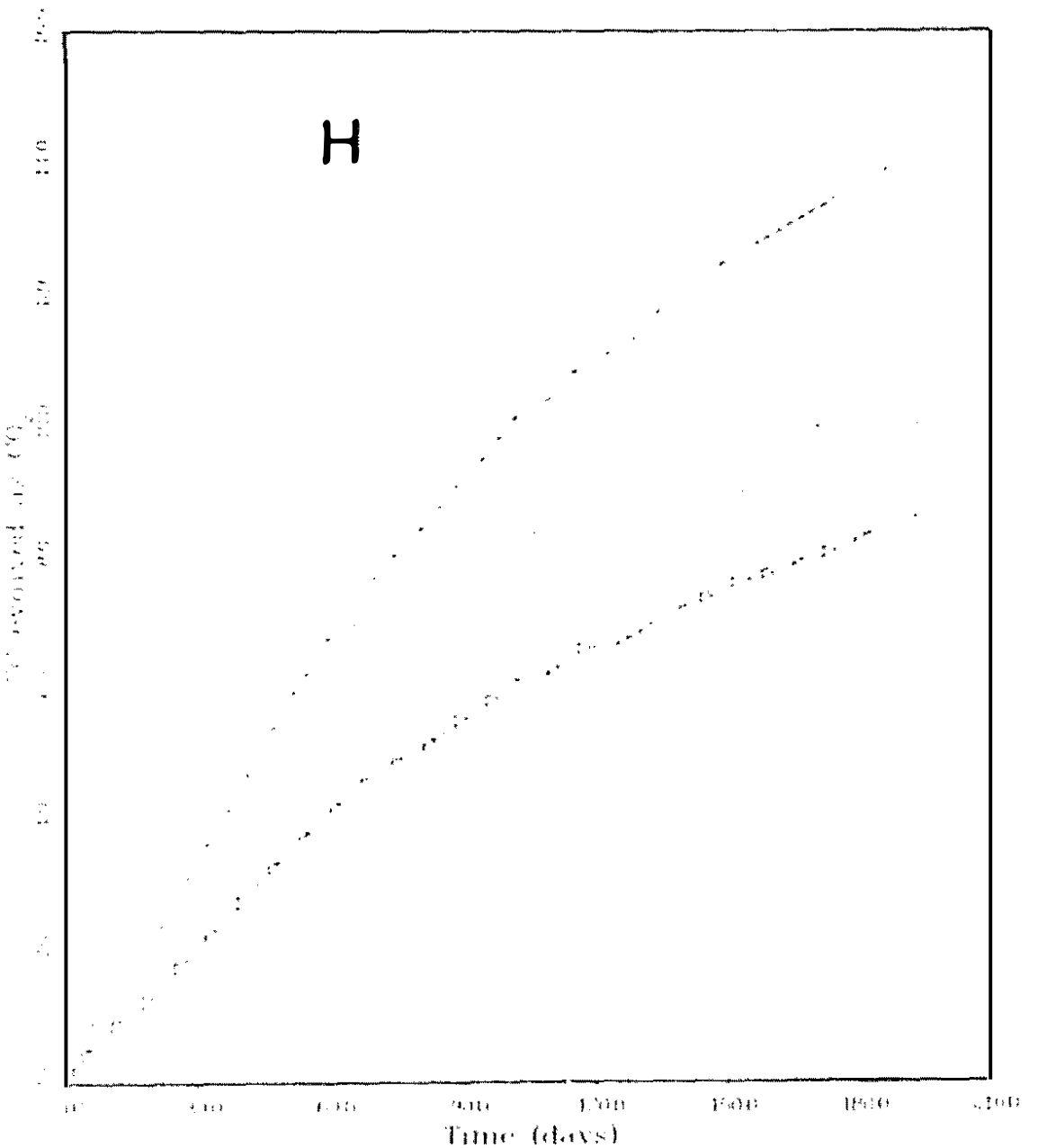

Figure 4.12 Probable range of biodegradation of TMI strippable coating in Hanford soil.

4.2.2.4 Summary of TMI Strippable Coating Sample Tests

Leach testing of the sample of ALARA 1146 coating from the TMI-2 GDE showed that Cs leaches from the coating immediately upon contact with water. The Sr activity was portioned more or less equally between the coating and the leachate whereas, only about a quarter of the Pu activity was leached. These results are consistent with the scoping radionuclide leach testing results and added the knowledge that Pu tends to be retained on the coating, at least for the low activity levels measured here.

The TMI coating samples biodegraded readily in Barnwell and Hanford land burial backfill soils. This susceptibility to microbe attack dictates that coating radwaste classed as B or C under 10 CFR Part 61 would require stabilization.

Decontamination using the ALARA 1146 strippable coating was shown to be effective (DF about 100) on the epoxy painted floor of the TMI-2 reactor building.[9,10] Its effectiveness in removing debris following low pressure water flushing was the best of the decontamination methods tested. Use of the strippable coating generated about 0.64 ft^3 of solid compactible waste per 100 ft^2 of application. Compaction would allow coating waste from decontaminating 2000-2500 ft^2 to be disposed of in one 55-gal drum.

The contamination taken up by the coating application in the decontamination test produced waste which, if compacted to fill a 55-gal drum, would have a contact radiation reading of less than 5 rem/h.[10] Equation 2.1 and the appropriate C.F.[22,23] for a compacted 55-gal drum of Normal Unit 2 Radwaste allows calculation of the activity contained in the hypothetical drum.

$$\text{Activity} = (0.557\ \text{mCi h/mrem})\ (5000\ \text{mrem/h})$$

$$\text{Activity} = 2800\ \text{mCi}$$

This activity in a 55-gal drum (210,000 cm^3) results in an activity density of 13.3 $\mu Ci/cm^3$, or 10.7 $\mu Ci/g$ based on the 1.24 g/cm^3 density for the coating. This activity is consistent with, though higher than, the measured activities in Tables 4.6 and 4.7.

The measured activity on the strippable coating radwaste used for decontamination in the GDE provides evidence that such decontamination applications may be expected to result in Class B radwaste order 10 CFR Part 61. As of this writing, it is not known whether the coating will be used for decontamination activities beyond its evaluation in the GDE testing. It is being used as a protective coating over cleaned surfaces in the upper part of the reactor building including the 305 and 347-ft elevations and the polar crane. Class B strippable coating radwaste would require stabilization under 10 CFR

Part 61. Imperial Professional Coating has experimental solidification systems* for this purpose; however, these solidification systems have not yet been evaluated for compliance with the stability provisions of 10 CFR Part 61.

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*Personal communication between J. Adams (BNL) and H. Lomasney (Imperial), July 30, 1982, "Solidification Systems for Strippable Coatings."

5. REFERENCES

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APPENDIX A

10 CFR PART 61.55: RADIOACTIVE WASTE CLASSIFICATION FOR DISPOSAL IN SHALLOW LAND BURIAL

disposal site before they leave the site boundary.

§ 61.54 Alternative requirements for design and operations.

The Commission may, upon request or on its own initiative, authorize provisions other than those set forth in §§ 61.51 through 61.53 for the segregation and disposal of waste and for the design and operation of a land disposal facility on a specific basis, if it finds reasonable assurance of compliance with the performance objectives of Subpart C of this part.

§ 61.55 Waste classification.

(a) Classification of waste for near surface disposal.

(1) *Considerations.* Determination of the classification of radioactive waste involves two considerations. First, consideration must be given to the concentration of long-lived radionuclides (and their shorter-lived precursors) whose potential hazard will persist long after such precautions as institutional controls, improved waste form, and deeper disposal have ceased to be effective. These precautions delay the time when long-lived radionuclides could cause exposures. In addition, the magnitude of the potential dose is limited by the concentration and availability of the radionuclide at the time of exposure. Second, consideration must be given to the concentration of shorter-lived radionuclides for which requirements on institutional controls, waste form, and disposal methods are effective.

(2) *Classes of waste.* (i) Class A waste is waste that is usually segregated from other waste classes at the disposal site. The physical form and characteristics of Class A waste must meet the minimum requirements set forth in § 61.56(a). If Class A waste also meets the stability requirements set forth in § 61.56(b), it is not necessary to segregate the waste for disposal.

(ii) Class B waste is waste that must meet more rigorous requirements on waste form to ensure stability after disposal. The physical form and characteristics of Class B waste must meet both the minimum and stability requirements set forth in § 61.56.

(iii) Class C waste is waste that not only must meet more rigorous requirements on waste form to ensure stability but also requires additional measures at the disposal facility to protect against inadvertent intrusion. The physical form and characteristics of Class C waste must meet both the minimum and stability requirements set forth in § 61.56.

(iv) Waste that is not generally acceptable for near-surface disposal is waste for which waste form and disposal methods must be different, and in general more stringent, than those specified for Class C waste. In the absence of specific requirements in this part, proposals for disposal of this waste may be submitted to the Commission for approval, pursuant to § 61.58 of this part.

(3) Classification determined by long-lived radionuclides. If radioactive waste contains only radionuclides listed in Table 1, classification shall be determined as follows.

(i) If the concentration does not exceed 0.1 times the value in Table 1, the waste is Class A.

(ii) If the concentration exceeds 0.1 times the value in Table 1 but does not exceed the value in Table 1, the waste is Class C.

(iii) If the concentration exceeds the value in Table 1, the waste is not generally acceptable for near-surface disposal.

(iv) For wastes containing mixtures of radionuclides listed in Table 1, the total concentration shall be determined by the sum of fractions rule described in paragraph (a)(7) of this section.

TABLE 1

Radionuclide	Concentration curies per cubic meter
C-14	8
C-14 in activated metal	80
Ni-59 in activated metal	220
Nb-94 in activated metal	0.2
Tc-99	3
I-129	0.08
Alpha emitting transuranic nuclides with half life greater than five years	[1] 100
Pu-241	[1] 3,500
Cm-242	[1] 20,000

[1] Units are nanocuries per gram.

(4) Classification determined by short-lived radionuclides. If radioactive waste does not contain any of the radionuclides listed in Table 1, classification shall be determined based on the concentrations shown in Table 2. However, as specified in paragraph (a)(6) of this section, if radioactive waste does not contain any nuclides listed in either Table 1 or 2, it is Class A.

(i) If the concentration does not exceed the value in Column 1, the waste is Class A.

(ii) If the concentration exceeds the value in Column 1, but does not exceed the value in Column 2, the waste is Class B.

(iii) If the concentration exceeds the value in Column 2, but does not exceed the value in Column 3, the waste is Class C.

(iv) If the concentration exceeds the value in Column 3, the waste is not generally acceptable for near-surface disposal.

(v) For wastes containing mixtures of the nuclides listed in Table 2, the total concentration shall be determined by the sum of fractions rule described in paragraph (a)(7) of this section.

TABLE 2

Radionuclide	Concentration curies per cubic meter		
	Col. 1	Col. 2	Col. 3
Total of all nuclides with less than 5 year half life	700	[1]	[1]
H-3	40	[1]	[1]
Co-60	700	[1]	[1]
Ni-63	3.5	70	100
Ni-63 in activated metal	35	700	7000
Sr-90	0.04	150	7000
Cs-137	1	44	4600

[1] There are no limits established for these radionuclides in Class B or C wastes. Practical considerations such as the effects of external radiation and internal heat generation on transportation, handling, and disposal will limit the concentrations for these wastes. These wastes shall be Class B unless the concentrations of other nuclides in Table 2 determine the waste to be Class C independent of these nuclides.

(5) Classification determined by both long- and short-lived radionuclides. If radioactive waste contains a mixture of radionuclides, some of which are listed in Table 1, and some of which are listed in Table 2, classification shall be determined as follows:

(i) If the concentration of a nuclide listed in Table 1 does not exceed 0.1 times the value listed in Table 1, the class shall be that determined by the concentration of nuclides listed in Table 2.

(ii) If the concentration of a nuclide listed in Table 1 exceeds 0.1 times the value listed in Table 1 but does not exceed the value in Table 1, the waste shall be Class C, provided the concentration of nuclides listed in Table 2 does not exceed the value shown in Column 3 of Table 2.

(6) Classification of wastes with radionuclides other than those listed in Tables 1 and 2. If radioactive waste does not contain any nuclides listed in either Table 1 or 2, it is Class A.

(7) The sum of the fractions rule for mixtures of radionuclides. For determining classification for waste that contains a mixture of radionuclides, it is necessary to determine the sum of fractions by dividing each nuclide's concentration by the appropriate limit and adding the resulting values. The appropriate limits must all be taken from the same column of the same table. The sum of the fractions for the column must be less than 1.0 if the waste class is to be determined by that column. Example: A waste contains Sr-90 in a

concentration of 50 Ci/m³ and Cs-137 in a concentration of 22 Ci/m³. Since the concentrations both exceed the values in Column 1, Table 2, they must be compared to Column 2 values. For Sr-90 fraction 50/150=0.33; for Cs-137 fraction, 22/44=0.5; the sum of the fractions=0.83. Since the sum is less than 1.0, the waste is Class B.

(8) *Determination of concentrations in wastes.* The concentration of a radionuclide may be determined by indirect methods such as use of scaling factors which relate the inferred concentration of one radionuclide to another that is measured, or radionuclide material accountability, if there is reasonable assurance that the indirect methods can be correlated with actual measurements. The concentration of a radionuclide may be averaged over the volume of the waste, or weight of the waste if the units are expressed as nanocuries per gram.

§ 61.56 Waste characteristics.

(a) The following requirements are minimum requirements for all classes of waste and are intended to facilitate handling at the disposal site and provide protection of health and safety of personnel at the disposal site.

(1) Waste must not be packaged for disposal in cardboard or fiberboard boxes.

(2) Liquid waste must be solidified or packaged in sufficient absorbent material to absorb twice the volume of the liquid.

(3) Solid waste containing liquid shall contain as little free standing and noncorrosive liquid as is reasonably achievable, but in no case shall the liquid exceed 1% of the volume.

(4) Waste must not be readily capable of detonation or of explosive decomposition or reaction at normal pressures and temperatures, or of explosive reaction with water.

(5) Waste must not contain, or be capable of generating, quantities of toxic gases, vapors, or fumes harmful to persons transporting, handling, or disposing of the waste. This does not apply to radioactive gaseous waste packaged in accordance with paragraph (a)(7) of this section.

(6) Waste must not be pyrophoric. Pyrophoric materials contained in waste shall be treated, prepared, and packaged to be nonflammable.

(7) Waste in a gaseous form must be packaged at a pressure that does not exceed 1.5 atmospheres at 20°C. Total activity must not exceed 100 curies per container.

(8) Waste containing hazardous, biological, pathogenic, or infectious material must be treated to reduce to the maximum extent practicable the potential hazard from the non-radiological materials.

(b) The requirements in this section are intended to provide stability of the waste. Stability is intended to ensure that the waste does not structurally degrade and affect overall stability of the site through slumping, collapse, or other failure of the disposal unit and thereby lead to water infiltration. Stability is also a factor in limiting exposure to an inadvertent intruder, since it provides a recognizable and nondispersible waste.

(1) Waste must have structural stability. A structurally stable waste form will generally maintain its physical dimensions and its form, under the expected disposal conditions such as weight of overburden and compaction equipment, the presence of moisture, and microbial activity, and internal factors such as radiation effects and chemical changes. Structural stability can be provided by the waste form itself, processing the waste to a stable form, or placing the waste in a disposal container or structure that provides stability after disposal.

(2) Notwithstanding the provisions in §§ 61.56(a) (2) and (3), liquid wastes, or wastes containing liquid, must be converted into a form that contains as little free standing and noncorrosive liquid as is reasonably achievable, but in no case shall the liquid exceed 1% of the volume of the waste when the waste is in a disposal container designed to ensure stability, or 0.5% of the volume of the waste for waste processed to a stable form.

(3) Void spaces within the waste and between the waste and its package must be reduced to the extent practicable.

§ 61.57 Labeling.

Each package of waste must be clearly labeled to identify whether it is Class A waste, Class B waste, or class C waste in accordance with § 61.55.

§ 61.58 Alternative requirements for waste classification and characteristics.

The Commission may, upon request or on its own initiative, authorize other provisions for the classification and characteristics of waste on a specific basis, if, after evaluation, of the specific characteristics of the waste, disposal site, and method of disposal, it finds reasonable assurance of compliance with the performance objectives in Subpart C of this part.

§ 61.59 Institutional requirements.

(a) *Land ownership.* Disposal of radioactive waste received from other persons may be permitted only on land owned in fee by the Federal or a State government.

(b) *Institutional control.* The land owner or custodial agency shall carry out an institutional control program to physically control access to the disposal site following transfer of control of the disposal site from the disposal site operator. The institutional control program must also include, but not be limited to, carrying out an environmental monitoring program at the disposal site, periodic surveillance, minor custodial care, and other requirements as determined by the Commission; and administration of funds to cover the costs for these activities. The period of institutional controls will be determined by the Commission, but institutional controls may not be relied upon for more than 100 years following transfer of control of the disposal site to the owner.

Subpart E—Financial Assurances

§ 61.61 Applicant qualifications and assurances.

Each applicant shall show that it either possesses the necessary funds or has reasonable assurance of obtaining the necessary funds, or by a combination of the two, to cover the estimated costs of conducting all licensed activities over the planned operating life of the project, including costs of construction and disposal.

§ 61.62 Funding for disposal site closure and stabilization.

(a) The applicant shall provide assurance that sufficient funds will be available to carry out disposal site closure and stabilization, including: (1) Decontamination or dismantlement of land disposal facility structures, and (2) closure and stabilization of the disposal site so that following transfer of the disposal site to the site owner, the need for ongoing active maintenance is eliminated to the extent practicable and only minor custodial care, surveillance, and monitoring are required. These assurances shall be based on Commission-approved cost estimates reflecting the Commission-approved plan for disposal site closure and stabilization. The applicant's cost estimates must take into account total capital costs that would be incurred if an independent contractor were hired to perform the closure and stabilization work.

(b) In order to avoid unnecessary duplication and expense, the Commission will accept financial sureties that have been consolidated with earmarked financial or surety arrangements established to meet

APPENDIX B

TECHNICAL DATA SHEET FOR ALARA 1146 DECON STRIPPABLE COATING

IMPERIAL
PROFESSIONAL COATINGS

ALARA 1146 DECON (TM)

DESCRIPTION: One package, water borne, strippable coating.

RECOMMENDED USES: Decontaminating radioactive polluted areas. Apply over contaminated steel, concrete, wood, aluminum, or painted surfaces via spray or roll. Can be used for sealing (fixing) potential airborne contaminants, for protection of personnel from "smearable" contaminants. Also useful with beta emitters for "shielding" purposes. Because this product produces an easily compacted (solid) waste, the workload on radwaste processing facilities can be substantially reduced.

PRODUCT DESIGN FUNCTION:

1. Apply material over contaminated surface. While material is wet it attracts, absorbs and chemically binds heavy metal isotopes. During application the coating migrates into micro voids of surface to contact contaminants. Upon cure, the product mechanically locks the contaminants into a polymer matrix. Stripping the film effectively cleans the substrate and produces a solid waste.
2. Apply material over clean surface to provide protective layer against future contamination.
3. Apply material over previously contaminated surfaces while still wet to inhibit potential airborne contamination during dry-out.

CAUTION: *DO NOT FREEZE.* Consult Imperial for specific instructions concerning high humidity applications. Refer to chart on reverse.

PHYSICAL PROPERTIES:

TYPE	Vinyl
COLOR	Yellow
FLASH POINT	N/A
NUMBER OF COMPONENTS	One
POT LIFE @ 75°F	N/A
DRYING TIME @ 75°F AND 75% RH	
A. TO TOUCH (FOOT TRAFFIC)	24 hours
B. TO FULL CURE	2 days
SHELF LIFE	4 months
NET WEIGHT - 1's	9.0 pounds
5's	45.0 pounds
PERCENT VOLUME SOLIDS	42%
RECOMMENDED DRY FILM THICKNESS	20-30 mils
THEORETICAL COVERAGE AT 25.0 MILS	27 sq ft/gal
THINNER	Water
TEMPERATURE RESISTANCE	120°F
FIRE DATA (ASTM-E-84-77) @ 20-25 MILS	20 (flame), 25 (fuel), 25 (smoke)
CHARCOAL FILTER	99.3% efficiency after 2 hrs. @ 40FPM face loading (methyl iodide)
DECONTAMINATION FACTOR	30-100 (varies with substrate)

APPLICATION EQUIPMENT RECOMMENDATIONS:

1. Airless - Standard industrial spray equipment (stainless steel parts desired) such as Graco or Binks, using a 30:1 pump ratio with 50-60 psi inbound pressure and a .021" to .025" fluid tip with reversible cleaning head
2. Roller 1/2" lambs wool with phenolic core.

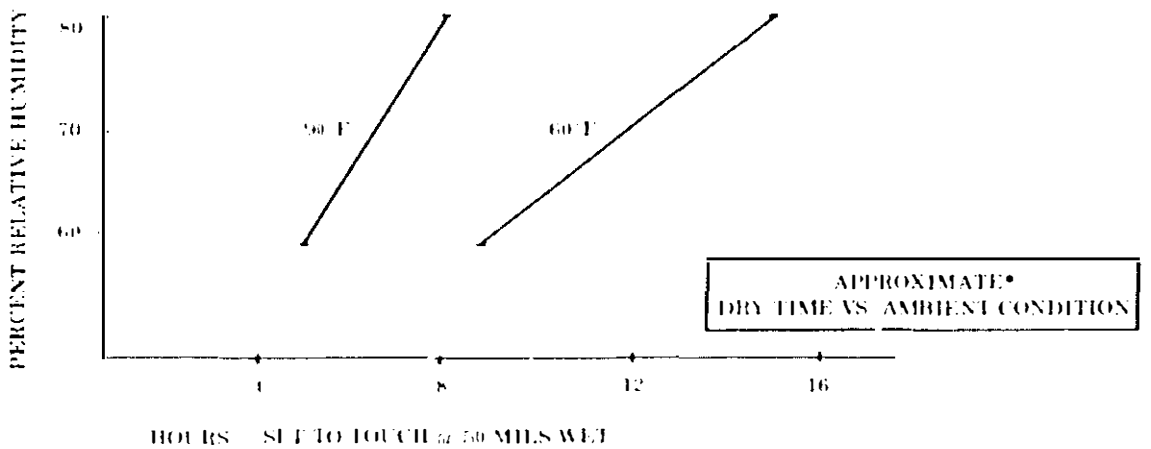
SAFETY EQUIPMENT RECOMMENDATIONS: #1146 is free of solvents and toxic materials. A particulate (gauze) mask is recommended to prevent inhalation of overspray. Consult Material Safety Data Sheet.

APPLICATION PROCEDURE:

1. Flush all equipment with fresh water prior to use.
2. Stir material thoroughly before and throughout application.
3. Do not thin except for workability, and then with no more than 10% by volume with fresh water.
4. Flush all equipment immediately after use with fresh water. Use butyl cellosolve or similar solvent for final cleaning.

STORAGE CONDITIONS:

40° to 90°F. DO NOT FREEZE.



*Dry time varies with air flow rate, temperature, relative humidity, film thickness and substrate conditions

NOTE *The technical data furnished is true and accurate to the best of our knowledge however, no guarantee of accuracy is given or implied. We guarantee our products to conform to Imperial Quality Control Standards. We assume no responsibilities for its handling, use, storage, the results obtained or any injury or damage resulting from its use.*

2-1-80

IMPERIAL PROFESSIONAL COATINGS

APPENDIX C

CUMULATIVE DOSE CALCULATIONS FOR THE ALARA 1146 STRIPPABLE COATING SAMPLES FROM THE TMI-2 REACTOR BUILDING GROSS DECONTAMINATION EXPERIMENT

The procedures in this Appendix, which are based on the analysis given in Appendix A of Reference 42, outline the methods used to calculate cumulative absorbed dose as a function of time.

Table C.1 lists the radionuclides and pertinent decay parameters used for the calculations. A waste container is assumed to be a 55-gal drum the approximate dimensions of which are radius = 25 cm and height 100 cm. The activity is assumed to be uniformly discributed throughout the volume of the drum. The activity densities used are from Table 4.6 and 4.7.

Table C.1

Nuclides and Relevant Decay Data Used in Calculation of Dose to Strippable Coating

Radionuclide	λ (yr^{-1})	$t_{1/2}$ (yr)	$\bar{E}$[a] MeV	Γ[a] (rad $cm^2 h^{-1} mCi^{-1}$)
Sr-90	0.025	28	0.200	no γ
(Y-90)[b]	(95)	(.0073)	(0.931)	(no γ)
Cs-134	0.33	2.1	0.152	8.7
Cs-137	0.023	30	0.195	3.3
Pu-239		24,100	5.19	no γ

[a]Reference 1.
[b]Daughter of Sr-90; for the purpose of calculation, Y-90 was assumed to decay coincident with parent decay.

The dose delivered by beta decay was calculated from the following equations. The dose delivered by the alpha decay of the Pu-239 was calculated using the same formulation as for beta decay. The initial beta dose rate of the ith radionuclide, D_i, is:

$$\dot{D}_i^{\beta} = A\, C_i \bar{E}_i$$

CUMULATIVE DOSE CALCULATIONS FOR THE ALARA 1146 STRIPPABLE COATING SAMPLES FROM THE TMI-2 REACTOR BUILDING GROSS DECONTAMINATION EXPERIMENT

C_i is the activity density of the ith radionuclide, $\bar{E}_i$ is the average beta energy and A is a proportionality constant. When C_i is in mCi/cm^3 and $\bar{E}_i$ is in MeV, A equals 2.1×10^3 rad cm^3 MeV^{-1} h^{-1}mCi^{-1} and D_i is obtained in rad per h. The total absorbed beta dose due to the decay of the ith radionuclide, $D_i^{\beta}(\infty)$, is

$$D_i^{\beta}(\infty) = \frac{D_i^{\beta} \times 8.76 \times 10^3 \text{ h}\cdot\text{yr}^{-1}}{\lambda_i}$$

where λ_i is the decay constant of the ith radionuclide in years^{-1}. The beta dose absorbed at any time may then be calculated by

$$D_i^{\beta}(t) = D_i^{\beta}(\infty)\ (1-e^{-\lambda_i t}).$$

The dose delivered by gamma decay was estimated from the following equations. The gamma dose rate is

$$D_i^{\gamma} = C_i \Gamma_i \bar{g}$$

where Γ_i is the gamma ray constant of the ith radionuclide and $\bar{g}$ is a geometric factor, which assumes tissue equivalency. Γ_i has the units rad cm^{2}mCi^{-1}h^{-1}, $\bar{g}$ has the unit cm^{-1}.

The value of $\bar{g}$ = 136 was taken from the table of values of $\bar{g}$ given in Reference 2 for a cylinder of radius = 25 cm and height = 100 cm.

The total gamma absorbed dose, $D_i^{\gamma}(\infty)$, is

$$D_i^{\gamma}(\infty) = \frac{D_i^{\gamma} \times 8.76 \times 10^3 \text{ h}\cdot\text{yr}^{-1}}{\lambda_i}$$

and the cumulative gamma dose was obtained from

$$D_i^{\gamma}(t) = D_i^{\gamma}(\infty)\ (1 - e^{-\lambda_i t})$$

The total cumulative absorbed dose for all nuclei and decay types is shown in Table C.2.

CUMULATIVE DOSE CALCULATIONS FOR THE ALARA 1146 STRIPPABLE COATING SAMPLES FROM THE TMI-2 REACTOR BUILDING GROSS DECONTAMINATION EXPERIMENT

Table C.2

Total Absorbed Dose to the Average TMI Strippable Coating in 300 Years Assuming a 55-Gal Drum and the Activity Densities in Tables 4.6 and 4.7

Radionuclide	Activity Denisty ($\mu Ci/cm^3$)	$D_i^{\beta}(300)$ (rad)	$D_i^{\gamma}(300)$ (rad)
Sr-90	0.25	3.6×10^4	----
Y-90	0.25	1.7×10^5	----
Cs-134	0.47	3.9×10^3	1.4×10^4
Cs-137	5.6	8.7×10^5	9.5×10^5
Pu-239	2.6×10^{-6}	75	----
	Total	$= 1.08 \times 10^6 +$	9.6×10^5
	Total	$= 2.04 \times 10^6$ rad	

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15 SUPPLEMENTARY NOTES

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16 ABSTRACT (200 words or less)

Decontamination activities from the cleanup of the Three Mile Island Unit 2 Reactor Building are generating a variety of waste streams. Solid wastes being disposed of in commercial shallow land burial include trash and rubbish, ion-exchange resins (Epicor-II) and strippable coatings.

The radwaste streams arising from cleanup activities currently under way are characterized and classified under the waste classification scheme of 10 CFR Part 61. It appears that much of the Epicor-II ion-exchange resin being disposed of in commercial land burial will be class B and require stabilization.

Strippable coatings being used at TMI-2 were tested for leachability of radionuclides and chelating agents, thermal stability, radiation stability, stability under immersion and biodegradability. Results indicated that both radionuclide contamination and chelating agents leach from strippable coating waste.

17 KEY WORDS AND DOCUMENT ANALYSIS

17b DESCRIPTORS

Decontamination, Radwaste, Ion-exchange resin, Strippable coatings, Waste classification

17c IDENTIFIERS/OPEN-ENDED TERMS

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