CONF-750902--12

SOLID WASTE GENERATION

IN

REPROCESSING NUCLEAR FUEL

80th National Meeting
American Institute of Chemical Engineers
Boston, Massachusetts

September 8, 1975

Presented By

E. D. NORTH

NUCLEAR FUEL SERVICES, INC.
6000 Executive Boulevard
Rockville, Maryland 20852

SOLID WASTE GENERATION
IN
REPROCESSING NUCLEAR FUEL

Reprocessing of nuclear fuel has been practiced for many years in this country and abroad. The wastes generated in the reprocessing operation have only recently been given much attention and this has principally come from the anti-nuclear people. The waste of greatest concern, one that has been adequately defined, is the high level waste containing better than 99% of the fission products from the spent fuel. This waste is self heating, and highly radioactive. Under the provisions of Appendix F, 10 C.F.R. 50, high level waste may be stored as a liquid for five years and then must be solidified and shipped to a Federal Repository within 10 years after generation. High level waste is not the major subject of discussion for this afternoon.

The subject for this session is all the other wastes from a reprocessing plant, and there is a lot of it, which must also be disposed of in an entirely safe manner for the protection of the public, both for the present and in the future.

These other wastes have not been the subject of specific regulations, so that a present concern is that engineering plans may be changed significantly by future regulations. There are various estimates of the quantities to be generated, but insufficient study has been made of the various sources, types, and levels of contamination to be expected in these wastes.

NFS Reprocessing Plant

NFS built the West Valley plant from 1963 to 1966 and operated the plant for six years during which time about 600 tonnes of fuel were processed. These fuels ranged from low burnup NPR fuel to high burnup (32,000 MWD/T) Yankee Rowe Fuel. NFS also operates a low level waste burial area for the State of New York in which low level radioactive waste generated on-site and waste from other off-site sources has been buried. A great amount of information has been accumulated over the years in handling and disposing of these wastes.

The plant as originally constructed was designed for a nominal one tonne of fuel per day. In 1972, the plant was shut down for modification and removal of some bottlenecks in the process which were limiting the throughput. When the modification program has been completed, the capacity will be increased to 750 tonnes per year. The data presented in this paper are based upon that annual production rate.

As a part of the modification program, NFS will build a plant to convert uranyl nitrate solution to uranium hexafluoride. A unit will be installed to convert plutonium nitrate solution to plutonium oxide, and in addition, a waste solidification facility will be constructed to calcine high level waste to a solid form for eventual shipment to the Federal Repository.

Each of these units will, of course, generate solid wastes which must be handled, packaged, consolidated and finally be disposed of in a safe and secure manner.

Table I shows the estimated annual generation of solid wastes from a 750 tonne per year plant. It can be seen at once that the wastes vary greatly in quantity and contamination level and that the total volume per year is quite sizable -- 111,000 cubic feet per year as generated and nearly 72,000 cubic feet per year after treatment and compaction.

Where are these wastes from, what are their characteristics, and what can be done with them?

Solid Waste Generation

High Level Waste

Column 1 covers the high level waste, undoubtedly the most difficult technical problem in the lot, but only a small proportion, volume wise, of the overall solid waste problem. It is estimated that there will be some 1400 cubic feet per year or 1.3% of the total waste generated and which after calcining and dilution will comprise some 2800 cubic feet per year. This is still only 3.9% of the final solid wastes.

Hulls

Column 2 shows data on the hulls. This material includes the end pieces and other hardware associated with the fuel elements. The hulls, while not self heating and not in the same class as high level waste, present a difficult radiation problem. They are highly radioactive, primarily from the induced cobalt-60 formed during exposure in the reactor. The hulls will be washed following removal from the dissolver to remove soluble uranium, plutonium and fission products. They will then be monitored for SNM content and will be placed in specially designed containers about four feet in diameter and four feet high. A shielded storage facility will be constructed for short term storage, but it is intended that hulls will be shipped off-site or otherwise removed from the plant as soon after separation as is possible. What is needed is a definition of the container dimensions and activity levels which ERDA will accept at a Federal Repository. With the short half life of cobalt-60, the former practice of local burial may well be perfectly adequate.

Intermediate Level Waste (ILW)

The next three columns - 3, 4, and 5 - highlight a group of wastes normally called intermediate level waste. The waste is primarily sodium nitrate from the solvent clean-up operation, but there is also sand and filter aid from fuel pool cleanup and spent resins from various operations such as low level liquid waste treatment plant. The high sodium content makes these wastes unsuitable for inclusion with high level waste since it is possible that these high level wastes may eventually be converted to glass, and the glass formulations presently in use will accept only a limited quantity of sodium. In any case, the sodium content must be complexed by some means such as calcium or aluminum before the wastes are calcined. The addition of complexing agents will thus increase the total volume and exacerbate an already difficult problem.

The intermediate level waste also includes mercuric iodate from the iodine recovery process and both the mercury and the iodine will volatilize in the calcining operation to present a difficult off-gas clean-up problem.

Mercuric iodate is almost as bad from a chemical standpoint as it is regarding radioactivity. The tentative EPA limit on mercury in fresh water is 2 parts per billion. A little mercury goes a long way! It may prove desirable to convert the mercuric iodate to sodium iodate and recycle the mercury to the scrubber column rather than sending it to the ILW system. The sodium iodate can be dried and packaged for disposal -- when it is decided where it has to go.

It is also expected that the intermediate level waste will contain a significant ruthenium content; another volatility problem with a hard gamma daughter product. NFS plans to neutralize these wastes and evaporate them to a dry sodium nitrate salt for disposal in containers similar to the hull container. The resins will be mixed with the filter aid and concrete and packaged in 55 gallon drums for disposal. The total volume is some 7,200 cubic feed or about 10% of the total solid waste.

Failed Equipment

Column 6 covers a problem that is easy to overlook -- failed equipment. The term "failed equipment" refers to those pieces of plant hardware that wear out in service. It does not connotate poorly designed or improperly installed process equipment. After all, a reprocessing plant represents a pretty hostile environment with strong acids and high radiation levels in the cells. It is not surprising, therefore, that repair or replacement of equipment is required from time to time. In the usual chemical plant, when equipment fails, the maintenance crew replaces the failed component and puts the item back into service. This is not the case in a reprocessing plant. The radiation levels on the failed item preclude the extensive decontamination required for contact work and in the decontamination process extensive occupational radiation exposure would be incurred. To avoid the occupational exposure, it is less expensive and less time consuming to remove the failed item entirely and replace it with a new item. The failed item is decontaminated by washing and is then packaged and buried on site.

Over the years, as would be expected, various items have failed and it is expected that they will continue to fail in future operations.

Both of the original dissolvers developed leaks over a five year period, probably due to stress corrosion set up by thermal cycling in normal operation. The design was modified in the replacement dissolvers to minimize the thermal stresses, but insufficient experience has been gained using the new design to determine that the correction is fully effective. Some of these items are of significant size and the occupational radiation exposure that would be incurred in dismantling and packaging for off-site disposition would be unacceptable.

Tube bundles in evaporators, condensors and factionators also have developed leaks and have been replaced.

Pumps, valves, tools, and MSM parts also have failed from time to time. These have been removed, washed, and packaged for on-site burial. The plant was, of course, designed to handle such equipment failures. Special provisions have been made to permit remote removal and decontamination of failed equipment.

The total volume of this waste is estimated to be about 10,000 cubic feet per year before packaging and 15,000 cubic feet per year when packaged for burial. Failed equipment is estimated to be about 22% of the total waste volume.

High Efficiency Particulate Absorbers

Column 7 covers HEPA filters. The volume is relatively small to begin with, 2000 cubic feet per year. It is estimated that these can be compressed to 500 cubic feet and disposed of in 55 gallon drums. Some 75% of the filters will show radiation levels of from 1-500 mR/hr gross beta and will not have been exposed to significant plutonium contamination.

The other 25% will originate in the plutonium oxide facility and will have lower or negligible gross beta, but will contain significant plutonium contamination at about 5 mCi Pu per gram. These filters will have to be very carefully handled in a glove box environment for compaction and packaging for disposal.

Spent Solvent

Column 8 shows the expected volume of spent solvent to be some 200 cubic feet per year. While the volume is small, the disposal problem is not insignificant. The present plans are to burn this material, but anybody who has ever tried to burn TBP -- kerosene knows that a heavy black smoke will surely be developed in copious quantities. This will surely plug the off-gas filters and will increase the quantity of filters to be disposed of.

An alternate method of disposal is distillation and recovery. This is not a highly developed technology and further work remains before this solution can be demonstrated.

The most simple approach would seem to be the one that has been used to date -- absorption on a porous solid such as vermiculite followed by burial.

Alpha Contaminated Combustible Waste

Column 9 brings us to a very troublesome waste - alpha contaminated combustibles such as paper, cloth, rubber gloves, plastic and miscellaneous trash. A significant portion of this will originate in the process areas in the plutonium oxide facility. It is estimated that some 17,000 cubic feet per year will be generated.

There are several methods available for reduction of this volume. One is incineration with all its attendant problems, not the least of which is filter plugging. The treatment here may be worse than the disease!

A second method which is being developed is acid digestion using nitric acid as an oxidant in hot sulfuric acid. The presence of PVC develops hydrochloric acid, thus giving a serious problem with materials of construction. The present work is being done in glass which must be considered somewhat less than desirable in handling plutonium. Corrosion studies are underway which indicate that tantalum shows excellent corrosion resistance, however, fabrication and costs present problems. Duriron and durichlor are marginally acceptable.

Atomics International has developed a molten salt process that merits serious consideration. In this process the waste is ground in a hammermill and blown into a pool of molten sodium carbonate at 750°C-1000°C. The alkaline sodium carbonate does not cause any corrosion problems. The waste is converted to CO_2 and water, with plutonium, uranium, chlorides, etc. being retained in the salt. The salt can be discarded as is or, alternatively, can be dissolved for recovery of plutonium. This process will be discussed in detail in a paper to be delivered at a later session during this meeting.

Low Specific Activity Waste (LSA)

The next three columns present data on about half of the total solid waste from the reprocessing plant. These wastes are of low specific activity in that they are less than 200 mR/hr and they contain little or no plutonium. These wastes originate in various parts of the plant where plutonium is either not present, or if present, is at such low levels that the resulting plutonium content of the waste is not of significance.

These wastes are the uniforms from operators who remain outside of contaminated areas, waste paper, plastic, calcium fluoride sludge from the uranium hexafluoride facility, neutralized regeneration acid from the low level liquid waste treatment plant, and other assorted trash of all kinds.

The combustibles will be reduced in volume by compaction, or incineration and will be packaged in 55 gallon drums for disposal. The calcium fluoride sludge will be packaged in 55 gallon drums either as is, 50% dry solids, or can be dried prior to packaging. The neutralized regeneration acid will be dried or adsorbed on vermiculite for disposal in drums.

The sum total of all of these wastes from reprocessing 750 tonnes per day is 111,000 cubic feet as generated, and 72,000 cubic feet per year after compaction and further treatment. This is a lot of waste! There may be some errors in the estimates, but the values are reasonable based upon NFS' experience.

Activity Level of the Various Wastes

Figure 1 shows the data from Table I in thousands of cubic feet per year to be disposed of versus the activity level in curies per cubic foot. The activity level varies over seven orders of magnitude from high level waste at 10^5 or 10^6 curies per cubic foot down to low specific activity waste at 10^{-2} or less curies per cubic foot.

The solidified high level waste with its high activity level and heat generation rate is a special problem and is not the subject of this discussion for we are primarily concerned with the other wastes.

Hulls are an interesting case. The activity level is two or three orders of magnitude less than high level waste and they are not self heating. Moreover, the hull activity mainly comes from induced activity from cobalt-60 with a 5.26 year half life. In 53 years (ten half lives) the activity level will have decreased to the level of ILW salt.

The activity level of the failed equipment varies over a wide range -- from about 0.1 curie per cubic foot up to 8 or 10 curies per cubic foot. This results from the wide range in the types of equipment which have to be replaced and also from the difficulty in cleaning the equipment prior to disposal. When equipment such as pumps or valves can not be decontaminated easily, it is placed in drums and encased in concrete prior to disposal.

The low specific activity wastes are only slightly contaminated but are large in volume. The three generation rates are additive with all at about the same activity level. This class of waste comprises some 33,000 cubic feet per year or some 46% of the total solid wastes.

It can be seen that these wastes come from many different sources and thus present many different problems; some of which are severe, some of which are not. The important point is that there are major differences in the waste categories and that they should not all be lumped together for one method of disposal.

Waste can be, and should be, segregated on the basis of point of origin within the plant, and the method of disposal should be based upon the various characteristics of the segregated waste. Some will, of necessity, be sent to the Federal Repository, but clearly some do not warrant the added costs and occupational radiation exposures that would be incurred were the material to be shipped off-site.

TABLE 2

RADIOACTIVE WASTES GENERATED ANNUALLY AT NFS DESIGN RATE — 750 METRIC TONNES PER YEAR

	1	2	3	4	5	6	7	8	9
			Intermediate Level Waste						
	High Level Waste	Leached Hulls	Salts	Slurry	Resin	Equipment	HEPA Filters	Solvent	Alpha Combu
Original Quantity	1,400 ft^3	9,200 ft^3	4,800 ft^3	1,635 ft^3	730 ft^3	10,360 ft^3	2,000 ft^3	200 ft^3	17,0
Percent of Total	1.3	8.3	4.3	1.5	0.6	9.3	1.8	0.2	1
Physical Form	Solid	Solid	Slurry	Slurry	Solid	Solid	Solid	Liquid	Soli
Chemical Form	Calcine Oxide of MFP and Added Inerts	Zr Alloy or Stainless Steel	Sodium Nitrate 40% Solution + Mercuric Iodate	Filter Aid	Organic & Mineral Resins	Metal	General Dust on Glass Filter	Diluent TBP	Paper, Cloth, bur, tic,
Radioactivity (ft^3/Final Volume)	99.95% of Activity in Fuel. ~$1x10^5$ Ci of Mixed FP/ft^3	0.05% of Activity in Fuel. 500 Ci Co-60/ft^3	20-40 Ci of Mixed FP. 0.01 Ci I-129. <0.1 g Pu/ft^3	1-10 Ci/ft^3 MFP. <10 nCi Pu/g.	1-10 Ci/ft^3 MFP. <10 nCi Pu/g.	<1,000 mr/hr. <10 nCi Pu/g.	1-500 mr/hr <0.5 Ci 8/ft^3 75% <10 nCi, 25% ~5 mCi Pu/g.	>100 Ci/ft^3 MFP <10 nCi Pu/g.	~500 ~3 g
Process	Calcine	-	Dry	Dry	Concrete	Decon - Concrete as Necessary	Compaction	Burn or Distill	Comp
Shipping Container	1-1/2' x 15' Canisters	Steel Canisters	Steel Canisters	55 Gallon Drums	55 Gallon Drums	Special Container As Reqd.	Special Container or Drum	Mix with ILW Waste	55 Ga Drums
Final Volumes	2,800 ft^3	9,200 ft^3	4,300 ft^3	1,470 ft^3	1,460 ft^3	15,500 ft^3	500 ft^3	10 ft^3	3,70
Percent of Total	3.9	12.8	6.0	2.0	2.0	21.6	0.7	0.0.	5.

TABLE 2

RADIOACTIVE WASTES GENERATED ANNUALLY AT NFS DESIGN RATE — 750 METRIC TONNES PER YEAR

6	7	8	9	10	11	12	
				Low Specific Activity Waste			ft^3
Equipment	HEPA Filters	Solvent	Alpha Waste Combustible	Combustible	Slurry	Salts	Total Volume
10,360 ft^3	2,000 ft^3	200 ft^3	17,000 ft^3	35,000 ft^3	23,300 ft^3	5,500	111,125
9.3	1.8	0.2	15.3	31.5	21.0	4.9	
Solid	Solid	Liquid	Solid	Solid	Slurry	Slurry	
Metal	General Dust on Glass Filter	Diluent TBP	Paper, Cloth, Rubber, Plastic, Misc.	Same	Water Treatment. Sludge, CaF_2	Sodium Nitrate 40% Sol.	
<1,000 mr/hr. <10 nCi Pu/g.	1-500 mr/hr <0.5 Ci β/ft^3 75% <10 nCi, 25% ~5 mCi Pu/g.	>100 Ci/ft^3 MFP <10 ~Ci Pu/g.	~500 mr/hr, ~3 g Pu/ft^3	<200 mr/hr <10 nCi Pu/g	<200 mr/hr <10 nCi Pu/g	<200 mr/1 <10 nCi Pu/g	
Decon - Concrete as Necessary	Compaction	Burn or Distill	Compaction	Compaction or Incineration	-	Dry or Absorb	
Special Container As Reqd.	Special Container or Drum	Mix with ILW Waste	55 Gallon Drums	55 Gallon Drums	55 Gallon Drums	55 Gallon Drums	
15,500 ft^3	500 ft^3	10 ft^3	3,700 ft^3	7,000 ft^3	20,700 ft^3	5,000	71,640
21.6	0.7	0.01	5.2	9.8	28.9	7.0	

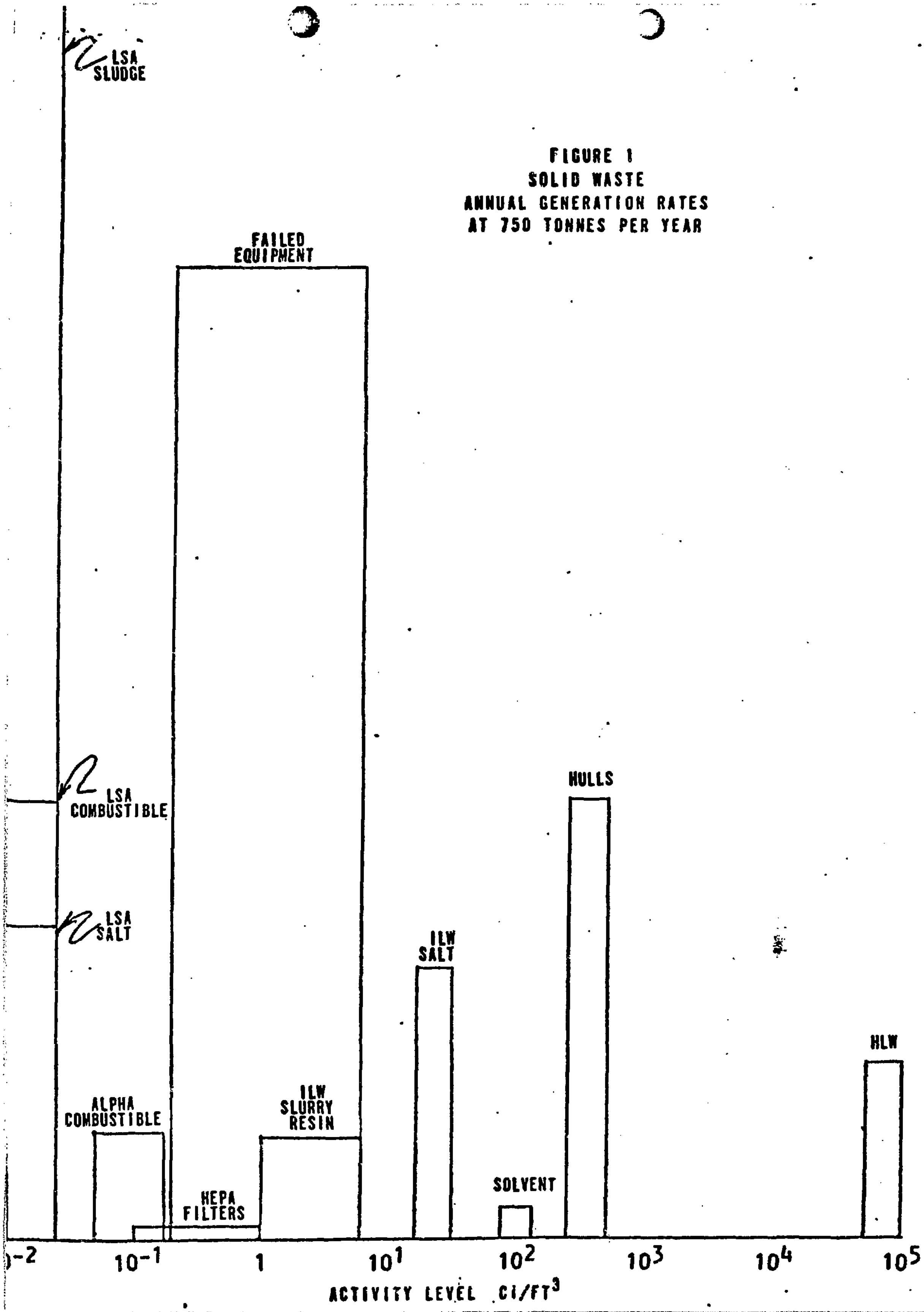
FIGURE 1
SOLID WASTE
ANNUAL GENERATION RATES
AT 750 TONNES PER YEAR
LSA SLUDGE
FAILED EQUIPMENT
LSA COMBUSTIBLE
HULLS
LSA SALT
ILW SALT
HLW
ALPHA COMBUSTIBLE
ILW SLURRY RESIN
SOLVENT
HEPA FILTERS
10^{-2}
10^{-1}
1
10^{1}
10^{2}
10^{3}
10^{4}
10^{5}
ACTIVITY LEVEL CI/FT^3