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**CHARACTERISTICS OF FUEL CRUD AND ITS IMPACT ON
STORAGE, HANDLING, AND SHIPMENT OF SPENT FUEL**

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SUMMARY

Corrosion products, called "crud," form on out-of-reactor surfaces of nuclear reactor systems and are transported by reactor coolant to the core, where they deposit on external fuel-rod cladding surfaces and are activated by nuclear reactions. After discharge of spent fuel from a reactor, spallation of radioactive crud from the fuel rods could impact wet or dry storage operations, handling (including rod consolidation), and shipping. It is the purpose of this report to review earlier (1970s) and more recent (1980s) literature relating to crud, its characteristics, and any impact it has had on actual operations.

Crud characteristics vary from reactor type to reactor type, reactor to reactor, fuel assembly to fuel assembly in a reactor, circumferentially and axially in an assembly, and from cycle to cycle for a specific facility. To characterize crud of pressurized-water (PWRs) and boiling-water reactors (BWRs), published information was reviewed on appearance, chemical composition, areal density and thickness, structure, adhesive strength, particle size, and radioactivity. Information was also collected on experience with crud during spent fuel wet storage, rod consolidation, transportation, and dry storage.

PWR crud is generally black or gray, being a partially substituted nickel ferrite, or spinel, having the generalized composition $Ni_xFe_{3-x}O_4$, where $0 \leq x \leq 1$. A typical metal content is about 78 wt% iron, 20 wt% nickel, with the remainder being other metals, including cobalt at about 0.03 to 0.11 wt%. Typical maximum values for areal density and thickness of crud on a PWR spent fuel rod are about 65 mg/dm² and 5 μ m; these maximums are found on rods in the top-center of the reactor core. These values decrease slightly towards the core periphery and substantially along the length of a fuel rod. The maximum areal density found by Westinghouse on old PWR spent fuel was 1214 mg/dm² (about 101 μ m thick assuming a density of 1.2 g/cm³).

BWR crud is generally a flocculent, red Fe₂O₃ with Fe being about 87 wt% of the metal content. Cu, Zn, Ni, and Mn are also found in some BWR cruds. The typical areal density of crud is about 500 mg/dm² for a BWR having a deep

bed polishing unit in the coolant condensate cleanup system and 150 mg/dm² for a BWR having a powdered resin unit.

Crud may exist in one or two layers on either PWR or BWR spent fuel rods. The outer layer of BWR crud is typically loose and porous; the inner layer, dense and tenacious. PWR crud, generally in a single layer, is much more adherent than the outer layer of BWR crud. In terms of shear force required for removal, quantitative measurements of the adhesive strengths of crud are unavailable. The best qualitative indication for shear strength is the tools used to remove crud samples: a nylon brush to remove loose deposits and a silicon carbide stone to remove tenacious deposits.

Individual crud particles are small, 0.1 to 2.0 μm , but these are agglomerated and may not easily become airborne to pose migration problems. Adhesive strength and particle size distribution, including that for aerosol particles, need quantification; modelers have identified that data are required for developing a source term model of a spent fuel transportation cask.

Cobalt-60 is the primary source of radiation from crud on spent fuel stored for a long period. For PWRs, the ⁶⁰Co activity range is about 0.1 to 140 $\mu\text{Ci/cm}^2$ at reactor shutdown; for BWRs, it is about 110 to 180 $\mu\text{Ci/cm}^2$.

Data concerning impacts of crud on spent fuel wet storage, handling, shipping, and dry storage operations is not as extensive as that for crud characteristics. Crud may be released from spent fuel during water storage pool manipulations, but it rapidly settles and can be removed by system cleanup equipment to satisfactory levels. After long-term water-pool storage, once-tenacious crud has been observed to loosen from spent fuel and to have a tendency to peel. During one demonstration of PWR rod consolidation, crud removed from the fuel during rod-pulling formed a "cloud" in the storage pool water. Although annoying and causing delay in operations, the dispersion had little effect on pool radioactivity or personnel exposure. Filtration methods have been improved to mitigate crud dispersion.

In a case involving shipping of BWR spent fuel from the West Valley Nuclear Services (WVNS) storage pool to a reactor pool, crud was released into the shipping cask during shipping or subsequent operations, and, in

spite of extensive flushing, a portion was retained in the cask. Later, during draining of the cask, crud was released to the drain hose, and because connecting the hose is a hands-on operation, radiation dose to the staff increased. In another program involving fuel shipping, small amounts of crud were released during shipping and handling of PWR spent fuel used for full-scale tests of the CASTOR-V/21 dry storage cask. Crud release was not a major problem and was localized and easily handled. During long-term, low-temperature (229°C), single-rod dry storage tests, using PWR and BWR spent fuel rods, only a small amount of crud was released. As a result, the researchers concluded that crud spallation during dry storage should be a manageable problem.

From experience with wet storage, rod consolidation, transportation, and dry storage, it appears crud spallation can be managed effectively, posing no significant radiological problems.

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INTRODUCTION

The word "crud" is used in the nuclear industry to designate the corrosion products (principally oxides of iron, nickel, chromium, copper, and cobalt) that form outside the core in primary coolant system heat exchangers, piping, pumps, etc.; that dissolve or are eroded away; or that circulate in the water coolant, and are deposited on fuel rods and non-heat-generating surfaces in the core and the reactor vessel. Crud does not refer to the dense, strongly adherent films formed by oxidation of fuel cladding, such as zirconium oxide.

Crud components on the surface of fuel rods become activated by nuclear reactions, for instance, $^{59}\text{Co} (n, \lambda) ^{60}\text{Co}$ and $^{58}\text{Ni} (n, p) ^{58}\text{Co}$ (Shaw 1986). Activated crud particles may be released from fuel rods to the coolant, redeposit at other locations on a fuel rod, or circulate and redeposit on primary coolant system surfaces outside the reactor core, where the crud can cause radiation exposure problems for reactor plant operators and maintenance personnel. The formation, transport, and deposition mechanisms have been discussed earlier by Carlson (1970), Berry (1971), Berry and Diegle (1979), and others. Crud can also remain on fuel rods and build up. This fuel crud may pose problems later: after the fuel is discharged from the reactor and stored in a water pool; when it is transferred in the pool for testing, reracking, rod consolidation, and other handling; or when it is loaded into a shipping cask, then shipped dry for transfer to other pools or placement into dry storage casks.

It is the purpose of this report to review some of the earlier (1970s) and more recent (1980s) literature as it relates to the impact of crud on storage, handling (including rod consolidation), shipment, and dry storage. Although fuel assemblies in service today in pressurized water reactors (PWRs) and boiling water reactors (BWRs) are much freer of crud than assemblies discharged and placed into storage many years ago, the older assemblies eventually may be consolidated or placed into dry storage. Consideration needs to be given to the potential impact of fuel rod-borne crud on future handling, storage, and shipment of spent fuel. Consequently, this report presents fuel crud characteristics related to both older and

newer spent fuel. Data collected from fuel-crud studies and general observations are given.

CONCLUSIONS

The following conclusions resulted from this study:

- Fuel crud has highly variable characteristics, which depend on the history of spent fuel operations. Typical fuel-crud characteristics and approximate measurable values are as follows:
 - PWR crud is generally black or shades of gray. It is usually a partially substituted nickel ferrite or spinel: $Ni_xFe_{3-x}O_4$, where $0 \leq x \leq 1$. The compounds NiO , SiO_2 , and CrO_3 and the element Ni have also been found. A typical metal content is 78 wt% Fe, 20 wt% Ni, 2 wt% other metals, including 0.03 to 0.11 wt% Co. Most crud on PWR fuel rods is at the top of the core. The typical areal density of PWR crud is about 48 to 65 mg/dm² (4 to 5 μ m thick) from the top periphery to the top center of the core. At midcore the areal densities are about 4 to 6 mg/dm² (0.3 to 0.5 μ m thick). Maximum areal densities that have been found for Westinghouse PWR fuel rods are 1214 mg/dm² (~101 μ m thick) at the top center of the core and 470 mg/dm² (~39 μ m thick) at the top periphery.
 - BWR crud is generally a flocculent, red Fe_2O_3 , with Fe being about 87 wt% of the metal content. Small amounts of Cu, Zn, Ni, and Mn are also found. The copper content was higher in crud on early BWR fuel rods, but its content has been decreased by equipment and process changes. However, a copper-rich crud (Cu about 50 wt% of metal content) associated with (U,Gd) O_2 rods was found in 1979. It was related to a mechanism called crud-induced localized corrosion (CILC); only a limited number of rods have been affected by the CILC mechanism. The areal density of more typical BWR crud is about 500 mg/dm² for a reactor having a deep bed polishing unit in the condensate cleanup system and 150 mg/dm² for a system having a powdered resin polishing unit. The heaviest deposits occur in the zone of subcooled boiling, typically about a third of the rod length from the coolant inlet and of the assembly.
 - Crud may be in the form of a loose deposit, a tightly adherent deposit, or both, in two layers on BWR rods. A tightly adherent

crud usually occurs on PWR rods. The outer layer of BWR crud is a loose, highly porous, flocculent layer; the inner layer is tenacious. In terms of shear force required for removal, quantitative measurements of the adhesive strength are unavailable. The best indication of adhesive strength is the tools used to remove crud samples, i.e., a nylon brush to remove loose deposits and a silicon carbide stone to remove hard, tenacious deposits. Adhesive strength for different crud needs further study and measurement; modelers have identified that data for these fuel crud characteristics are required for a spent fuel transportation cask source term model.

- Scanning electron microscopic (SEM) examinations made on loosely adherent and tenacious crud samples from BWR fuel rods showed clusters of small particles in the 0.1 to 0.3 μm range, and larger particles in the 0.5 to 2.0 μm range. Due to agglomeration, the particles may not easily become airborne. Like the lack of data for adhesive strength, the effect of agglomeration needs quantification.
- Cobalt-60 is the primary out-of-reactor source for radiation from crud on spent fuel stored for a long period. Other radioisotopes that may be present on fuel upon reactor discharge are ^{58}Co , ^{54}Mn , ^{65}Zn , ^{59}Fe , and ^{51}Cr , but these have much shorter half-lives than ^{60}Co and, thus, diminish more quickly than ^{60}Co . Radioactivity measurements range widely. For PWRs, the ^{60}Co range found in terms of area is 0.1 to 140 $\mu\text{Ci}/\text{cm}^2$. One literature reference expressed the ^{60}Co composition of a typical PWR crud as 25,000 to 100,000 $\mu\text{Ci}/\text{mg}$ parent, i.e., ^{59}Co , rather than in activity per unit of rod area. Another reference discussing BWRs gives the following as typical for fuel crud ^{60}Co activities: BWR with deep bed polishing in the condensate cleanup system, 180 $\mu\text{Ci}/\text{cm}^2$; BWR with powdered resin polishing, 110 $\mu\text{Ci}/\text{cm}^2$.
- Public safety implications of fuel-crud release and dispersal are minimal for an independent spent fuel storage installation and reactor pools. Although crud may be released from spent fuel and form a

temporary cloud during pool manipulations, it rapidly settles and is easily removed to satisfactory levels by system cleanup equipment.

- After long-term water-pool storage, once-tenacious crud may loosen from spent fuel and have a tendency to peel.
- Crud clouds have been observed in spent fuel water-storage pools during rod consolidation. Although annoying and causing delay in operations, the dispersion had little effect on radioactivity of the pool or personnel exposure. Filtration methods have been devised to mitigate crud dispersion.
- During testing of the CASTOR-V/21 PWR spent fuel storage cask at the Idaho National Engineering Laboratory, small amounts of crud were found in the shipping cask after dry shipment of spent fuel and its removal, and on the personnel work platform located between the casks following transfer of the spent fuel to the storage cask. The small amount of release crud, however, posed no significant problem and was easily cleaned up.
- In another case involving the return shipment of stored spent BWR fuel from the West Valley Nuclear Services (WVNS) water-storage pool to a reactor pool, even after thorough flushing crud was released from the shipping cask during spent fuel removal at the reactor pool, and again from the cask during its return to the WVNS water pool. Activity spikes were observed at the latter pool. Additional radiation exposure to personnel may have occurred during cask draining operations.
- Only very small amounts of crud spallation have been observed in dry storage tests of spent fuel. As a result of observations made during single-rod tests, the researchers concluded that crud spallation during dry storage should be a manageable problem.

CRUD CHARACTERIZATION

The character of crud is diversified. It has varied from reactor type to reactor type, reactor to reactor of the same vendor type, fuel assembly to fuel assembly in a reactor, circumferentially and axially in an assembly, and, perhaps most importantly, from cycle to cycle for a specific facility. The latter variations are due to several factors: improved operation, replacement of equipment with that using less corrodible materials (e.g., titanium or stainless steel instead of copper alloys in BWR heat exchangers), and use of improved, controlled water chemistry. The history of LWR reactor operations shows that with the use of improved technology, less crud is produced. The following sections discuss the appearance of crud, its chemical composition, areal density and thickness, structure, and radioactivity.

APPEARANCE

Spinel of the magnetite (Fe_3O_4) and nickel ferrite ($\text{Ni}_x\text{Fe}_{3-x}\text{O}_4$) types are the major components of PWR crud. Because of these oxides, crud on PWR fuel rods generally appears black or in shades of gray due to the reducing environment caused by hydrogen addition to PWR primary coolant. Because of the presence of a small amount of oxygen in the coolant, crud deposits on BWR fuel rods are usually reported as being red since hematite ($\alpha\text{Fe}_2\text{O}_3$) is the principal deposit which has been found; but Fe_3O_4 is also present (Berry and Diegle 1979). Variations in color are not unusual, however. To help establish initial conditions of spent fuel rods before dry storage testing, Einziger and Cook (1984b) observed that crud on PWR fuel rods was black, gray, reddish brown, reddish orange, and brown tan; and that crud on BWR fuel rods was brown, reddish brown, and orange. They cautioned, however, that visual observations are an unsatisfactory means of crud evaluation because hot cell and underwater lighting affect the colors that appear on the rods. Davis (1980) observed Turkey Point (PWR) fuel rods (average core burnup: 25.7 MWd/kgU) and noted gradual color changes in crud from black to shades of gray from the bottom to the top of the rods. The material observed, however, may have been zirconium oxide, the corrosion product of the cladding. The color variations suggest that the average crud chemical composition may also

vary along the length of fuel rods. In fact, Riess (1976) has reported that chemical composition changed considerably along the length of fuel rods removed from the PWR reactor at Obrigheim (KW0) after two cycles of operation. At the bottom of a fuel rod, the chemical composition of crud was 40% Fe₂O₃, 25% NiO, and the remainder SiO₂, Cr₂O₃, etc.; at the top it was 20% Fe₂O₃, 70% NiO, and the remainder SiO₂, Cr₂O₃, etc.

CRUD COMPOSITION

PWRs

Generalized compositions for crud deposits on fuel assemblies have frequently been described. Strasser, Sheppard, and Santucci (1985) state that crud particles in PWRs, either in the coolant or deposited on fuel rods, have an approximate metal composition as follows:

Fe	78%
Ni	20%
Cr, and other metals	2%

This composition differs from the relative weighted surfaces of stainless steel and Inconel in a typical large PWR:

Fe	14%
Ni	70%
Cr, and other metals	16%

They further say that the major constituent of PWR crud has been found to be partially substituted nickel ferrite: Ni_xFe_{3-x}O₄, where 0 ≤ x ≤ 1. With x greater than 0.5 there would be an excess of Ni for nickel ferrite, and either Ni or NiO would be expected. The typical composition of fuel crud is Ni_{0.6}Fe_{2.4}O₄ with the Ni/Fe ratio being 0.25.

Bergmann, Roesmer, and Perone (1983) also give typical core-deposit data for the first two cycles of a PWR. These and other PWR fuel-crud data are given in Table 1. The same major chemical compound was given for crud, but with a slightly higher range for nickel, 0.4 ≤ x ≤ 0.9. They also state that Ni and NiO are sometimes present.

TABLE 1. Characteristics of PWR-Core Crud^(a)

<u>Element,</u>	<u>wt%</u>	<u>Radiochemical Compositions, $\mu\text{Ci/mg Parent}$</u> ^(b)	
Fe	39-47	^{58}Co	800-1300
Ni	19-24	^{60}Co	25,000-100,000
Cr	0.8-2.5	<u>Surface Concentration, mg/dm²</u>	
Co	0.11	10-350	
<u>Thickness, μm</u>		<u>Density, g/cm³</u>	
~0.8-29		~1.2 (estimated)	

(a) Typical data per rod for Plant B, Cycles 1 and 2.

(b) Origin element for radionuclides; fuel age unspecified.

BWRs

Iron oxide, often in two layers, in the form of $\alpha\text{-Fe}_2\text{O}_3$ is the primary constituent of crud deposited on BWR fuel rods; only trace quantities of other iron oxide crystalline structures, such as Fe_3O_4 and iron hydroxides, have been observed. Significant quantities of metallic elements other than iron (e.g., Co, Ni, and Cu) are present in some deposits (Anstine and Naughton 1981). A typical analysis of standard BWR fuel crud is given in Table 2 (Baily, Marlowe, and Proebstle 1985).

The primary sources of crud in a present-day BWR are the carbon steel surfaces in the feedwater system, which corrode to produce iron oxides; other sources are stainless steel surfaces of the feedwater and reactor coolant systems, and copper alloy surfaces of the condenser. Even though it has less surface area in these systems than stainless steel, carbon steel is the principal source for crud because of its higher corrosion rate (Strasser, Sheppard, and Santucci 1985). Anstine and Naughton (1981) state that 60% to 80% of the corrosion products entering the primary system from the feedwater

TABLE 2. Typical Analysis of Standard BWR Fuel Crud^(a)

<u>Element</u>	<u>Standard Crud, wt%</u>
Iron	87
Copper	2
Zinc	4.4
Nickel	3.3
Manganese	2.2
Chromium	1.1
Cobalt	0.3

(a) Principal compound Fe₂O₃

train deposit rapidly on fuel surfaces. Niki et al. (1985) reports that over 90% of the feedwater-borne crud is deposited on the fuel of the Shimane nuclear power station in Japan.

In early BWRs, corrosion of copper alloy heaters used in the feedwater system led to introduction of copper and nickel into the primary system at nearly the same rate as that for iron. The resulting crud deposits on fuel rods were hard and tenacious and had poor heat-transfer characteristics. Since 1965, ferrous-alloy systems (or titanium) have been used successfully to significantly reduce the amount of corrosion products other than iron to the primary system by way of the feedwater (Blok, Sawochka, and Snyder 1973).

Differences in composition of fuel-deposited crud have also been found due to the two types of water cleanup systems used (Berry and Diegle 1979). Iron composed 94% of the metallic elements present in fuel crud in BWRs having deep bed polishing units in the condensate cleanup system. On the other hand, Fe composed only 65% of these metallic elements in a BWR using powdered resin units. The latter fuel crud had high zinc levels (Berry and Diegle 1979) probably from Admiralty brass heat exchanger tubing. The first type of corrosion product is generally flocculent and easy to remove; the second, generally thinner but tenacious. Strasser, Sheppard, and Santucci (1985) also describe the difference between deposits on BWRs having deep-bed or powdered resin filter units. After three operating cycles, crud found on

fuel rods of Nine Mile Point-1 (NMP-1), which uses the deep-bed units, was flocculent. This crud was easily dislodged by brushing; no hard, tenacious deposits were found. After two cycles, crud layers on fuel rods of Monticello, which uses the resin filter units, were ten times thinner than crud layers found on NMP-1 fuel rods and consisted of a relatively hard, tenacious sublayer and a more loosely adhering outer layer. Nylon brushing could remove only about half of the deposits. The zinc content of the Monticello fuel crud was also relatively high. Compositions of crud removed by using a nylon brush (B) or a silicon carbide stone (S) are shown in Table 3.

Marlowe et al. (1985) and Baily, Marlowe, and Proebstle (1985) report finding in 1979 a new and unexpected mechanism of localized fuel cladding corrosion failure in some BWRs. The failures were highly concentrated in (U,Gd)₂O₂ rods having exposure levels of 15 to 20 MWd/kgU. Cladding breach by this mechanism, however, has occurred in only a small fraction of General Electric fuel rods that have experienced BWR service. The crud-induced localized corrosion (CILC) has been limited exclusively to plants with copper alloy condenser tubes and filter-demineralizer condensate cleanup systems. Rather than the typical flocculent iron oxide crud, a flaking deposit was found that was very dense. This deposit, having high copper concentrations, was found closely associated with a white zirconium oxide, nodular corrosion product often found on BWR fuel rods.

About 50% of the cation content of this type of crud was copper, rather than over 90% iron, as in typical fluffy crud. Adamson et al. (1986) described a typical analysis for a crud causing CILC failures:

<u>Element</u>	<u>Wt%</u>
Fe	21
Ni	3
Cr	3
Mn	3
Co	1
Cu	53
Zn	11

TABLE 3. Elemental Composition of BWR-Fuel Deposits

	<u>Monticello (midcore)</u> <u>16,070 EFPH(a) (Cycles 1 & 2)</u>		<u>Nine Mile Point-1 (midcore)</u> <u>18,900 EFPH (Cycles 1, 2 & 3)</u>	
	<u>B(a)</u>	<u>S(a)</u>	<u>B</u>	<u>S</u>
Fe	82.0	63.0	97.0	96.0
Cu	2.3	6.4	0.5	0.7
Ni	1.9	5.6	1.7	1.5
Co	0.16	0.45	0.1	0.1
Cr	1.7	2.9	0.5	0.6
Mn	1.8	4.8	0.6	0.8
Zn	6.0	14.0	0.3	0.3
Mg	1.4	1.1	-	-
Ca	2.5	1.6	-	-

(a) EFPH - Effective full power hours, B - nylon brushed sample, S - silicon carbide stoned sample

Comments on Crud Compositions

The metal elemental compositions of PWR and BWR cruds given by numerous references are presented in Table 4. The data have been arranged approximately from fairly recent age to earlier ages (e.g., 1965) for spent fuel, insofar as identifiable, to show any effects of improvements in water chemistry, reactor coolant system structural material, etc. on crud composition. In many cases a complete material balance was not given in the references. In some instances, an oxide percentage was given rather than element weight percentage. Hence, values must be compared with some caution. Nevertheless, approximate and relative comparisons can be made.

Copper content of crud was high for some BWR spent fuel discharged before 1971, for example, at Big Rock Point (ca 1968) and Dresden-1. The copper oxide cruds were hard and tenacious. Problems from copper except those recently associated with CILC were later essentially eliminated by changes in equipment materials and improved water coolant chemistry.

TABLE 4. Elemental Compositions of Fuel-Crud Samples from Various PWR and BWR

Reactors	Date	Elements, wt% of Total								Reference
		Fe	Ni	Cr	Mn	Co	Cu	Zn	Other	
<u>PWRs</u>										
Typical		78	20	2						Strasser et al. (1985)
Plant B, typical, CY(a)1, 2		39-47	19-27	0.8-2.5		0.11				Bergmann et al. (1983)
H. B. Robinson 2 (Rod B0-5-H9)	1974									
15 in. from rod top		68	28	4	3					Einziger (1984a)
137 in.		74	18	6	3					
148 in.		81	17	2						
Zorita (40 GWD/MTU)	1972	61	26	2					Zr: 6	Einziger (1984a)
Zion 1, EOC 2	before 1979	40±10	37±12	4±1	1.2				16 (incl Zr)	Einziger (1984a)
EOC 3		59± 5	21± 3	7±1					10 (incl Zr)	Einziger (1984a)
Oconee 1 Destructive Test	ca 1979	34	39	27						Einziger (1984a)
Nondestructive Test		40	57	3						
Oconee 1 run	ca 1978	40-70	25-60	4-8					Zr: 1	Berry (1979)
Obrigheim (KW0) (EOC 2)	ca 1975									
Top of Rod		20 ^(b)	70 ^(b)							Riess (1976)
Bottom of Rod		40 ^(b)	25 ^(b)							
Beznau I (NOK) CY1	1971	47	14.4	1.3	0.14	0.034				Solomon, Roesner (1976)
CY3	1974	45	21	0.78	0.13	0.03				
Point Beach I, CY1	ca 1972	30	12.5	1.6	0.06	0.025				Solomon, Roesner (1976)
Yankee Rowe	ca 1965	59	8	0.6	0.3	0.2				Berry (1979)
Saxton	ca 1965	40	8	11	10	0.09				Berry (1979)

(a) Cy - Cycle, EOC - End of cycle, EFPH - Effective full power hours, EFPD - Effective full power days.
 (b) Oxide percentage.

Reactors	Date	Elements, wt% of Total								Reference
		Fe	Ni	Cr	Mn	Co	Cu	Zn	Other	
BWRs										
Typical (present day)		87	3.3	1.1	2.2	0.3	2	4.4		Baily et al. (1985)
Nine Mile Point-1	1971-73									
CY1,2,3 (18,900 EFPH)										
Brush removed (loose)		97.0	1.7	0.5	0.6	0.1	0.5	0.3		Strasser et al. (1985)
SiC stone removed (tight)		96.0	1.5	0.6	0.8	0.1	0.7	0.3		
Monticello	1974									
CY1,2 (16,070 EFPH)										
Brush removed (loose)		82	1.9	1.7	1.8	0.16	2.3	6.0	Mg, Ca	Strasser et al. (1985)
SiC stone removed (tight)		63	5.6	2.9	4.8	0.45	6.4	14.0	Mg, Ca	
Oskarsham 1	ca 1978									Berry (1979)
26,000 EFPH										
Loose layer										
(>50% of deposit)		68-76	7.1-8.3	1.7-3.7	2.0-2.2		3.9-9.0	8.9-11		
Adherent layer		47-69	6.2-8.4	2.0-5.8	2.1-3.0		8.7-23	12-13		
Dresden-2		96	2	<1	<1	<1	1	<1		Adamson et al. (1986)
BWR with deep bed polishing unit										Berry (1979)
20,000 EFPH		94	3			0.1	1.0	0.3	2	
BWR with powdered resin polishing unit										
16,000 EFPH		65	6			0.4	4.0	14.0	10	Berry (1979)
Millstone 1	before 1975	94.7	2.4	<1	<1	<1	1.4	<1		Einziger (1984a)
Tsuruga	ca 1973	96								Einziger (1984a)
Peach Bottom-II	1974									Einziger (1984a)
(Rod PH2 PH-482-E8)										
20 in. from rod top		62	16	1	7		4	11		
60 in.		60	11	1	4		15	8		
156 in.		69	15	1	3		6	6		
Dresden-1	before 1971	14-21	66-74	0.74-2.5			4.7-15.2			Einziger (1984a)
1568 EFPD										
Big Rock Point	ca 1968						Majority(a)			Lunde 1975

(a) Essentially pure copper components in feedwater system later avoided by removal of copper components and better feedwater chemistry.

AREAL DENSITIES AND THICKNESSES OF CRUD ON FUEL ASSEMBLIES

Knowledge of the quantity, thickness, and distribution of crud on fuel rods and means to diminish crud buildup are of major interest for reactor operations. Diminishing crud buildup on fuel rods is desirable to prevent rod overheating (Shaw 1986), which may arise from crud-induced poor heat transfer, and corrosion and breaching of fuel cladding. Improved water coolant chemistry, upgrading of water coolant system equipment (e.g., replacement of copper alloy tubing with stainless steel or titanium in BWR systems), and advanced processing of the coolant have all led to much lower deposition of crud on fuel during the past decade or so. Improvements are continuing to be developed, as shown by recent reports and guidelines on water chemistry (Fejes, Ivars, and Svensson 1984; Strasser, Sheppard, and Santucci 1985; Shaw 1986).

The quantity, thickness, and distribution of crud are also important for handling, storage, and other operations following reactor discharge and cooling of the fuel. This is especially so if the fuel is to be consolidated, placed into casks, shipped, removed from shipping casks, and stored dry in storage casks. Release of loose, small-particle crud or spalling of relatively dense, once-tenacious crud could potentially increase radiation exposure of personnel during these handling operations and during subsequent maintenance of fuel handling equipment and facilities.

Measurements or results of observations of areal densities (mg/dm^2) and thickness (μm) of crud on fuel discharged from reactors from the late 1960s to the early 1980s are presented in Table 5.

Crud areal densities have been determined by removing crud from measured areas of fuel rods and weighing the collected material. Nylon brushes have been used to remove loose crud and a scraper, such as a silicon carbide abrasive stone, has been used to remove strongly adherent material (Strasser, Sheppard, and Santucci 1985). Solomon and Roesmer (1976) describe a sampling tool developed by Westinghouse Electric Corporation for use in determining areal densities and other crud properties. Utility of this device has been reaffirmed by Solomon et al. (1984).

TABLE 5. Crud Areal and Thickness Measurements, Averages and Ranges

Reactor	Date	Areal Density, mg/dm ²	Thickness, μm	Comments	Reference
<u>Pressurized Water Reactors (PWRs)</u>					
Westinghouse studies of 150 plants, representing 28 cycles	12 yrs of evaluation 1971-1983				Solomon et al. 1984
Average Values (Maximum Values)					
Top core center		8 (1214) ^(a)	-5 ^(b) (-101) ^(b)		
core periphery		48 (470)	-4 ^(b) (-39)		
Middle core center		4 (12)	-0.3 ^(b) (-1)		
core periphery		8 (32)	-0.5 ^(b) (-2.7)		
Bottom		No Measurements			
Plant B (typical) CY(c)1,2	ca 1983	10-350	0.8-29	Density: 1.2 g/cm ³ (estimated)	Bergmann 1983
KWU PWRs, typical	ca 1985	Crud deposits practically zero			Knaab 1985 (also Peehs 1986)
KWU Gosgen-Daniken (KKG)	after 1982	No measurable deposits found			Garzarolli 1985
PWRs with FRAGEMAs fuel, >30,000 MWD/MTU	ca 1985	Fuel crud very light or nonexistent		Use coordinated Li/B chemistry for coolant	Thomazet et al. 1985
Trojan EOC 1	1978	110-200	9-13	Deposits non-uniform axially. Low lithium, low hydrogen in coolant	Strasser et al. 1985
EOC 3	ca 1982	2-8	~0.2-0.5	New assemblies, high lithium	Strasser et al. 1985

(a) Maximum values are in parentheses.

(b) Calculated using density of 1.2 g/cm³, a value estimated by Westinghouse.

(c) CY - Cycle, EOC - End of cycle, EFPH - Effective full power hours, EFPD - Effective full power days.

<u>Reactor</u>	<u>Date</u>	<u>Areal Density, mg/dm²</u>	<u>Thickness, μm</u>	<u>Comments</u>	<u>Reference</u>
Beaver Valley EDC 1	1979	40 avg.	~3	Deposits fairly uniform axially; factor of 2 between maximum and minimum	Strasser et al. 1985
Belgian 3(BR-3) CY4B	1980		80-120 max		Strasser et al. 1985
Ringhals 2 EDC 1	1979		75		Strasser et al. 1985
Zorita CY2	1972		100 max		Strasser et al. 1985
CE Plant D CY1		1000 max	~100		Strasser et al. 1985
Surry Cycle 1 Cycle 2	before 1979		0.03-0.4 1.5-3.3		Einzigler 1984a
Oconee I	before 1979		2.5		Einzigler 1984a
Zion I EDC 2 EDC 3	before 1979		0.5-10 0.5-19		Einzigler 1984a
Obrigheim (KW0)	ca 1975	800 max	(~67)	Thickness varied from top to bottom with maximum at top. Hydrogen overpressure inadequate.	Riess 1976
H.B. Robinson-2 (Rod B0-50-H9) 15 in. from rod top 137 in. 148 in.	1974	48 7 81	5 0.76 7.6		Einzigler 1984a
Stade (KKS) after 2 cycles	ca 1975	negligible deposits			Riess 1976
Turkey Point 26.7 MWd/kgU	1972		3.8-18		Davis 1980
Point Beach Cycle 1	ca 1972	0.7-46	0.05-3.8	Very thin, relatively uniform; average density ~1.2 g/cm ³	Solomon, Roesmer 1976

<u>Reactor</u>	<u>Date</u>	<u>Areal Density, mg/dm²</u>	<u>Thickness, μm</u>	<u>Comments</u>	<u>Reference</u>
Beznau I (NOK) CY1	1971	47-470	3.8-38	Samples from core top; high values at periphery	Solomon 1978
CY3	1974	0.6-48	0.9 avg		
Yankee Rowe, Core I	ca 1965	68-152	2.3		Berry 1979
Core I reused		1010-1450	100		
Saxton Core I	ca 1965	53-83	3.3-5.3		Berry 1979
<u>Boiling Water Reactors (BWRs)</u>					
BWR with deep polishing unit 20,000 EFPH	ca 1978	500			Berry 1979
BWR with powdered resin polishing 16,000 EFPH	ca 1978	150			Berry 1979
Scandinavian BWRs	ca 1984	100 (typical)			Fejes et al. 1984
KWU (Kraftwerk Union) BWRs	1987	Very low, generally			Knaab 1985
Peach Bottom-II Rod PH-462-E6 in. from Top:	1974				Einzigler 1984a
20		0	0		
60		21	1.8		
156		16	1.3		
Monticello EOC 2-4	1974- 1977	20-200			Strasser et al. 1985
Nine Mile Point-1 1st 3 cycles	1971- 1973	600-1000 (mg Fe/dm ²)			Strasser et al. 1985

<u>Reactor</u>	<u>Date</u>	<u>Areal Density, mg/dm²</u>	<u>Thickness, μm</u>	<u>Comments</u>	<u>Reference</u>
Brunswick Unit 2 EOC 1	ca 1977	400-700 avg. 3600 peaks			Strasser et al. 1985
TVO-1 CY1		300 (peak) 74 (avg)			Strasser et al. 1985
Nuclenor, Tarapur		2000			Strasser et al. 1985
Oskarsham 1 26,000 EFPH	ca 1976	2-60			Berry 1979
KRB, Gundremmingen	ca 1973		100		Lunde 1975
Millstone I	before 1975		41-61	Based on density of 1 mg/cm ³	Einzigler 1984a
Tsuruga	before 1975		71-250	Based on density of 1 mg/cm ³	Einzigler 1984a
Dresden-1 1568 EFPD	before 1971		28		Lunde 1975
<u>Other Reactor Types</u>					
Bruce (CANDU)	ca 1977		0.03	Based on density of 1 mg/cm ³	Einzigler 1984a
Douglas Point (CANDU)	ca 1977		1, 0.1, 0.5	Based on density of 1 mg/cm ³	Einzigler 1984a
SGHWR	ca 1973		75 20-50	Some fuel failures	Lunde 1975

Crud thicknesses have been determined by using metallographic techniques or, more recently, by a nondestructive eddy-current test method. Solomon and Roesmer (1976) estimated that the rough average density of PWR crud is about 1.2 g/cm^3 (others have used 1.0 g/cm^3), which gives a specific weight of 12 mg/dm^2 (equivalent to $1 \text{ }\mu\text{m}$ thick). They have used this value to estimate PWR-crud thicknesses.

Crud deposition is non-uniform. Solomon et al. (1984) conducted a systematic twelve-year Westinghouse Electric Corporation study started about 1971. It focused on core deposits on 150 PWR fuel assemblies in seventeen plants representing twenty-six cycles. They concluded that most crud deposits are near the top of the core. These Westinghouse data are shown in Table 5. Roesmer (1984) observed that Beaver Valley and other PWR cores had deposit thicknesses inversely proportional to axial fuel assembly power. Similar observations had been reported earlier by Riess (1976). Roesmer and Rootham (1978) stated that deposits found on a given core are often heavier after the first cycle than after subsequent cycles. Individual fuel rods have been observed to have a variety of crud distribution patterns: flow outline, circumferential, raked, spot, and bamboo effect (Solomon and Roesmer 1976).

Deposit distribution also varies for BWR fuel rods. Lunde (1975) discovered that the deposition of crud on a cladding surface depends on temperature, heat flux, and flow conditions, and that thickness varies over fuel-rod length. Persson, Mutler, and Stenberg (1981) discerned that wide variations occur between different fuel assemblies in a given reactor, indicating a high influence of local power density conditions. It was noted that the heaviest deposits occur in the zone of undercooled boiling. It was shown that the heaviest deposits were typically located roughly one-third of the fuel-rod length from the coolant inlet end of the fuel assembly. Blok, Sawochka, and Snyder (1973) had similar findings: crud peaked at about 30 inches above the coolant inlet end of fuel assemblies and was negligible near the coolant outlet end. They also found that radially, the heaviest crud deposit was in the peripheral, low-flux regions of the core, and that the deposit amount could be correlated reasonably well with wall fluid shear: i.e., heaviest in low shear regions of the fuel assembly; lightest in the

high shear, center core regions near the coolant outlet end of the assembly. Niki et al. (1985) also state that at the Shimane BWR nuclear power station in Japan thicker deposits occurred near the bottom of fuel assemblies and thinner at the top.

With the recent improvements in water chemistry at both PWR and BWR reactors (methods described by Strasser, Sheppard, and Santucci 1985), visual examinations are showing that crud deposited on fuel rods is diminishing. [Note the recommended specifications given by Shaw (1986) for PWRs, and Baily, Marlowe, and Proebstle (1985) for BWRs. See, too, Garzarolli et al. (1985) and Knaab and von Jan (1985) regarding KWU PWRs and BWRs; Dehon et al. (1985) discussing thirty-eight PWRs operating with fuel supplied by FRAGEMA, and Olsen (1985) describing both PWR and BWR fuel rods.]

Thus, the amount of crud and its distributions on PWR and BWR fuel rods are highly variable. Older fuel assemblies with thicker crud deposits that have been exposed longer to water storage may be more susceptible to crud loosening during handling and fuel management operations such as shipping, rod pulling for rod consolidation, and dry storage. Dispersal of crud during operations and equipment maintenance may increase dose levels for the personnel involved.

Although crud distributions are highly variable throughout reactor cores, the total core deposited crud is of interest for fuel handling operations after reactor discharge. Some estimates have been made.

Roesmer and Rootham (1978) give estimates of weights of elements in crud in the table below for a 1000 MWe PWR plant after twelve months of operation. The estimates are based on average crud compositions of Cycle 1 cores of Wisconsin Electric Power (WEP) Unit 1 and Beznau-1 (NOK) nuclear reactors.

<u>Element</u>	<u>WEP, kg</u>	<u>NOK, kg</u>
Fe	15.1	24.0
Ni	6.5	7.3
Cr	0.82	0.68
Co	0.013	0.017

Strasser, Sheppard, and Santucci (1985) have estimated the total amounts of iron on fuel rods of Monticello and Nine Mile Point-1 cores. The calculated

estimates of iron balances are based on iron distributions found on fuel rods at ends of fuel cycles 1 and 2. These BWRs are rated at 570 MWe and 620 MWe, respectively. The data are below:

	<u>Monticello(a)</u>		<u>Nine Mile Point-1</u>	
	<u>EOC(b)1</u>	<u>EOC2</u>	<u>EOC1</u>	<u>EOC2</u>
Effective Full Power Hours (EFPH)	10,170	16,070	12,100	18,900
Total Iron on Fuel Core, kg	20	35	149	165
Crud Type	Hard, tenacious sublayer; more loosely adhering outer layer		Easily dislodged by brushing; no hard, tenacious deposits found	

(a) Monticello is recognized as a relatively "clean" plant.
 (b) EOC - End of cycle.

CRUD STRUCTURE

Types of Crud

Two types of crud that may form in layers on PWR and BWR fuel rods are discussed in the literature: one a loose, flocculent, or fluffy layer; the second, a tightly adherent layer. The two-layer crud is associated with BWR fuel rods. The loose, flocculent crud also is more frequently associated with BWR fuel than PWR fuel (Bailey et al. 1982).

PWR crud is a non-stoichiometric nickel ferrite, a nickel-substituted magnetite determined to have a spinel lattice by x-ray diffraction spectroscopy (Roesmer 1984, Solomon et al. 1984). Berry and Diegle (1979) state that it has long been known that the corrosion product forming on boiler steels under low oxygen conditions (ppb range) is magnetite (Fe_3O_4 spinel). The hydrogen levels used in PWR coolant to produce low oxygen content ensure that spinels are stable over the operating temperature range of the reactor plant.

Wood (1986) reports several crud deposit types for BWRs; tightly adherent or loose layers of Fe_2O_3 or both. The adherent layer is attached to the cladding oxide, which in the case of zirconium oxide is essentially a permanent part of the cladding. Two tightly adherent, dense cruds have been identified: one a mix of Fe_2O_3 and Fe_3O_4 , the second, a mix of Fe_2O_3 and copper.

Anstine and Naughton (1981) point out that significant amounts of elements other than iron are present in some BWR deposits. These elements--Co, Ni, Cu--are generally at higher concentrations in the tenacious inner layer. Berry and Diegle (1979) listed Oskarsham-1 BWR two-layer data:

	<u>Fe, wt%</u>	<u>Cu, wt%</u>
Loose layer	68 - 76	3.9 - 9.0
Adherent layer	68 - 76	8.7 - 2.3

These data are for fuel having 26,000 EFPH (effective full power hours) and a deposit range from 2 to 60 mg/dm². The inner layer deposits can constitute about one-half of the quantity of BWR fuel deposits. Berry reported the loose layer to comprise more than 50% of the total for Oskarsham; Strasser, Sheppard, and Santucci (1985) indicated the loose layer to consist of about

one-half the total deposit at Monticello; Niki et al. (1985) stated that the inner layer deposits are 60% of the total at the Shimane nuclear power station.

Lin et al. (1981), commenting on the two deposit layers, said that the outer, loose oxide layer can be removed from fuel rod surfaces with a nylon brush, but that the inner, tenacious layer required a silicon carbide stone for removal. Experimental data indicate that the relative thickness of the inner, tenacious layer is greater in reactors using filter-demineralizer condensate treatment systems, and the Co/Fe ratio in the inner layer is also higher in those plants.

In early BWRs, use of copper-alloy heaters in the feedwater system led to introduction of copper and nickel at about the same concentration as iron to the primary coolant system. The spinels with these metals formed as tenacious deposits on fuel cladding surfaces (Blok, Sawochka, and Snyder 1973). The amount of copper and nickel has been substantially diminished and now deposits are about 95% iron, largely as α -Fe₂O₃, in a flocculent, loosely adherent deposit. This deposit is usually very porous and can have numerous boiling channels (also called chimneys or wicks) about 2-4 μ m in diameter, which allow easy interchange of steam and water at the fuel-rod surface (Lunde 1975).

Spalling, flaking, and patchy crud has often been reported. A recent case described by Fejes, Ivars, and Svensson (1984) occurred at Ringhals 1. In 1980, the crud deposition rate was observed to be increasing, and, because of the relatively high copper in the feedwater, the copper concentration in fuel-deposited crud rose to above 50%. Heavy spalling of the crud and the underlying ZrO₂ was observed on some rods. The problem was overcome, subsequently, by efficient removal of copper in the condensate cleanup system and by retubing the turbine condenser with titanium tubes.

The CILC crud (Marlowe et al. 1985, and Baily, Marlowe, and Proebstle 1985) discussed earlier was tightly adherent, of high density, with low heat-transfer capability, and composed of Fe₂O₃ and CuO with copper at 50% of the total cation content rather than the typically flocculent Fe₂O₃ crud. CILC has been observed to cause failure in only a small fraction of fuel rods in BWR service. Improvements in plant water chemistry and cleanup systems have

eliminated high concentrations of copper in reactor water and subsequent CILC failures.

Adhesive Strength of Crud

It is apparent from review of the literature that the adhesive strength of crud, as measured by the shear force required for removal, has not been determined quantitatively using suitable measuring devices. Thus, the best indication for adhesive strength is qualitative, and is based on the abrasiveness of the tool required to remove samples, namely:

soft, loose deposits	nylon brush (low abrasion)
hard, tenacious deposits	silicon carbide stone (high abrasion)

Strasser, Sheppard, and Santucci (1985) describe amounts of crud removed from fuel rods of two BWRs using these tools (Table 3). The crud of Nine Mile Point-1, being soft and loose, was easily dislodged by brushing. On the other hand, the crud of Monticello was two-layered and required use of both tools; the ratio of deposit weights was 50%/50%.

Lack of quantitative measurements of crud adhesive strength also may be due to probable difficulties similar to those that arise in taking crud samples. Solomon and Roesmer (1976) describe the tool and technique used by Westinghouse to remove a sample of crud from a rod of a spent fuel assembly. First, a crane is used to suspend a fuel assembly in a reactor site spent fuel pit. A sampling tool attached to a 25-foot-long aluminum conduit is manually held to contact the rod to be sampled and is moved along the rod between two grid spans. In this way, loosened crud is gathered. Ten traverses are made to collect one crud sample.

Particle Size

Due to possible release of small particles as a radioactivity-containing aerosol, the particle size of crud is of interest during any handling, shipment, wet or dry storage of spent fuel having deposits. The source of fuel crud is the reactor coolant. Both soluble and particulate crud are transported in the coolant with typical concentrations of about 10-20 ppb for both PWRs and BWRs (Berry and Diegle 1979). "Soluble" crud is generalized as being the portion which will pass through a 0.45 μm microporous filter and will include colloidal particulate material (Ponting

and Rodliffe 1983). Both soluble and particulate crud may deposit on fuel rod surfaces and agglomerate. Particle size and corrosion-product activity distribution appear to depend on reactor plant-operating conditions. Berry and Diegle (1979), commenting on Oconee 1 PWR coolant, said that only 23% of the corrosion product activity was filterable on a 0.45 μm filter in 1974, but 70% was filterable in 1975.

For loosely adherent crud and combined loosely adherent and scraped crud for fuel rods from Monticello Cycles 1 and 2, scanning electron microscopic (SEM) examinations (Strasser, Sheppard, and Santucci 1985) showed clusters of small particles in the 0.1 to 0.3 μm range and larger particles in the 0.5 to 2.0 μm range. SEM examinations of deposits on Nine Mile Point-1 fuel rods showed that the deposits consisted of agglomerates of about 0.1 μm particles.

Aerosols are microscopic particles in a size range of about 0.1 to 10 μm . The discrete crud particles easily are in this particle range, and if released, would have to be confined or controlled. The size of fuel crud particles that could become airborne is indicated qualitatively by a description of crud particle dispersal in storage-pool water during a fuel-rod consolidation demonstration (Rasmussen 1986). During the rod consolidation, crud was scraped off some rods and suspended in fuel pool water, forming a cloud. Visibility problems prevented continuation of operations until the small, cloud-forming particles had settled. Although discrete crud particles on fuel rods are small, they appear to be agglomerated, and may not be readily dispersed in air as individual particles.

CRUD RADIOACTIVITY

Crud scraped or spalled from fuel rods may be a source of increased radiological exposure to personnel during subsequent handling operations of fuel rods for consolidation, transfer to or from shipping casks, or storage under wet or dry conditions, or during facility or equipment maintenance. Because of its relatively large quantity and a half-life longer than that of radioisotopes present in fuel crud, cobalt-60 is the primary source^(a) for

(a) Fission products such as cesium-137 from breached fuel rods may be present, but these are not considered to be crud.

radiation from crud on fuel after long storage periods. Blok, Sawochka, and Snyder (1973) found that after several weeks' decay, cobalt-60 represented about 75% of the activity of fuel rod crud deposits. The activity decay of radioisotopes in fuel crud, which is potential interest to fuel handling operations, is shown in Figure 1. A list of the more important radioisotopes commonly found in fuel crud and their half-lives are in Table 6.

Like crud areal density and thickness, the inventories of radioisotopes present in crud on fuel rods is highly variable. Bergmann, Roesmer, and Perone (1983) have presented typical characteristics for PWR crud, including radioactivity of the two principal radionuclides after two cycles of operation of a PWR; their data are in Table 1. Berry and Diegle (1979) give fuel crud activity for two BWRs, one using a deep bed polishing unit in the condensate cleanup system, the other using a powdered-resin polishing system. Data for 12,000 EFPH reactor operation are in Table 7.

TABLE 6. Radioisotopes Commonly Found in PWR and BWR Fuel Crud and Their Half-Lives

<u>Radioisotope</u>	<u>Half-Life, Days</u>
51Cr	27.7
54Mn	312
58Co	70.8
59Fe	44.6
60Co	1924
65Zn	244

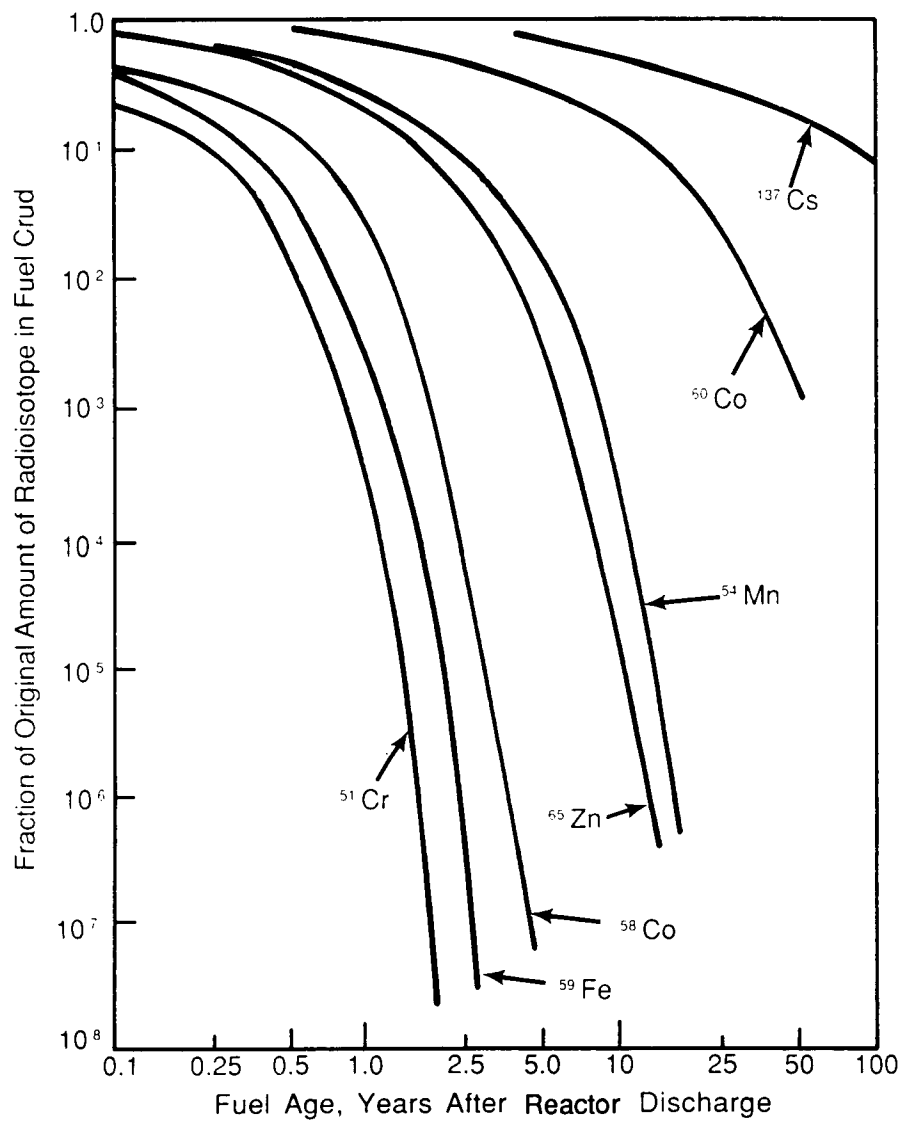


FIGURE 1. Activity Decay of Nuclides Found in Fuel Crud

TABLE 7. Corrosion Product Activities on BWR Fuel Rods

<u>Isotope</u>	<u>Condensate Cleanup System</u>	
	<u>Deep-Bed Polishing, $\mu\text{Ci}/\text{cm}^2$</u>	<u>Powdered- Polishing, $\mu\text{Ci}/\text{cm}^2$</u>
^{60}Co	180	110
^{65}Zn	15	210
^{58}Co	50	40
^{59}Fe	80	15
^{54}Mn	90	15

Einzigler and Cook (1984a) collected fuel-crud activity data from various sources. Some of this data for PWRs and BWRs is reproduced in Table 8. In addition, they performed radiation analyses of crud on BWR and PWR fuel rods used in their program, and back-calculated the amounts of radioisotopes present at reactor discharge of the fuel assemblies. Data for two principal fuel crud radioisotopes, shown in Table 9, illustrate the effect of nine years of decay following fuel discharge.

Crud radioactivity has ranged widely, as shown by the data in this section, but has diminished substantially as reactor operations and water chemistry have improved. The range given in Table 8 for cobalt-60 on PWR fuel is 0.1 to 140 $\mu\text{Ci}/\text{cm}^2$. The range for cobalt-60 on BWR fuel is 97 to 1250 $\mu\text{Ci}/\text{cm}^2$. The cobalt-60 data of Einzigler and Cook (1984b) for PWR and BWR fuel discharged in 1974 (Table 9) are at the lower ends of these ranges, a maximum of 19.3 $\mu\text{Ci}/\text{cm}^2$ for a PWR fuel rod and 10.6 $\mu\text{Ci}/\text{cm}^2$ for a BWR fuel rod. The maximum areal densities were 0.81 mg/cm^2 for the PWR rod and 0.21 mg/cm^2 for the BWR rod; these areal density data are also at the lower portions of the areal density data ranges.

These data indicate that crud on older spent fuel may have had higher radioactivities on reactor discharge than more recently discharged fuel having relatively little crud. As Figure 1 indicates, however, decay will have to be taken into account (see Figure 1) for any present-day handling of the older fuel.

TABLE 8. Activity of Fuel-Crud Radioisotopes

Reactor	Reactor Type	Sample Technique	Zero Time(a)	Fuel-Crud Radioisotopes, $\mu\text{Ci}/\text{cm}^2$					
				^{58}Co	^{60}Co	^{54}Mn	^{51}Cr	^{59}Fe	^{95}Zr
Yankee Rowe	PWR		Reactor Shutdown	1400	140	380	240	300	
B&W Reactors	PWR		Reactor Shutdown	2-400	0.4-100		0.7-170	0.1-30	0.4-1.0
Point Beach 1	PWR		Reactor Shutdown	0.7-13	0.1-2.0	0.1-2.0	0.9-16	0.1-2.0	
Beznau 1	PWR		Reactor Shutdown	0.9-215	0.1-16	0-13	1.6-391	0.1-25	
Zion 1-EDC-3	PWR	Steel Scraper	Reactor Shutdown	121±26 $\mu\text{Ci}/\text{mg}$	31±8 $\mu\text{Ci}/\text{mg}$	16±3 $\mu\text{Ci}/\text{mg}$	58±15 $\mu\text{Ci}/\text{mg}$.8±2 $\mu\text{Ci}/\text{mg}$	16±13 $\mu\text{Ci}/\text{mg}$
Tsuruga	BWR				140-1250	105-625			
Millstone 1	BWR	Brushed & Scraped		15-63	97-390	41-85	16-35	34-87	
Nine Mile Point	BWR	SiC Stone	Several weeks after reactor shutdown		~75% of activity				
Various GE reactors	BWR		Reactor Shutdown	40-50	110-180	15-90		15-80	

(a) Either measured at indicated time or corrected to reactor shutdown

Source: Einziger and Cook (1984a)

IMPACTS OF CRUD ON SPENT-FUEL HANDLING OPERATIONS

If crud is present in relatively large quantities, there is a potential for it to impact spent-fuel handling operations following reactor discharge. These subsequent operations may include wet storage, fuel-rod consolidation, transportation, dry storage, and equipment and facility maintenance. There is little quantitative data in the literature for crud release and its impact during these operations. Some descriptive information, however, is available and is summarized below.

WET STORAGE

Zima (1978) concluded that public safety implications of fuel crud are minimal for water storage of spent fuel at an independent spent-fuel storage installation (ISFSI). Fuel assemblies must survive reactor site handling, storage, transfer, and cleanup before being placed in an ISFSI water pool (away from the reactor). However, compared with radioactivity contained within the fuel, crud deposited on fuel assemblies is a relatively minor source of radioactivity.

TABLE 9. Radiation Analysis of Crud on Fuel Rods of a PWR and a BWR

<u>Fuel Rod Locations and Dates</u>	<u>Fuel-Crud Radioisotopes(a), $\mu\text{Ci}/\text{cm}^2$</u>	
	<u>^{60}Co</u>	<u>^{54}Mn</u>
Peach Bottom (BWR)		
Rod PH-462-E6		
April 1983	1.17 - 4.24	7.3×10^{-4} - 1.75×10^{-2}
May 1974 Reactor Shutdown ^(b)	2.9 - 10.6	0.21 - 5.06
H.B. Robinson (PWR)		
Rod B0-5-H9		
April 1983	0.33 - 6.05	8.2×10^{-4} - 1.2×10^{-3}
May 1974 Reactor Shutdown ^(b)	1.0 - 19.3	1.03 - 1.50

(a) Isotopes from fuel also present: ^{125}Sb , ^{134}Cs , ^{137}Cs , ^{154}Eu

(b) Based on back-correction of isotopes observed April 1983. Other short-lived isotopes may have been present at time of reactor discharge of fuel.

(Source: Einziger and Cook 1984b)

Zima's findings continue to be appropriate. (a) Crud may be released from fuel while it is being delivered or during routine fuel operations. If released by bumping or jerking of the fuel, crud may form a temporary cloud, obscuring view of the pool floor, and interrupt pool operations. Nevertheless, it has little impact on pool water because of its low solubility, and because it readily settles and is easily cleaned up to satisfactory levels by system equipment.

Bailey and Johnson (1983) observed that crud layers on five BWR assemblies in a wet storage facility had changed in tenacity during five to eight years of storage; crud that appeared adherent in 1978 was flaky and had a tendency to peel by 1982. They noted that loosening and spallation would more likely occur if the assemblies have undergone dry shipment, dry handling, or dry sipping. They further noted that studies under DOE's Commercial Spent Fuel Management (CSFM) program indicate crud-loosening may be occurring at one or two other sites.

SPENT FUEL ROD CONSOLIDATION

Crud posed problems during the Duke Power Company - Westinghouse Electric Corporation rod consolidation demonstration program in late 1982 (Rasmussen 1986; Bailey 1985a). Spent fuel used in the program was discharged from Oconee-2 PWR in 1977. Equipment used in the demonstration was built to pull all 208 fuel rods from an assembly at the same time. During rod pulling, a large amount of crud was scraped from the fuel rods, as expected, and became temporarily suspended in the surrounding water due to inadequate filtration. Vision was obscured and operations ceased until the particles settled. Although annoying, the dispersion had no appreciable effect on radioactivity level or personnel exposure (Bailey 1985b).

A subsequent rod consolidation program performed at the West Valley Nuclear Services facility water pool for Rochester Gas and Electric Company avoided the crud release problem (Furierer 1986). Crud release was controlled and water clarity maintained by a shroud around the fuel assembly;

(a) Personal communication with E. E. Voiland, General Electric Co., Morris, Illinois, April 1987.

a pump and filter system was attached to the shroud. No detectable pool activity increase occurred.

There has been no experience with extended wet or dry storage of consolidated fuel rods, but problems are not expected (Bailey 1985a).

TRANSPORTATION

For full-scale dry storage tests (Creer et al. 1986) of the Castor-V/21 PWR Spent-Fuel Storage Cask at the Idaho National Engineering Laboratory (INEL), twenty-one spent fuel assemblies of standard 15x15 Westinghouse design were shipped dry by truck in Transnuclear TN-8L shipping casks from the Virginia Power Co. Surry nuclear power station. Seven shipments of three assemblies each were made, the first and the seventh in the same cask. Following these two shipments, crud and smear samples were collected at INEL from the sides and bottom of the TN-8L cask to determine whether crud spallation occurred during fuel shipment and handling operations. Analyses made at Lawrence Livermore National Laboratories showed that no large amounts of radioisotopes common to crud were found, and that no fission products were detected. In fact, the cask, which was cleaned between shipments, was cleaner after the seventh than the first shipment.

The CASTOR-V/21 storage cask was loaded with fuel assemblies in the hot shop of the INEL Test Area North (TAN) cask-testing facilities. Fuel assemblies were transferred through air between the shipping and storage casks. During the operation, minor contamination occurred to the personnel work platform between the shipping and storage casks in three of the seven shipments. The contamination was localized; cleanup took about four hours. Black-and-white video tapes and 35-mm color photographs were taken of the fuel before and after its shipment to INEL and after other INEL operations. Although the photographs had insufficient detail to fully characterize crud on fuel rods, no noticeable changes were revealed in characteristics or adherence of crud resulting from shipping or INEL handling operations. In general, the fuel rods were in excellent condition, with a very adherent crud layer (Creer et al. 1986).

Dry storage performance tests are also being conducted with the Transnuclear, Inc., TN-24P PWR Spent Fuel Storage Cask at INEL (McKinnon et

al. 1987). This cask was loaded with 24 PWR spent fuel assemblies shipped from the Surry nuclear power station in TN-8L shipping casks. As with loading of the Castor-V/21 cask, minor contamination occurred to the personnel work platform between the casks. Again, contamination spread was not a major problem. Analyses of crud and smear samples taken from the TN-8L cask used for both the first and final spent fuel shipments showed an increase in the buildup of crud in the bottom of the cask during the shipping campaign. In contrast, the amount of crud on the cask wall decreased.

Johnson^(a) made some observations and deduced possible causes and impacts for crud released from long-term water-stored BWR spent fuel assemblies following their shipment from West Valley Nuclear Services (WVNS) to the Oyster Creek (OC) Nuclear Power Plant storage pool and return of the shipping cask to WVNS. The cask was dried before shipping. In some cases, a reddish crud cloud formed on opening the shipping cask underwater at OC for removal of the fuel assemblies; no cloud, however, was previously seen in the WVNS pool when the fuel was inspected and prepared for shipment. Before return of the cask to WVNS, it was thoroughly flushed; but when it was reopened underwater at WVNS, crud again emerged, reddening a large area of the pool.

Johnson^(a) made further observations of the causes and impacts:

- Activity spikes, principally from ^{60}Co and ^{137}Cs , occurred when the returned casks were opened at WVNS. This contamination could lead to increased activity in the "bathtub ring" on the pool wall.
- Before each shipment, the cask was drained to the pool and dried at 40 mbar (0.6 psia). Crud was drawn into the cask drain hose during the operation and raised radiation levels. Because connecting the hose is a hands-on operation, radiation to operating staff could have increased.
- The following are possible explanations for the crud phenomena:
 - Some of the normally tenacious part of the BWR Fe_2O_3 crud (sometimes 25 to 50 μm thick) may have loosened during extended wet storage.

(a) A. B. Johnson, Jr.: Trip report on visit to West Valley Nuclear Services, West Valley, New York, March 1985.

- Dry conditions and vibrations during shipping may have further loosened crud from fuel rods.
- Water ingress during cask refill also may have loosened some crud and redeposited it as a pasty layer on cask and fuel-basket surfaces. Only a fraction was removed by flushing.
- Partial dryout of crud and vibration during the cask return trip to WVNS may have loosened crud from cask and basket surfaces.
- Water flooding of the cask on its placement into the WVNS pool and turbulence in the cask when another set of fuel assemblies was inserted may have suspended crud, causing the subsequent turbidity in the pool.

The cloud of crud was most severe with the first shipment of fuel to OC (out of a total of about 30); lesser amounts of crud were noted in the other shipments(a). In the case of the first shipment, the cask (TN-9) loaded with fuel sat for several weeks with the fuel in a dry environment before it was shipped. Subsequent shipments were shipped more promptly and a better flushing technique was used on the casks. At WVNS, a hydrolasing technique was tried on the OC fuel, but the crud did not come off easily. No quantitative data on crud release from these shipments is available. The qualitative information and possible impacts presented for a specific situation may, however, be representative for other spent fuel having long-term water storage followed by dry shipping.

DRY STORAGE

Westinghouse Hanford Co. and EG&G Idaho, Inc., have performed a long-term, low-temperature, single-rod test program using four PWR and four BWR spent fuel rods to determine the performance of the rods under a variety of possible dry storage conditions (Einziger and Cook 1984a,b). Crud along the length of a sibling rod of the PWR rods used in the exposure test had an areal density range of 7 to 80 mg/dm² and a thickness of 0.76 to 7.6 μ m. Little variation in crud deposition was observed among the PWR rods. The ranges for crud areal density and thickness along the length of a BWR sibling

(a) PNL memorandum: W. J. Bailey to J. M. Creer, May 13, 1987.

rod were respectively, 0 to 21 mg/dm² and 0 to 1.8 μm. The rods were heated in inert gas or air atmospheres in individual capsule tests in an electric furnace to maintain the temperature at 229°C. After an exposure of 5962 hours, only one rod had visual or dimensional indications of rod degradation. This was a BWR rod with deliberately defected cladding exposed to air. Little crud had spalled from the rod and little fuel fell from a crack. Less than 0.1% of the materials released to the capsule became airborne in the respirable range of 2 to 15 μm. Einziger and Cook (1984a) concluded that the behavior of the fuel used in their test program should be representative of a large portion of the spent fuel population. They also concluded that crud spallation during dry storage should be a manageable problem.

Another test of fuel stored in air was conducted at the Fuel Temperature Test Facility (FTT) at the Engine Maintenance and Dissassembly (EMAD) site in Nevada (Johnson, Gilbert, and Dobbins 1986). Turkey Point (PWR) Assembly B02 was used in the testing. This assembly had previously been stored in water pools, shipped a few times, and stored dry. The maximum cladding temperature reached in the previous handling was <150°C during transit and vault or silo storage. The maximum cladding temperature in the 24-month air storage test in the EMAD FTT was 275°C; temperature decreased to 220°C. No unusual appearance of the fuel rods was observed before the FTT tests. After the FTT air storage, the crud on the Turkey Point B02 assembly was observed, through hot cell windows, to have taken on a reddish tinge. This was presumed due to formation of hematite (Fe₂O₃) by reaction of the spinels (e.g., NiFe₂O₄) with O₂. No other unusual appearance was observed.

Performance tests were conducted on a Ridihalgh, Eggers & Associates REA 2023 BWR spent fuel storage cask using BWR spent fuel assemblies from Nebraska Public Power District's Copper Nuclear Station (McKinnon et al. 1986). The cask was tested under vacuum, backfilled with helium, and backfilled with nitrogen. The maximum fuel-cladding temperature reached in the tests conducted for 4½ months was 241°C. There were no noticeable changes in characteristics to the very adherent reddish brown crud as a result of the dry storage test or handling operations.

Spent fuel dry storage tests have also been performed in the Federal Republic of Germany (Peehs, Bokelmann, and Fleisch 1986). The discharge burnup of Kraftwerk Union (KWU) fuel rods and assemblies tested was about

40 to 45 Gwd/tU. The cladding oxide layer thicknesses on fuel rods at this burnup are in the range of 50 to 100 μm for PWRs and 30 to 70 μm for BWRs. Crud deposition normally is small for KWU BWRs and practically zero for KWU PWRs. Single rod tests, conducted in an oxygen-free system, ranged up to 450°C for a storage duration of 16.5 months. The maximum temperature for fuel-assembly tests reached 300°C; after sixty days, when the run was ended, the temperature had fallen to 270°C. Fuel assembly tests were carried out in a specially-designed dry storage box in a reactor pool. Crud and oxide layers proved to be stable and adherent during all experiments and demonstration storage tests. In no cases was spallation observed.

GENERAL OBSERVATIONS

It appears from wet storage, rod consolidation, transportation, and dry storage evaluations that crud spallation, when it occurs, can be managed effectively, posing no significant radiological problems.

Crud quantities on PWR rods used in the dry storage tests are about average or at the lower end of the typical deposit quantity range. The quantities of crud on BWR rods used in these tests, on the other hand, were in the lower range for a typical BWR. If the amount of crud had been at the upper portions of these ranges instead (as it may be on the oldest stored spent fuel rods), more spallation might have occurred.

Bailey et al. (1983) have noted that, in general, experience with transportation and subsequent handling of over 5100 LWR fuel assemblies at reprocessing and/or storage facilities did not involve any uncontrolled release of significant amounts of volatile, aerosol-borne, or other solid particulate radionuclides.

AN APPROACH TO CLASSIFYING CRUD TYPES AND REVEALING THEIR POTENTIAL CONSEQUENCES

As shown previously, crud occurs in a broad range of amounts and compositions. Based on his personal observations over many years, Johnson^(a) suggests a generalized approach to classify the crud types and to reveal their potential consequences. The approach covers handling PWR or BWR spent fuel in wet storage, shipment, or dry storage. The intent is to provide an improved focus for a rather diverse subject.

Five classes of PWR and BWR fuel crud given in Table 10 are proposed. Class I involves a thick, hard copper oxide crud, principally CuO with embedded particles of magnetite. This crud class caused early fuel cladding failures in Big Rock Point (BRP) and the Steam Generating Heavy Water Reactor (SGHWR) at Winfrith, U.K. Replacement of copper alloy heat exchangers with stainless steel has essentially eliminated Class I crud. Most of the BRP fuel assemblies were reprocessed at the West Valley, New York facility, but about 85 of the BRP assemblies currently remain in wet storage at West Valley. These assemblies are destined for dry storage. Based on discussions with staff members of the shipper of fuel from West Valley and the receiving utilities, crud on certain fuel stored at West Valley appeared to have loosened from fuel during about fifteen years of wet storage. The condition of crud on BRP fuel, which has been stored since 1968, is not yet determined because the fuel has yet to be handled for shipment.

Class II is the general case for BWR crud, involving three subclasses: 1) a duplex layer of flocculent reddish brown Fe₂O₃ and a tenacious, inner layer; 2) a single layer, largely reddish brown Fe₂O₃; and 3) a thin, sometimes darker layer of Fe₂O₃.

Class III crud represents the crud-induced localized corrosion (CILC) case. The crud is mostly Fe₂O₃ with areas of CuO. The CILC crud has resulted in a small number of fuel failures. Reactors having this class of crud include Hatch 1 and 2, Vermont Yankee, and Browns Ferry. Exposure of

(a) A. B. Johnson, Jr., Pacific Northwest Laboratory, in a personal communication.

TABLE 10. Water-Reactor Crud Classifications

<u>Class</u>	<u>Reactor Type</u>	<u>Crud Compositions</u>	<u>Crud Thickness, μm</u>	<u>Comments</u>
I	Early BWR Early SGHWR	$\text{CuO} + \text{Fe}$ Oxides	25 to 175	Primarily limited to early Big Rock Point & SGHWR fuel
II	General BWR and Current SGHWR	Fe_2O_3	1 to 100	Frequently occurs as loose outer layer and tenacious inner layer
III	Selected BWRs	Fe_2O_3	--	Occurs on fuel at plants with copper alloy condensers and powdered resin demineralizer systems
IV	Early PWRs	$\text{Ni}_x\text{Fe}_{3-x}\text{O}_4$	0 to 85	Crud effects probably a function of thickness, not composition
V	Current PWRs Current PHWR	$\text{Ni}_x\text{Fe}_{3-x}\text{O}_4$	0 to 10	

spent fuel rods of this class has been limited mostly to wet storage; only a few rods have been shipped dry.

Class IV crud represents early PWRs. This class includes Turkey Point, Ginna, H. B. Robinson, Point Beach, and Oconee.

Class V covers recent PWRs and BWRs, such as those of the Federal Republic of Germany (FRG). Little crud has been found on fuel rods of these reactors and no crud problems have been observed.

Observations of the effects of wet storage, dry shipment, and interim dry storage on the five classes of crud are shown schematically in Table 11. In some cases loosening of crud has been observed in wet storage; in other cases crud has remained adherent even during dry storage.

Based on his personal observations, Johnson said that there have been very few cases where crud has caused any impact in over twenty-seven years in which nuclear fuel has been handled or shipped. There have been no incidents of serious impact.

Johnson has conceived a means to illustrate observations of the effects of wet storage, dry shipment, and dry storage on the adherence of fuel crud of each of the five classes. The concept is illustrated by Table 11.

First, the table gives the five classes of crud, reactor type, and the number of fuel cycles for fuel assemblies observed. Second, the table gives wet storage duration and comments on observations made on adherence of crud. A time line with an arrow shows the approximate number of years that fuel assemblies have been in wet storage (e.g., Big Rock Point fuel has been in wet storage up to 18 years; Cooper fuel assemblies observed, 41 months). On each time line is a reference to a footnote that gives further details regarding a particular fuel. Below the time line a brief comment is given on the observed adherence of the crud or whether the condition is unknown. In some instances, more than one time line is given for a crud class. This is because there is experience with fuel from different reactors having the same class of crud on the fuel. Third, a column is presented to show any observed effects of dry shipment; the time line shows simply that dry shipment occurred. Lastly, observations of the effects of dry storage are given. The time line shows whether fuel was stored for a few months or about two years. Observed crud adherence and dry storage temperatures are given.

Knowledge is still partial about adherence of crud under wet or dry storage or dry shipment. As more information is obtained from observations, it can be added to the table to improve its usefulness.

TABLE 11. Case Studies of Fuel Crud Observations

Class	Reactor Type	Fuel Cycles	Wet Storage At and Away from Reactor			Dry Shipment	Interim Dry Storage	
			To 5 yr	5 To 10 yr	Over 10 yr		Few Months	Up to 2 yr
I	BWR	1 to 3			(a)*->			
II	BWR	3 to 5	(b)-> Adherent	Condition unknown			(b) Adherent; 241°C	
III	BWR	--	Adherent (d)		(c)> Loose(?)	Loose(?)		
IV	PWR	--	Condition unknown for 7 to 8 year storage					
V	BWR and PWR (FRG)	--	(e)->		Adherent		(e)->	Fairly Adherent; 275°C
			(f)->					
			(g)->		Some loosening at 12 to 15 years			
			(h)->			Adherent		352 to 405°C
			(i)->			Adherent(?)		Unknown
			Adherent					200 to 450°C

*See footnotes following

Footnotes to Table 11

- (a) Big Rock Point fuel in wet storage up to 18 years; present crud condition is unknown.
- (b) Cooper fuel - There was no substantial loss of adherence after a series of fuel transfers and little spalling; fuel stored up to 41 months in water.
- (c) Oyster Creek - Wet storage at reactor, wet shipment to West Valley, wet storage at West Valley 12 to 14 years. Not much crud lost at West Valley. Dry shipment to Oyster Creek. Empty cask returned to West Valley. Crud was released from the cask into the pool to give a "tomato-juice-appearing cloud."
- (d) Experience just wet storage.
- (e) Turkey Point fuel discharged early 1970s. Not much crud. Stored dry at the Engine Maintenance and Disassembly (EMAD) facility of the Nevada Test Site for several years in several modes. Crud remained fairly adherent following exposure at 275°C.
- (f) Early PWRs: Ginna and Point Beach - at reactor storage, wet shipment to West Valley, wet storage 12 to 15 years at West Valley, dry shipment to reactor, wet storage at reactor. In one case some evidence of crud loosening observed after fuel placed back into reactor pool.
- (g) Early PWR: Oconee - Involved 8 years of wet storage in reactor pool; crud primarily adherent, but some evidence of flaking or loosening observed.
- (h) Surry - Adherent during at-water reactor storage and dry shipment. Peak clad temperature 352 to 405°C, effect unknown.
- (i) FRG fuel stored at reactor in pool, shipped dry to a hot cell, and rods stored dry at 200 to 450°C for 1 to 1½ years.

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