



Synroc for Plutonium Disposal

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SUMMARY

A pyrochlore-rich titanate ceramic has been chosen by the US DOE for excess weapons Pu immobilisation in the USA. The development of this wasteform was based on the synroc strategy which aims to immobilise radioactive waste in durable multiphase titanate ceramics with phases chosen to be similar to titanate minerals that exist in nature and have immobilised U and Th for billions of years. The evolution of the pyrochlore-rich ceramic for Pu immobilisation from earlier synroc variants is described and the choice of process steps is discussed. Leaching studies demonstrate that the release rate of Pu from the wasteforms in aqueous media is very low and similar to those of U and the neutron absorbers Gd and Hf that will ensure avoidance of nuclear criticality in repository environments.

1. INTRODUCTION

In the aftermath of the Cold War, the US and Russia have agreed to large reductions in their stockpiles of nuclear weapons. The US is pursuing a two-track strategy for the disposition of 50 metric tonnes of surplus weapons plutonium:

- as mixed-oxide fuel in existing commercial power reactors to render the material non-weapons-usable
- immobilisation and geologic disposal.

Both approaches will ensure that the overall goal of the disposition program is met by "rendering the plutonium as unattractive and inaccessible for retrieval and weapons use as residual plutonium in the spent fuel from commercial reactors", ie the US National Academy of Science's (1) "spent fuel standard". Following an extensive screening study of more than 70 technologies and waste forms previously studied for high level waste (HLW) immobilisation, a synroc-based pyrochlore-rich titanate ceramic was recommended (2) to the US Department of Energy (DOE) for the immobilisation of surplus weapons plutonium for use with can-in-canister technology. In this approach, cans with 7.5cm diameter and about 51cm long would be loaded with ceramic discs containing the immobilised plutonium. These cans

in turn would be loaded into standard Defense Waste Processing Facility (DWPF) canisters for HLW glass and the molten glass would be poured around them to provide an external gamma radiation barrier.

The Australian Nuclear Science and Technology Organisation (ANSTO) has been developing titanate ceramic wasteforms since 1980 through its synroc program. ANSTO is participating with the Lawrence Livermore National Laboratory (LLNL), the lead laboratory on plutonium immobilisation for the US DOE office of Fissile Materials Disposition (MD). Other members of the team are Argonne National Laboratory, Pacific Northwest National Laboratory and the Westinghouse Savannah River Company Technology Center. The US DOE has expressed a preference for siting the immobilisation facility at the Savannah River site and current plans envisage that the facility would be fully operational by 2006/7.

ANSTO's involvement in the Pu immobilisation program commenced late in 1994. ANSTO provided LLNL access to some of the results of its synroc development program centred on the Synroc Demonstration Plant for the screening process and the subsequent Programmatic Environmental Impact Statement on Storage and Disposition of Weapons-Usable Fissile Materials (3). This screening process resulted in borosilicate glass and "titanate-based Synroc-like" ceramics being ranked first and second, respectively. This choice was reversed late in 1997 (2) following a period of intense wasteform development and formal independent evaluations.

The final choice by the DOE of a pyrochlore-rich titanate ceramic rather than a lanthanum borosilicate glass (LaBS) under development by WSRC for storage of americium and curium, was made against a number of criteria developed by DOE MD. These included: (1) resistance to Pu theft, diversion and recovery by a terrorist organisation or rogue nation; (2) resistance to recovery and reuse by a host nation; (3) technical viability, including technical maturity, development risk and acceptability for repository disposal; (4) environmental, safety and health factors; (5) cost effectiveness; and (6) timeliness.

2. SYNROC-BASED TITANATE WASTEFORMS

The synroc strategy aims to immobilise radioactive wastes in durable multiphase ceramics, with the phases chosen to be similar to titanate minerals that exist in nature and have immobilised U and Th for billions of years. Most of the early development of synroc wasteforms and process technologies focused on the Synroc-C formulation for immobilising liquid HLW from the reprocessing of commercial Light Water Reactor (LWR) spent fuel. The actinides in Synroc-C partition into zirconolite ($\text{CaZrTi}_2\text{O}_7$) and perovskite (CaTiO_3). Perovskite is mainly designed to immobilise Sr present in HLW. Barium hollandite ($\text{BaAl}_2\text{Ti}_6\text{O}_{16}$) is included in Synroc-C to primarily immobilise cesium although it also accepts K, Rb, and Ba. The basic Synroc-C formulation is flexible and a single precursor composition can be used to immobilise HLW loadings in the range 0 to 30 wt% without significant effects on its chemical durability in aqueous media. This flexibility is due largely to the extended solubility of the radionuclides in the titanate phases and the excess of titanium oxide as reduced rutile in the formulation. Even at 30 wt% HLW, the principal host phases, zirconolite, perovskite and hollandite, are undersaturated with respect to the key radionuclides in the HLW from LWR fuels.

The solubility of Pu in Synroc-C is extensive and 11.8 wt% PuO_2 (^{238}Pu) has been successfully incorporated (4) in solid solution in the host phases of Synroc-C, in experiments to study accelerated α -decay damage. The normal rare earth loadings were eliminated by molar substitutions with the Pu. Nevertheless, to increase the PuO_2 loadings in parallel with neutron absorbers it was necessary to look for alternative formulations to Synroc-C. Since zirconolite is the most durable phase of Synroc-C, the focus of these newer formulations was on the zirconolite family of phases, including pyrochlore which can incorporate as much as 50wt% of PuO_2 and/or UO_2 .

The avoidance of nuclear criticality is essential during wasteform processing, during its storage, and over the very long term in the repository environment. Such avoidance is ensured in the Pu-rich titanate wasteforms by having high loadings of neutron absorbers such as Gd, Sm and Hf in solid solution in the Pu host phases.

3. Pu IMMOBILIZATION IN TITANATE CERAMICS

A. Zirconolite-Rich Titanates

Zirconolite-rich Synroc has been developed for actinide wastes in general (5,6) and more particularly since 1994 as a candidate ceramic host for the immobilization of excess weapons Pu (7). Kesson et al (8) found that zirconolite solid solutions can separately accept as much as 27 wt% UO_2 , 20 wt% ThO_2 and 29 wt % rare earth oxides, particularly if charge coupled multiple cation substitutions are utilised.

Zirconolite-rich ceramics (7) containing about 10 wt% each of hollandite and rutile have been fabricated to contain 13.5 wt% PuO_2 as well as 10 wt% HfO_2 , 4 wt% Gd_2O_3 and 4 wt% Sm_2O_3 . The hollandite was included in the formulations as a host phase for radioactive cesium to provide an internal gamma radiation source in contrast to the present reference can-in-canister approach. The Gd and Sm oxides were each incorporated in a 1:2 molar ratio with respect to Pu. The HfO_2 content could have been increased to provide additional neutron absorption since it only replaced one third of the Zr in zirconolite in the above case. Full substitution of Hf for Zr is possible in zirconolites.

These ceramics were produced by two methods, either by (a) sintering of blended high-fired oxides in air for 4 hours at 1375°C or (b) by hot isostatic pressing (HIPing) at 1280°C/150 MPa of calcined alkoxide route powders in which the Pu was introduced as a nitrate solution prior to calcination at 750°C in argon. The HIPed material was fully dense ($\sim 5.1 \text{ g/cm}^3$) with all the Pu incorporated in the zirconolite. The sintered material had a grain size of $\sim 4 \mu\text{m}$ compared with $\sim 1 \mu\text{m}$ in the HIPed material and a density of $\sim 93\%$ of theoretical. Less than 2% of the Pu inventory remained atomically unincorporated in the sintered zirconolite - as a reacted oxide (Hf, REE, Pu) O_2 .

B. Pyrochlore-Rich Titanates

After the preliminary compositions of actual Pu streams for immobilization became available, it was apparent that the feed streams had on average about equal amounts of uranium and plutonium. This uranium content could be incorporated in a zirconolite-rich ceramic together with the Pu but with a significant loss of rare earth neutron absorbers because of total solubility limits. Consequently, it was decided to focus on a pyrochlore-rich wasteform in which:

- the uranium to plutonium ratio would be 2 to 1. High ^{238}U loadings would ensure additional criticality control through limitation of the long term $^{235}\text{U}/^{238}\text{U}$ ratio with the decay of ^{239}Pu to ^{235}U
- the plutonium-to-gadolinium-to-hafnium mole ratio was 1:1:1.

Pyrochlore and zirconolite in titanate ceramics are closely related structurally and are derived from ordered anion-deficient fluorite structures. In the cubic pyrochlore with the empirical formula unit of $\text{A}_2\text{Ti}_2\text{O}_7$ the A site is 8-coordinated and can be occupied by Ca^{2+} , Gd^{3+} , U^{4+} , Pu^{4+} and Hf^{4+} , amongst others. The Ti^{4+} is 6-coordinated. The monoclinic zirconolite ($\text{CaZrTi}_2\text{O}_7$) has five distinct cation sites permitting incorporation of a wider variety of cations of different charge and ionic radius than pyrochlore. Moreover, a particular cation may enter more than one site in zirconolite.

The baseline pyrochlore-rich ceramic was designed to incorporate about 11.5 wt% PuO_2 and 23 wt% UO_2 to yield about 95% of pyrochlore and 5% of rutile in which some substitution of Ti by Hf was anticipated. The actual product, formed after sintering for 4 hours at 1350°C , contains some brannerite (UTi_2O_6) which also contains Pu, Gd and Hf. As for the zirconolite-rich titanates mentioned above, a small (<2%) amount of the Pu inventory remains unincorporated in the sintered pyrochlore made from high-fired oxides, as a reacted oxide (Hf,REE,Pu,U) O_2 , whereas in materials made from alkoxide powders the actinides are fully dissolved in the titanate phases. The theoretical density of the baseline pyrochlore ceramic is about 6.0 gm/cm^3 and the observed density of the ceramics is about 5.6 gm/cm^3 .

The mineralogy and partitioning of Pu, U and the neutron absorbers among the phases in the pyrochlore-rich ceramic have been studied extensively. The phase relationships are depicted in the simplified ternary diagram in Fig 1. The baseline ceramic has six oxide components that need to be reduced to three variables on the basis of the following simplifying assumptions:

- TiO_2 is always in excess, so the TiO_2 activity is fixed at unity.
- UO_2 and PuO_2 behave sufficiently similarly in pyrochloric-rich titanates that they can be treated as one oxide, AnO_2 .
- Gd_2O_3 is distributed relatively evenly among the actinide-bearing phases, enabling it to be proportioned out of the phase diagram.

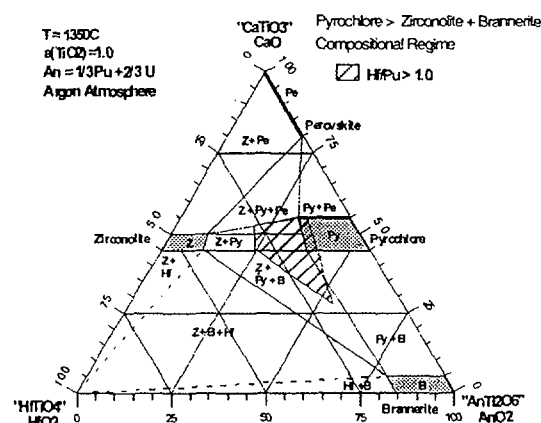


Figure 1. Depiction of the compositional regime (diagonally hatched) expected for impurity-bearing pyrochlore-rich ceramics.

The UO_2/PuO_2 ratio does show some variations amongst the phases. Pyrochlore and brannerite have UO_2/PuO_2 ratios close to 2:1 whereas it is about 1:3 in zirconolite. All primary Pu-containing phases have been found to accommodate more neutron absorbers (Gd and Hf) than plutonium on a molar basis.

The mineralogy of the pyrochlore-rich ceramic is influenced by impurities in the Pu feed stocks destined for immobilisation. The plutonium assay in the candidate feeds varies from under 10 wt% to over 99 wt%, ie from trace depleted uranium in plutonium to trace plutonium in fully enriched (93% ^{235}U) uranium. Impurities in the existing feed stock include: aluminum, carbon, calcium, chlorine, iron, fluorine, gallium, potassium, magnesium, molybdenum, sodium, silicon, tantalum, tungsten and zinc.

With the exception of carbon and the halides, fairly high levels of all the impurities in currently envisaged Pu feed stocks are tolerated by the immobilization form. Feed blending may be used to create impurity concentrations closer to the average which has been shown to be acceptable. Carbon contents and high halide element concentrations could be removed or depleted by appropriate pre-treatment of the particular Pu material prior to blending. The observed volume abundances of pyrochlore (60-90%), brannerite (0-22%), zirconolite (0-25%) and rutile (0-16%), depend on the impurity levels. Zirconolite is stabilized relative to pyrochlore by a number of divalent and trivalent metal oxides.

Silica, boron, phosphorus and the alkalis Na and K, if present at significant levels, lead to the formation of glasses. These glasses do not contain significant

Pu, U and neutron absorbers and are effectively micro-encapsulated by the resistant titanate phases.

4. TITANATE WASTEFORM FABRICATION

The Synroc-C process involves rotary calcination of a titanate precursor mixed with liquid HLW followed by uniaxial hot-pressing of the resultant calcine in bellows containers at about 1180°C/20 MPa (9). The hot-pressed disks containing about 30 kg of Synroc were about 30 cm in diameter. These disks were considered inappropriate for the preferred can-in-canister immobilisation option and could have impeded the flow of the HLW glass into the canister.

An alternative was to use HIPing rather than hot uniaxial pressing. ANSTO had demonstrated with zirconolite-rich formulations described earlier that long and slender cans could be so produced, containing up to 30 kg of ceramic. This would have eliminated the obstruction of the flow of the HLW glass and the risk of a negative impact on the quality of the glass.

The final choice of a process based on cold pressing and sintering was derived from the industrially mature technologies in glove-box plants used by the MOX fuel industry in Europe to satisfy the DOE MD criteria 3, 5 and 6 noted in the Introduction. There are substantial differences between the ceramic immobilisation product and the MOX product in terms of size and composition. The nominal size of the ceramic product (puck) is 6.3 cm in diameter by 2.5 cm high. The pyrochlore-rich ceramic sinters adequately at 1350°C whereas MOX fuel is produced by sintering at about 1700°C. Each ceramic puck contains about 58 g Pu and 20 sintered pucks are loaded in a can with 28 cans per HLW canister.

The most important steps in the fabrication process, in terms of impact on the final form, are the milling/blending and agglomeration steps. Two technologies relevant to the needs of the form processing have been established industrially. The "short binderless route" using attritor mills has been developed by British Nuclear Fuels Limited (10) whereas COGEMA, France uses ball milling (11). Both processes use dry milling to achieve sufficient micronisation of powders, homogenisation and conditioning to produce free flowing aggregates for the pressing operation.

Grinding and homogenisation are important steps because some residual actinide oxide particles (Hf, REE, Pu, U)O₂, tend to remain in ceramics fabricated from high-fired oxides whereas they are not part of the mineralogy of ceramics prepared from reactive alkoxide powders.

Sintering atmospheres ranging from reducing (Ar + 3.5% H₂), neutral (argon) and air have been examined during the development of the wasteform. Both air and argon are acceptable to produce the desired mineralogy. Under reducing conditions perovskite may form. Since perovskite is somewhat less durable in aqueous media than the other titanate phases it is less desirable.

Apart from benefiting from the proven industrial experience of MOX fuel fabrication, there are two other important factors in support of the cold press and sinter route:

- i) on line non-destructive evaluation of product density, mineralogy and composition is facilitated by the absence of cans used in hot-pressing;
- ii) fissile materials accountancy is more precise for bare pucks than canned material.

Nevertheless, HIPing remains a fall back option and has the benefits of a lower fabrication temperature, greater homogeneity and density, and would eliminate the need for automatic puck handling equipment.

5. WASTEFORM DURABILITY AND CRITICALITY SAFETY

Criticality safety during fabrication is ensured by limiting batch sizes of PuO₂ and by early blending with a precursor containing Gd₂O₃ and HfO₂, which are effective neutron absorbers. The safest approach to assuring criticality safety in the repository is to minimise the release of Pu, U and the neutron absorbers in the first place, i.e. to have low leach rates in aqueous media. Additionally, it is desirable for the individual release rates from the wasteform not to differ significantly and hence not generate uncertainty that the fissile materials and neutron absorbers separate after they are released. This separation may otherwise occur within a waste package, in the near-field repository environment, or in the far field.

Plutonium releases from Synroc-C (12), and zirconolite-rich and pyrochlore-rich ceramics (7) in deionised water are shown in Fig 2. These are normalised leach rates determined by α -counting and include contributions from ²⁴¹Am activity that could not be distinguished from Pu. The data in Fig 2 show the usual decrease of Pu leaching with time in MCC-1 tests and indicate that the Pu releases from the zirconolite-rich and pyrochlore-rich ceramics follow the long-term trend of Synroc-C. The "total" normalised release rate contains contributions from soluble Pu, colloids, and material absorbed on the leach vessel walls. The only uncertainty relates to dissolved elements that are reincorporated in very thin hydrous titania films

formed on the specimen during alteration. The total normalised release rates of Pu asymptote to a long term value of $5 \times 10^{-6} \text{ g.m}^{-2}.\text{d}^{-1}$ at 70 to 90°C. Such a low release rate corresponds to a long-term alteration rate of 0.001 mm/day or 0.5mm in 10^6 years. Consequently, the titanate wasteforms act as an independent immobilisation barrier.

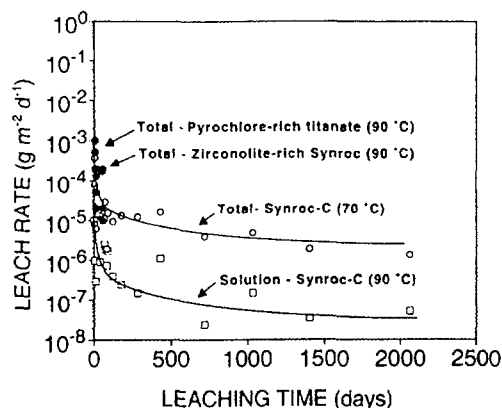


Figure 2. Normalised Pu leach rates measured in MCC-1 tests in deionised water at 70 and 90°C.

In 7 day MCC-1 leach tests at 90°C in deionised water, the Pu and U release rates varied by about a factor of 2 with impurity loadings from 1 wt% up to ~ 13 wt%. These results show that the baseline pyrochlore-rich ceramic is tolerant to the expected impurities and changes in mineralogy within the process regime adopted. In the same short term tests the total average leach rate ($\text{g.m}^{-2}.\text{d}^{-1}$) varied from ~ 2×10^{-4} for Pu, ~ 4×10^{-4} for U, ~ 5×10^{-4} for Gd and less than 4×10^{-5} for Hf. These leach rates demonstrate that the absolute differences between the releases of Pu and neutron absorbers are very small. These differences will be even smaller in the longer term.

A principal concern with crystalline wasteforms is that α -decay damage may render them amorphous and lead to significant changes in physical and chemical properties. The results of Weber et al (13), however, suggest that the Pu leach rate would increase only by a factor of about 10 from titanate ceramics in accelerated damage tests after amorphisation. The leaching behaviour of actinide doped specimens in accelerated damage tests could be influenced by α -radiolysis effects that would not be important in Pu immobilisation, given the longer half lives of ^{239}Pu and ^{235}U than that of ^{238}Pu .

Recent analysis by Gottlieb et al (14) concludes that no external criticality is possible from any Pu concentration mechanisms in the near field of Pu-containing can-in-canister packages employed in the Yucca Mountain potential repository. They also conclude that criticality is not likely in the far field.

6. DISCUSSION

The need for a high Pu loading in the wasteform was driven by economics since the Pu wasteform impacts the displacement of DWPF glass resulting in the MD program having to pay a fee for each additional HLW canister placed in the repository. Current costs for each additional HLW canister are about \$US500,000. The high Pu loading combined with high density of the pyrochlore ceramics (5.6 gm/cm^3) versus LaBS glass (3.85 gm/cm^3) resulted in a life cycle cost advantage to the ceramic of as much as \$US70M.

Significant public and worker health and safety benefits, as well as unquantified economic impacts, resulted from a total radiation dose field of seven to eight times higher for LaBS glass than the titanate ceramic, predominantly from (α, n) reactions on boron, a key constituent in the LaBS glass. This higher neutron dose would have necessitated additional shielding and may have prevented "hands on" maintenance of equipment in the glove boxes.

The primary resistance to diversion arises from the can-in-canister concept that provides a strong gamma radiation field. The canisters are large - 0.6m in diameter, 3m long and weigh about 2.5 metric tons. A shielded flask would be required to transport a canister containing ceramic cans and there would be no easy identification of co-located canisters with and without PuO_2 . The pyrochlore ceramic is not soluble in nitric acid and hence recovery of Pu is not possible using the well established PUREX process without additional steps at the head end.

The ability to adapt the mature ceramic process technologies from MOX fuel production Pu immobilisation was helpful in meeting criteria on technical viability and timeliness.

The durability of the synroc-based pyrochlore waste form which contained Pu, U and high loadings of neutron absorbers in solid solution of the titanate phases was seen to be an advantage for avoiding nuclear criticality possibilities in a repository. The data base available from ANSTO's synroc research for over fifteen years on Pu behaviour in the relevant titanate phases was a significant factor in judgements of eventual repository acceptance of the ceramic form. Finally, the extensive data base on the durability of natural mineral analogues of the titanate phases with high U and Th contents provided assurance of long term safety.

7. CONCLUSIONS

The pyrochlore-rich titanate ceramic chosen for excess Pu immobilisation in the US has evolved from earlier Synroc wasteforms to produce an optimised ceramic to meet the various demands of the fissile materials disposition program. The pyrochlore-rich ceramics can incorporate in a multi-phase microstructure high loadings of Pu, U and neutron absorbers such as Gd and Hf. The wasteform is tolerant of impurities in the Pu streams designated for immobilisation. Current leaching tests show that pyrochlore-rich ceramics have Pu releases in aqueous media that closely follow the behaviour of Synroc-C in which the total Pu release rate after 2000 days is as low as $5 \times 10^{-6} \text{ g.m}^{-2}.\text{d}^{-1}$ at 70 to 90°C, equivalent to alteration of about 0.5mm in 10^6 years. The low Pu release and the small absolute differences in Pu release rates compared to those of U, Hf and Gd indicate that criticality is not credible in repository environments.

ANSTO continues to support the Plutonium Immobilisation Project as part of a team led by LLNL. Our activities are centred on process, envelope development including impurity tolerance as part of the process to gain regulatory acceptance. The plutonium wasteform qualification process will assist in opening up for ANSTO further opportunities for development of titanate ceramics in other radioactive waste remediation projects.

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