NUCLEAR REACTOR WASTES: DEVELOPMENT CONSIDERATIONS FOR A CONDITIONING PROGRAM FOR LONG TERM STORAGE OR DISPOSAL

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ABSTRACT

Present Ontario Hydro practices require reactor wastes to be stored in interim concrete storage facilities. The concrete structures provide the containment barriers required to prevent dispersion of the radioactive contents. A significant portion of the radionuclides have hazardous lifetimes in excess of the predicted storage structure life.

A conditioning program is described whose objective is to give all categories of reactor wastes satisfactory characteristics of: retrievability, stability, leachability, and minimum volume such that a transition from present interim storage to long-term storage or final disposal is possible. The development program (1977 - 1982) involves processes such as evaporation, reverse osmosis, incineration, compaction, crushing and immobilization in a system which considers the waste in an integrated fashion from the reactor to its final resting place.

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A Canadian Electric Utility with 4655 MWe of Operating Nuclear Capacity (February 1978).

INTRODUCTION

Ontario Hydro presently operates 4655 MWe of CANDU-PHW* reactors with a further 9310 MWe under design or construction. A full description of our present waste processing, handling and storage techniques can be found in references (1),(2), and (3). A brief summary of the system, sources, forms and storage practices now used is included here for completeness.

Other than wastes traditionally considered as 'decommissioning material', two types of radioactive products arise from the operation of a CANDU station:

(i) irradiated fuel;

and

(ii) contaminated wastes produced in the daily operation and maintenance of a nuclear station.

The program in Canada to develop and demonstrate a means of dealing with the irradiated fuel by-products is described in reference (4). The second category (ii) is normally referred to as 'reactor waste' and includes a wide variety of waste materials with a wide range of radioactivity concentrations.

Reactor wastes, their handling, processing and packaging for transport is the subject of this paper.

CURRENT REACTOR STATION PRACTICES

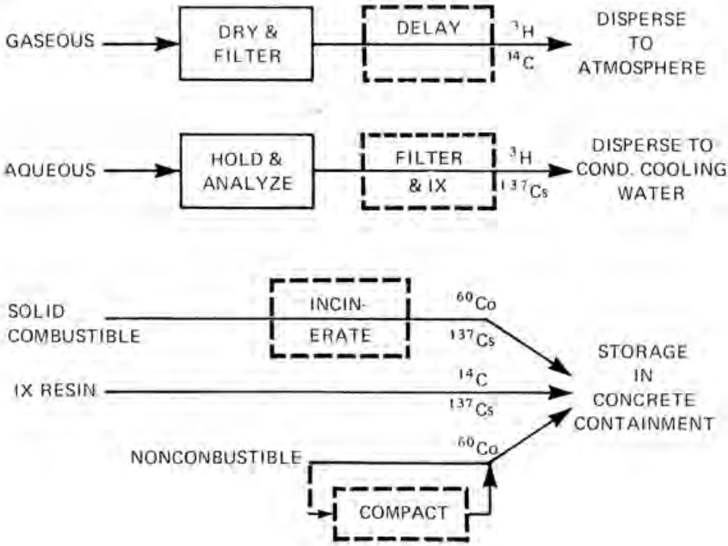
There are three forms of reactor wastes to be managed: liquid, gaseous and solid. (Fig. 1)

Liquid

Ontario Hydro CANDU reactors are indirect cycle, D_2O moderated and cooled pressurized water reactors. Due to the economic penalty associated with the loss of heavy water, a

* CANDU-PHW - Canada Deuterium Uranium - Pressurized Heavy Water

FIGURE 1
CURRENT REACTOR WASTE MANAGEMENT



great deal of effort is spent in all stages of design and operation to incorporate heavy water recovery and recycle systems into the normal operating cycle (area dryers, valve packing interspace leak-off connections, etc.). The separate moderator and heat transport systems are purified by individual ion exchange clean-up systems. The spent resins from these and other in-station purification systems are slurried to large in-station storage tanks. D_2O make-up to, and leakage from, the main systems are purified by ion exchange columns.

With on-power refuelling (natural Uranium Dioxide Fuel), the minimum excess reactivity requirement is met with minor shim concentration changes. Ion Exchange purification of the moderator system is used to effect these changes.

Due to the above design features and the excellent fuel integrity performance, the quantity and activity concentration of liquid waste streams have to date been low.

The liquid wastes are collected in holding tanks, analyzed, and then metered at an appropriate rate into the condenser cooling water for dilution and dispersion. At the present time there are no facilities at Pickering or Bruce Nuclear Generating Stations for the treatment of the liquid wastes. Under design, however, is a system using ion-exchange columns and filters to clean up occasional batches of waste which analysis has shown to have a higher than normal concentration of activity. The treatment system to be used at our next station is presently under review. The most significant nuclides in the effluent are tritium and cesium-137.

Presently, small liquid volumes of high-chemical content and relatively high activity content (i.e.decontamination wastes) are segregated from the above high-volume, low chemical/radioactivity wastes and immobilized using a solidification procedure in 200 ℓ drums.

Tritium in liquid wastes is more widespread at detectable concentrations in CANDU wastes than in LWR wastes. Routine measures to avoid loss of heavy water have been adequate to date to restrict tritium releases to a small fraction of the derived release limits.

An intensive program has been underway for the last 18 months to look at ways to limit internal doses to station staff and to limit tritium releases to the environment. A progress report on this work is the subject of a paper at this conference (5).

Gaseous

Gaseous wastes are dried to recover D_2O and associated tritium, filtered to remove radioiodines and particulates and then dispersed to the atmosphere. In addition, in the near future selected streams will also be passed through charcoal absorbers to delay the contained noble fission gases and allow most of them to decay before release. Once this modification is installed, the most important radionuclides remaining in the gaseous effluent will be tritium, carbon-14 and Argon 41.

In limiting the release of radioactivity from its nuclear generating stations to levels as low as reasonably achievable, Ontario Hydro has established a design and operating target of one percent (1%) of the Derived Release Limit (DRL) per 4 Unit Station for both gaseous and liquid radioactive effluent streams. To date radioactivity releases via the gaseous route have been consistently well below 1% DRL.

Solid

Solid reactor wastes can be classified into a number of categories, but for purposes of this paper I will refer only to combustible and non-combustible wastes, and ion exchange resins.

Low-level solid wastes are a mixture of contaminated paper and plastic trash plus discarded equipment and supplies. The majority is combustible and is packaged in clear polyethylene bags. Cobalt-60 (decay half-life = 5.3 years) and cesium-137 (half-life = 30 years) are the most important of the associated radionuclides. An incinerator is used to reduce the volume of this category of waste.

The non-combustible waste is a mixture of:

(1) combustible material which is either too active, corrosive or which may clog the incinerator;

(2) spent, disposable ion exchange columns;

(3) disposable filter cartridge columns;

(4) a variety of contaminated tools, reactor system components (valves, etc.);

(5) miscellaneous low-level activated or contaminated solid-waste materials.

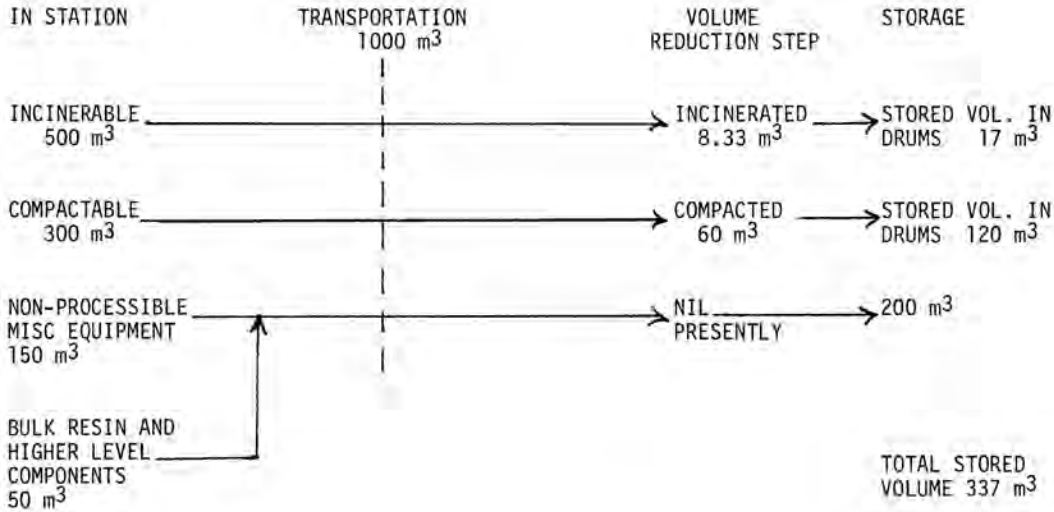
The non-combustible waste is presently packaged in LSA or Type 'A' containers, and stored either in containers or as loose pieces in storage structures.

Ion exchange resins are primarily mixed-bed, bead-form resins discharged from the purification systems of the heavy water moderator, reactor coolant and spent-fuel bay systems. The coolant purification resin is the major source of radioactivity in reactor wastes. Cesium-137 is the predominant radionuclide, with a transuranic content of less than 10 nCi/g (370 Bq/g). Moderator purification resin is essentially free of fission products, but contains major amounts of neutron-activation products (Co^{60}, Fe^{59}, Cr^{51}, etc.). The external radiation field is largely from cobalt-60, but a major radiological concern may be carbon-14 (half-life = 5700 years). Because a CANDU-PHW reactor (natural uranium) has a large static volume of moderator and reflector relative to a LWR, carbon-14 production from the neutron reaction with oxygen-17 in the water is much more important, giving about 500 Ci/GWe-year (18 TBq/GWe-a). The carbon-14 appears to accumulate on the ion-exchange resin as a carbonate or bicarbonate, and is discharged with the resin.

The present volumes of solid-reactor wastes produced at a 4 unit, 2000 MWe CANDU-PHW station is given in Table 1.

TABLE I

Existing Reactor Waste Categories
and Volumes (yr-1)-4 Unit, 2000 MWe



Storage

Ontario Hydro has in use or in final commissioning stages both in-ground and above-ground storage facilities. In-ground facilities have been used for a number of years, while the above-ground facilities are a more recent approach. (3)

Water, either in the form of precipitation surface or subsurface water, presents the greatest concern with respect to containment integrity and safe waste handling and storage operations. Elimination of water, with its leaching and carrier potential, would ensure negligible risk to the environment from these operations. A number of design concepts and operational approaches have been adopted to minimize any risk.

For in-ground facilities the dual envelope feature is provided by the use of reinforced concrete structures in conjunction with subsurface drainage and monitoring systems. The structures, which have an external asphalt coating for added protection, are placed on undisturbed grey till with a drainage system located under and around their lower perimeters. The drainage piping embedded in granular material ensures rapid draining of any water which might seep into the granular backfill around the structures from the adjacent natural till or from the ground surface. Any in-structure water that may escape to the subsurface as a result of structural failure would be intercepted by these systems. The various 'legs' of the drainage systems lead to collection manholes and then to sampling stations where water samples are automatically taken in proportion to flow. These samples, which are collected and analyzed weekly for tritium and gross beta/gamma activity, provide a record of the amount of radioactivity leaving the site in the subsurface drainage.

The dual-envelope feature for above-ground facilities is provided by the use of two, independent-structural, concrete barriers.

Currently, all solid-reactor waste from Ontario Hydro nuclear generating stations is placed in interim storage in concrete containment as described above. Since the hazardous life of much of the waste is expected to exceed the period for which the integrity of the containment can be guaranteed, this storage cannot be considered permanent. A co-ordinated development program is being carried out by Atomic Energy of Canada Ltd. (6) and Ontario Hydro, whose objective is to develop an integrated system to condition wastes at source (the reactor) or centrally, in order to produce a material form which can be easily retrieved and whose physical/chemical properties are adequate for final disposition.

DEVELOPING A NEW CONDITIONING PROGRAM

In order to plan for transition from an interim-storage approach to one in which the wastes may require direct or subsequent movement to a long-term storage structure or to permanent isolation, the factors required for each type of sturcture should be evaluated.

Current interim storage practices rely on engineering barriers to retain the radionuclides, backed up by favourable site characteristics which would attenuate the effect of any leakage from containment. Structural integrity is continually checked by monitoring. A fenced exclusion area serves to keep the public away from the wastes.

For permanent disposal, the aim is to assure isolation for periods longer than the hazardous life of the waste. This could be achieved by a combination of three impedances to migration.

A. All longer-lived wastes should be converted to a stable leach-resistant form.

B. The waste (A) should be emplaced in a way that protects it from moving groundwater.

C. A location should be chosen with characteristics that ensure that the pathways to man are long compared to the half lives of any migrating radionuclides.

Characteristics of Waste Form

One of the barriers to migration required in a system as described in "DEVELOPING A NEW CONDITIONING PROGRAM" is the form of the waste itself.

The form of the waste should be:

- stable, so that its characteristics are retained for at least the hazardous life of the waste.

- leach-resistant, so that it contributes a significant barrier to the migration of the associated radionuclides.

- of minimum volume, to lower final storage/disposal space requirements.

- in a form which is retrievable for transfer if required.

Methods under development for converting the waste to a stable form are outlined in Fig. 2. For all the conversion processes there are actually two steps in going from the original to final form: a <u>preconditioning step</u>, which normally includes volume reduction, to put the waste into a more stable and manageable form, followed by an <u>immobilization step</u> which provides a protective and consolidating envelope.

Preconditioning Steps

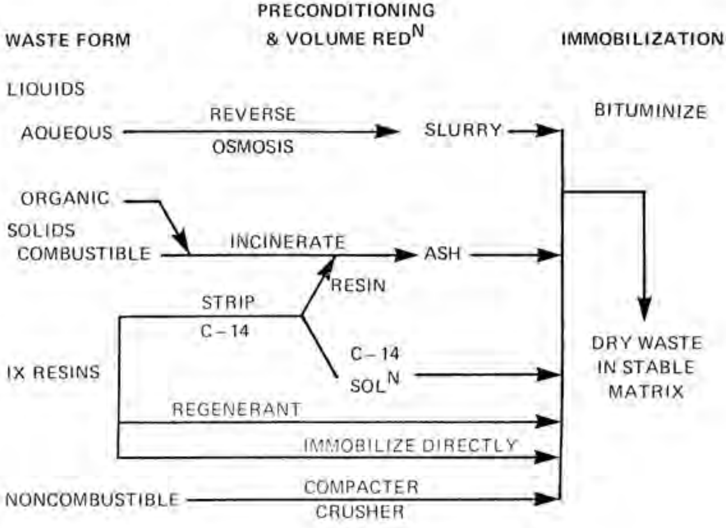
For aqueous-liquid wastes, which tend to be very dilute, AECL is developing reverse osmosis as a method for concentrating the waste to a slurry form. Reverse osmosis offers some potentially significant advantages over the more widely used evaporation process, particularly with respect to redundant capacity and energy requirements. However, at this early stage, no option is to be dropped until demonstrated shortcomings are confirmed.

Incineration is the existing preconditioning process for combustible wastes (both solid and organic liquid). The present system is a batch process which adds some flexibility to the operation.

A technology for preconditioning all non-combustible solid waste by a single process is not available. Compaction can reduce the volume of many solid wastes but cannot deal with more dense material.

This category of dense, non-combustible solid waste is composed of miscellaneous waste forms (valves, tubing, glass, insulation materials, tools, spent components and tools, etc). The storage of this category of waste in drums, assorted containers or as loose pieces in a trench, results in a large void factor. In an engineered storage system wasted volume costs big bucks!

FIGURE 2
WASTE CONVERSION



By using a high pressure hydraulic crusher, this category can be reduced to easily moveable compact lumps of material which can be efficiently stored and later retrieved if required.

For solid-metal masses such as valve bodies and other blocks, volume reduction can be achieved by packing more workable wastes into the voids around them. A typical process might be to place solid masses into the bottom of containers, fill with light-grade waste and compact. The lighter waste will be compacted and squeezed to fill voids in and around the solid lumps.

This will result in a high volume reduction factor (ranges from 5:1 to 10:1).

The hydraulic crusher is very similar to the compactor. The only difference is it exerts a far greater pressure so the ram can crumble more dense objects. Development work is proceeding on this solid preconditioning step.

The shape of the final waste form is important. Present use of non-rectangular drums is inefficient in space utilization when placed in rectangular structures. An efficient integrated system would put square pegs in square holes. Rectangular shipping and storage containers are being developed as part of this program.

Spent ion-exchange resins are presently stored in large stainless-steel tanks in the nuclear stations. A program of gradual removal from the stations in the bulk loose form in steel containers with storage in above ground "Quadricell" structures, is about to commence. Ion exchange resins contain the bulk of the activity (approximately 85%) in all reactor wastes. Studies are underway which are looking at several preconditioning steps:

1. simple stripping (including C^{14} and all f.p.'s and activated corrosion products);

2. acid digestion;

3. incineration.

The prime criterion is to produce a minimum volume and chemically stable form.

Immobilization Step

The immobilization techniques which are being assessed include concrete, polymers (both urea-formaldehyde and polyester), bitumen, glass, and ceramics. Including the factors outlines in "Characteristics of Waste Form", the assessment must take into account the versatility of each form in accommodating all expected types of waste and the probable reliability of the conditioning process. On this basis, bitumen appears to be the most attractive matrix for investigation. Concrete and polymers are considered somewhat deficient in leach resistance; glass and ceramic were thought to have limited versatility as well as a significant cost disadvantage for reactor wastes. Bitumen rates well in leach-resistance, versatility and relatively low volumes. Also, the demonstrated natural stability of bitumen over millions of years in an underground environment was considered a real advantage over man-made materials.

For immobilization with bitumen we are investigating two types of equipment - a twin-screw extruder and a thin-film evaporator. Since we do not want to pursue both options to full-scale demonstrations, one of our first aims is to assess their relative merits for our applications and choose one for further development. The quality of the product from both processing techniques, i.e., its stability and leach resistance, must be assessed.

The immobilization of all streams (liquid and solid) coming from the preconditioning step will be investigated. Interface problems are expected to be different for each waste stream. The main preconditioned feeds are expected to be:

(1) concentrated-aqueous wastes (RO and/or evaporators);

(2) incinerator ash.

The form in which ion-exchange resin will be processed remains to be determined. Preconditioning of the resin by incineration is attractive except for those resins containing significant amounts of carbon-14. One possibility is to strip the carbon-14 as carbonate from the resin, and combine this with the aqueous waste. The resin could then be incinerated. The alternative is to feed the resin directly to the bituminizing equipment. The choice will be affected by the relative leach resistance of the products from the two routes.

FINAL CONDITIONED PRODUCT CHARACTERISTICS

Retrievability, chemical/physical stability, leach resistance and volume reduction realized are the characteristics of reactor-waste form most important for long-term storage or final disposal. Until the final storage/disposal site characteristics are known, only qualitative judgements can be made as to the required conditioned wastes' characteristics. As the program continues, the required waste characteristics will become evident.

ACKNOWLEDGEMENTS

The CANDU radwaste processing philosophy as practiced today and being developed for tomorrow is due to the co-operative efforts of AECL personnel and Canadian Nuclear Utilities. A major part of the development work described here is being carried out at the Chalk River Nuclear Laboratories of AECL by a team under Dr. D.H. Charlesworth. His leadership in this work is gratefully acknowledged.

The contributions of other areas of the Design, Operations, Health Physics and Research within Ontario Hydro are essential to the development program described here.

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