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SUBJECT: The Alkaline Method for Treatment of
High Radiation Level Aluminum Wastes

TO: F. L. Culler

FROM: I. R. Higgins

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1.0 INTRODUCTION

The proposed utilization of commercial and military atomic power requires an answer to the problem of fission product disposal. It is essential that this radiation hazard be permanently isolated from the biological environment to the lowest possible level. The disposal method is a significant cost factor in an atomic power economy, therefore, the method must be simple. Consideration should be given to the recovery and use of certain valuable constituents, both new elements formed in fission, and special alloy components of the fuel element.

The proposed process most directly concerns aluminum-bearing wastes, but a simplified version would cover Purex wastes and stainless steel wastes. Hafnium-free zirconium and niobium may be sufficiently valuable to warrant head-end treatment for their separation from the long-lived radioisotopes. Further work will be carried out to study the details of the proposed method.

2.0 ASSUMPTIONS RELEVANT TO THE PROPOSED PROCESS

2.1 Permanent Disposal

It is assumed that storage tanks are satisfactory for permanent disposal if bulk salts are removed down to "carrier" quantities. It is suggested that fission product concentration and storage in a permanent type tank by the proposed method may be more economical than transport of the untreated waste to a permanent non-recoverable site. It is considered more desirable to have the fission products confined and under control than deposited anywhere out of control.

2.2 Temporary Disposal

It is assumed that relatively large volume cheap storage is

available for holdup for low-level, short-lived, and the less biologically dangerous isotopes.

3.0 PHILOSOPHY FOR DEVELOPMENT OF THE TREATMENT METHOD

The first step is a head-end type treatment adapted to ease of operation at high radiation levels and not subject to radiation damage. High separation efficiency is desirable but not expected or required. The function of this step is to reduce the radiation energy level by at least a factor of 10 or 100.

The second step should not suffer significantly from intermediate levels of radiation and should provide complete separation of the bulk salt or liquid from the long-lived and biologically most dangerous isotopes.

The final treatments are optional, as opposed to dumping in pits or pools. The low activity, short-lived dilute waste may be subject to simple steps, such as precipitation, fixation of bulk activity, recycle of liquid and chemicals, etc.

4.0 THE ALKALINE ALUMINUM FLOWSHEET

4.1 Headend Precipitation

The high radiation level waste from solvent extraction is usually a concentrated $\text{Al}(\text{NO}_3)_3$ solution containing practically all the initial radioisotopes. From high power level reactors some of the fission products are present in sufficient quantity to form precipitates without carriers. If iron is not already present it is added (Fig. 1), as Fe^{+3} to ~ 1 g/l, as the only added carrier. The $\text{Al}(\text{NO}_3)_3$ solution

Fig. 1 Alkaline Treatment of High Radiation Level $\text{Al}(\text{NO}_3)_3$

is added to a dilute NaOH solution to give a final solution of about 0.3 M NaNO_3 , 0.1 M NaAlO_2 , <0.1 M NaOH. The bulk of the fission products is isolated by centrifuging in a relatively small volume of ferric hydroxide precipitate carrying 99.9% of the R.E.'s (supposedly the same for Am and Pu), 99% of the Sr, 75-90% of the Zr, Nb, and Ru, and none of the Cs. The hydroxide precipitate must be dewatered and this step is more easily accomplished with hydroxide precipitates than by decomposition of salts. The fluffy powder may be stored as such, or be hydraulically pressed into compact billets. These billets are slipped into a steel cylinder, a steel plug added on the end, and welded in. A volume reduction of 400 may be expected from 1.0 M ANN. The cylinder is now mobile and can be disposed of in any way. Heat removal is required. Therefore, the suggested disposal is in an underground, asphalt-lined swimming pool.

4.2 Ion Exchange Removal of Long-Lived Isotopes

In alkaline solution cesium has an unusual affinity for phenolic cation exchange resin. The phenolic resins are also more stable to radiation than styrene resins. This unusual affinity for the monovalent cesium and the normally higher affinity for di- and trivalent ions makes ion exchange an efficient cleanup for Cs, RE, Sr, and supposedly Am and Pu. About 10,000 gal/day of NaAlO_2 solution would be formed from 1000 gal/day of 1.0 M $\text{Al}(\text{NO}_3)_3$. The fission products are stripped from the resin with 6 M HCl. For 600 gal/hr/ft² a 1-ft-dia ion exchange contactor is required. With a loading D.C. of 55 and a strip of 2, about 350 gal/day of 6 M HCl cesium solution results. The strip acid is recycled following evaporation and the Cs salt stored in cylinders for disposal or commercial use. An alternate strip solution may be alum, which may be recycled after the standard Cs removal step by crystallization. This ion exchange method is considered more efficient and less costly than the diban method previously considered (ORNL-1984).

4.3 Al(OH)₃ Precipitation

If cheap low-level waste storage space is available no further treatment of the NaAlO₂ solution is required. However, by using a few simple steps storage space may be conserved. Aluminum hydroxide can be precipitated as a granular, fast-settling, fast-filtering solid by bubbling CO₂ (or adding dry ice) into a sodium aluminate solution. The dry powder from 1 mole of Al(OH)₃ occupies a volume of about 300 ml, or is 3.3 molar. The Al(OH)₃ also carries the bulk of the remaining Zr and Nb activities.

4.4 Al(OH)₃ Effluent Treatment

After precipitation of Al(OH)₃ from NaAlO₂, the remaining solution is about 0.3 M NaNO₃ and 0.1 M Na₂CO₃ containing low levels of activity most of which is Ru. The solution may be stored in a holdup pond or evaporated to a smaller volume. To avoid the dispersion of any liquid, it is proposed that the salts be run through a membrane electrolytic cell to form NaOH and HNO₃. The 0.4 M NaOH needs no volume reduction and may be used over and over to form more NaAlO₂. The 0.3 M HNO₃ may not be as economical to evaporate and use over as it is to buy new HNO₃ for fuel element dissolution, but it eliminates the problem of releasing this low activity level waste to the environment. For 1000 gal of 1.0 M Al(NO₃)₃ per day, new caustic at 5 cents per pound would cost ~\$65/day. At 1 cent/kwhr, the power requirement at 80% efficiency for a membrane cell would be \$10. In addition, 2800 sq.ft. of membrane cell space would be required, with a \$6000 power supply. After evaporation the concentrated HNO₃ would be worth \$110/day (70%) at 5 cents/pound.

5.0 ADAPTATION OF THIS PROCESSING METHOD TO NON-ALUMINUM WASTE

For stainless steel and Purex wastes that contain little or no

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aluminum the same treatment method may be used with slight modifications. If NH_4OH is used instead of NaOH , the NH_4NO_3 and NH_4OH waste after the Cs ion exchange step may be treated with CaO and the NH_4OH boiled out for reuse. The resulting $\text{Ca}(\text{NO}_3)_2$ is probably not worth further treatment, and may be held up in ponds. For stainless steel fuel elements, the storage of bulk chemicals with fission products will be higher than an aluminum element with a NaAlO_2 treatment, but the dilute waste volume to be treated would be much smaller.

I. R. Higgins
I. R. Higgins

IRH/jr

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