

The Synroc strategy for HLW management

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Synroc is an advanced titanate ceramic comprising an assemblage of phases chosen for their compatibility, geological/geochemical stability and collective ability to immobilise the various elements and radionuclides present in high-level nuclear wastes. A major technical advantage of the Synroc strategy is that it possesses considerable flexibility in adapting to the requirements of special wastestreams and different preferred disposal options.

Synroc-C is the 'parental' formulation developed for the immobilisation of HLW from the reprocessing of spent fuel from commercial power reactors. It consists mainly of zirconolite $\text{CaZrTi}_2\text{O}_7$, hollandite $\text{Ba}(\text{Al},\text{Ti})_2\text{Ti}_6\text{O}_{16}$, perovskite CaTiO_3 , and minor titanium oxides. Together these phases accept into solid solution or microencapsulate most of the elements present in HLW. (eg Ringwood *et al.*, 1988; Blackford *et al.*, 1992).

Extensive evidence for the chemical durability of Synroc has been obtained from leach tests with simulated HLW, with active radioactive waste and from accelerated alpha-decay damage studies. This database was acquired not only in Australia but also as a result of collaborative international research programs (eg. Boulton *et al.*, 1990; Mitamura *et al.*, 1992). Moreover the process technology for Synroc fabrication has been successfully demonstrated on a commercial scale (10 kg/hr) with simulated non-radioactive HLW, at ANSTO's site in Sydney, Australia (Levins *et al.*, 1986).

Synroc-D, developed by Ringwood *et al.*, (1980) for the immobilisation of US Defense wastes at Savannah River, demonstrates the flexibility of the Synroc concept for handling different wastestreams. High concentrations of process contaminants such as sodium, silica and ferric oxides in Defense HLW could be accommodated by a Synroc phase assemblage in which nepheline and spinel coexist with zirconolite and perovskite, whereas hollandite is absent.

Alternative strategies aimed at minimising the volume of US Defense wasteforms requiring final disposal are currently under investigation at

Hanford (Straalsund *et al.*, 1992) Preliminary investigations at ANSTO suggest that the overall volume could be substantially reduced via partitioning of this wastestream coupled with the selective use of both Synroc and glass.

Recent political developments pose serious questions about the management of unwanted weapons-grade plutonium. Vitrification and irretrievable deep-borehole disposal has been canvassed by USNAS (1994) as a serious option. However vitrification of large quantities of Pu may encounter rather challenging technical problems. In contrast, a custom-designed formulation of Synroc would be ideal for this purpose. Synroc could accommodate high levels of Pu along with a neutron poison such as Gd to suppress criticality. Moreover ^{137}Cs could be deliberately incorporated in this variant of Synroc so as to inhibit future diversion of Pu. Finally, the proven chemical durability of Synroc at high temperatures permits serious consideration of ultra-deep borehole disposal to further reduce any risk of Pu retrieval.

There is renewed interest in advanced reprocessing strategies that involve the partitioning of actinides and their subsequent transmutation (or special disposal) so as to reduce long-term radiotoxicity and environmental hazards. Zirconolite-rich Synroc formulations have been developed for the ANSTO/JAERI co-operative program with this specific objective. Recently Lefevre *et al.* (1993) independently suggested that Synroc could be used for long-term storage of ^{244}Cm , ^{137}Cs and ^{90}Sr separated from French HLW. (These radionuclides are the principal sources of heat generation). This concept is unexpectedly attractive because the enhanced Cs and Sr loadings in a Synroc formulation designed for such a purpose should be high enough to anneal out much of the alpha-decay damage caused by ^{244}Cm .

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References

- Blackford, M.G., Smith, K.L. and Hart, K.P. (1992) *Materials Research Soc. Symp. Proc.*, **257**, pp 243–9.
- Boult, K.A. *et al.* (1988) UKAEA Harwell Report - AERE R 13318.
- Lefevre, J., Baudin, G., and Salvatores, M. (1993) In *Proceedings of Safewaste 93, Avignon, France*, June 14-18, Vol 1, pp 416–29.
- Levins, D.M. *et al.* (1986) In *Proc. of 2nd Int. Conf. on Radioactive Waste Management, Winnipeg, Canada, Sept. 7-11, 1986*, pp 576–83.
- USNAS: National Academy of Sciences, Committee on International Security and Arms Control, 1994, *Management and Disposition of Excess Weapons Plutonium*, National Academy Press, Washington, D.C.
- Ringwood, A.E., Kesson, S.E. and Ware, N.G. (1980) *Scientific Basis for Nuclear Waste Management*, Northrup, C., ed., Plenum Press, N.Y., Vol 2, pp 265–72.
- Ringwood, A.E. (1988) In *Radioactive Waste Forms for the Future*, (W. Lutze and R.C. Ewing, eds.), North-Holland, Amsterdam, pp 233–334.
- Straalsund, J.L. (1992) *Battelle PNL Report - PNL - 8388 Vol 1 and Vol 2*.