SYNROC

THE AUSTRALIAN METHOD FOR IMMOBILIZING HIGH LEVEL NUCLEAR WASTE

The main centres of SYNROC research and development in Australia are:

1. AUSTRALIAN ATOMIC ENERGY COMMISSION (AAEC) LUCAS HEIGHTS RESEARCH LABORATORIES, SYDNEY

- Site of the AAEC/ANU demonstration plant project.
- Laboratory fabrication studies on SYNROC.
- Leach testing of SYNROC.
- Radiation damage testing of SYNROC using fast neutrons.
- Fabrication and testing of small SYNROC specimens containing actinide and, separately, fission product elements.
- Study of SYNROC fine structure by electron microscopy and X-ray diffraction.

2. AUSTRALIAN NATIONAL UNIVERSITY (ANU), CANBERRA (RESEARCH SCHOOL OF EARTH SCIENCES)

- Continuing fundamental research on the scientific principles underlying SYNROC.
- Innovation and small-scale development of key process technology for the AAEC/ANU SYNROC demonstration plant.
- Development of new formulations of SYNROC for specific applications.
- Studies of radiation damage effects in natural mineral analogues of SYNROC phases.
- Laboratory fabrication studies and leach testing of SYNROC and SYNROC phases.

3. GRIFFITH UNIVERSITY, BRISBANE (SCHOOL OF SCIENCE)

- Studies on the fine structure of SYNROC before and after leach testing using a range of advanced microstructural surface analytical techniques.
- Crystallography of SYNROC phases.

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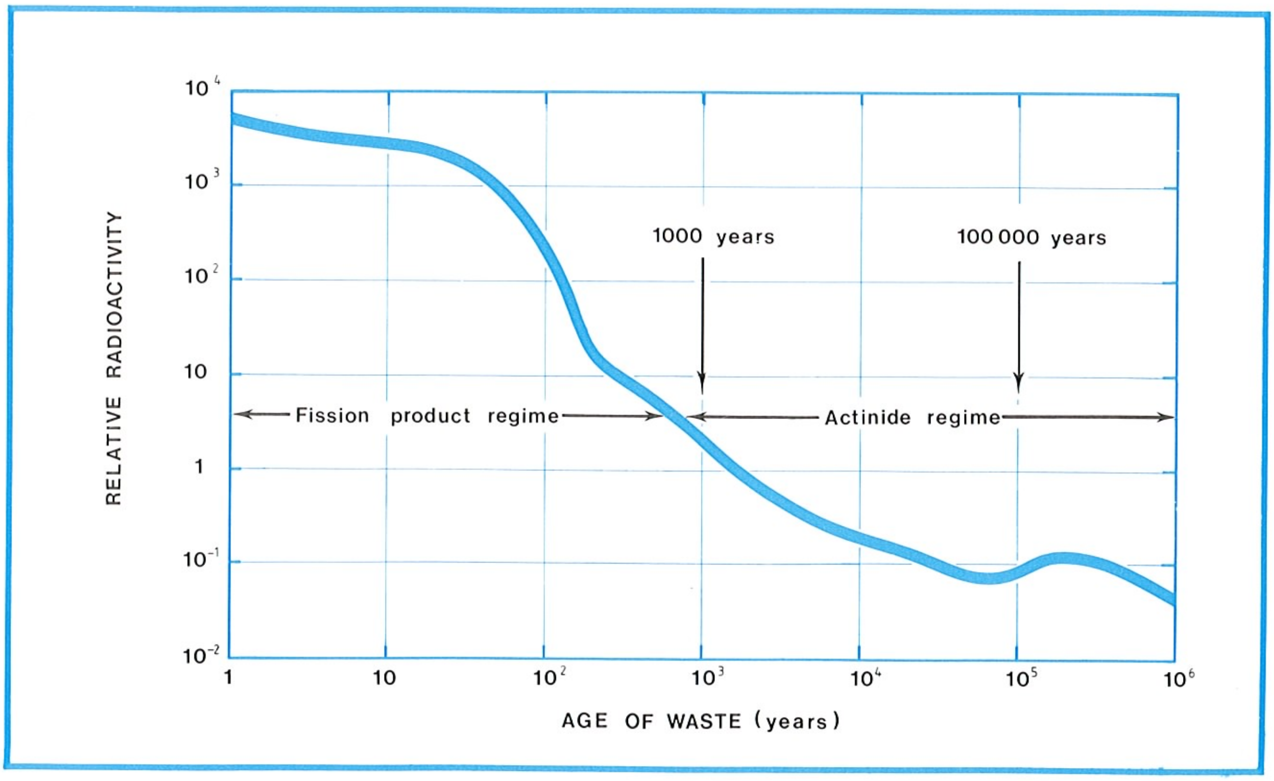
SYNROC

What is SYNROC?

SYNROC is a synthetic rock composed of three titanate minerals, hollandite $BaAl_2Ti_6O_{16}$, zirconolite $CaZrTi_2O_7$ and perovskite $CaTiO_3$, plus rutile TiO_2, and a small amount of metal alloy. Almost all of the elements in high level nuclear reactor wastes can be made to form an integral part of the crystal lattices of these very stable SYNROC phases. The natural radioactivity of common rocks and minerals is contained in just the same way. Moreover, close analogues of the titanate phases in SYNROC also occur in nature. These ancient natural minerals have survived extreme geological conditions for millions of years, yet they still contain their original content of radioactive elements. Such evidence, provided directly by nature, strongly suggests that SYNROC can immobilise high level nuclear wastes for the long periods needed for their radioactivity to decay to safe levels. SYNROC was first developed and characterised in 1978 by Professor Ted Ringwood and his colleagues at the Australian National University in Canberra. The properties and performance of SYNROC have been studied in scientific laboratories in many countries including Australia, UK, USA, Canada, Federal Republic of Germany and Japan.

What is high level waste?

A typical nuclear power station generating 1000 megawatts (one million kilowatts) of electrical energy produces 30 tonnes of spent fuel per year. After uranium and plutonium have been recovered for future recycling in reactors, about one tonne of high level waste is left behind. Almost all of the radioactivity originally in the spent fuel ends up in this high level waste. The diagram below shows how the radioactivity from fission product elements like caesium and strontium decays in about 1000 years. Thereafter the radioactivity comes from the decay of actinide elements such as neptunium, plutonium, americium and curium. Their



The radioactivity of the high level waste arising from the reprocessing of spent nuclear fuel decreases with time radioactivity takes about 100 000 to a million years to decay to acceptably low levels.

How is high level waste managed?

The general strategy is to convert the high level waste into a compact solid form and to bury this deep underground while the radioactivity decays. The geology of the burial site should meanwhile prevent any escaped radionuclides from reaching the biosphere. In addition the waste form itself should be very stable in the geological environment. The main requirement is that it should not dissolve (or 'leach') in groundwater to any significant extent.

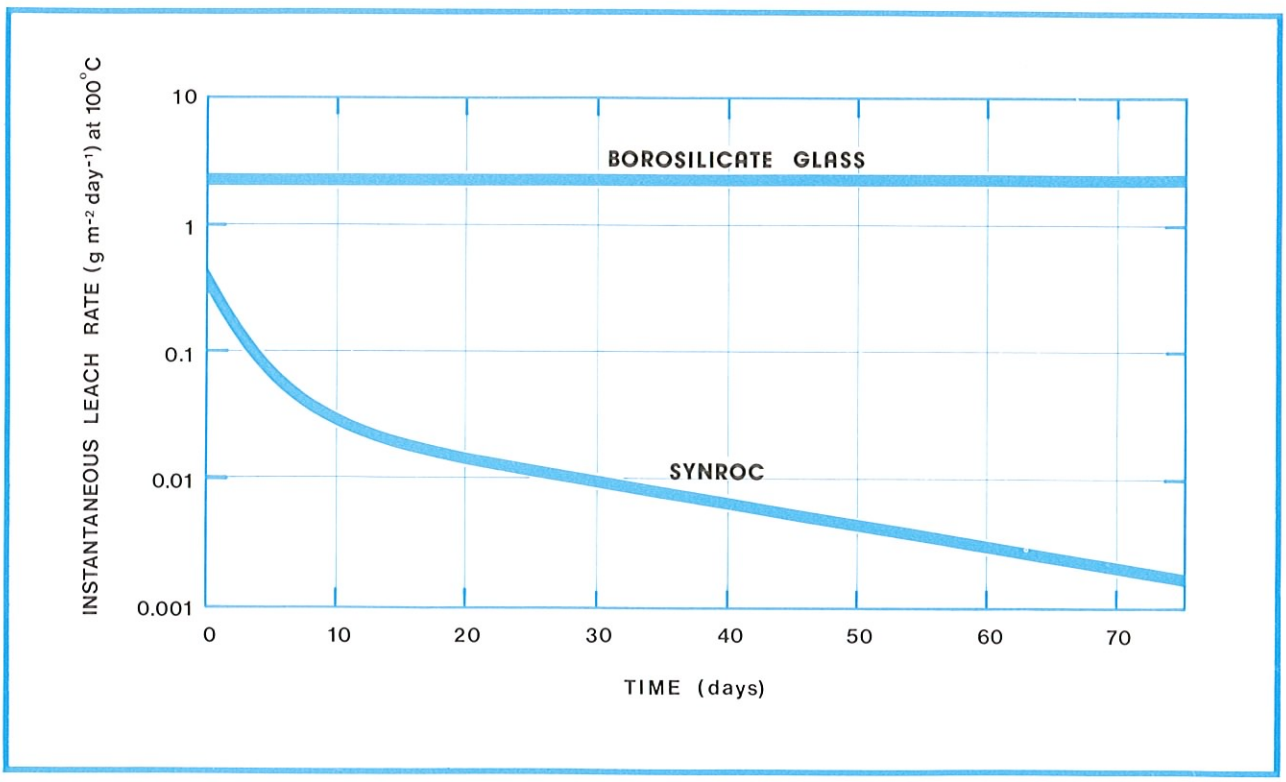
High level waste has not yet been disposed of geologically in any country, but in France it is being incorporated into a stable glass (known as borosilicate glass) on a commercial scale and then stored in air-cooled vaults for perhaps 30 to 50 years to allow it to 'cool' significantly (as regards both radioactivity and heat) before eventual disposal. Similar plans have been adopted in other nuclear power producing countries.

How good is SYNROC?

Leach tests in hot water at 100 °C have shown that SYNROC is at least 1000 times more resistant to leaching than a typical borosilicate glass wasteform, and that this advantage increases at higher temperatures (see next page). The test specimens show that glass flakes and cracks badly after a few days' exposure to high pressure water at 300 °C, whereas SYNROC is virtually unaffected.

What are the advantages of using SYNROC rather than borosilicate glass?

SYNROC is exceptionally resistant to ground-water leaching, even at high temperatures. This means that:



Waste form leach rates

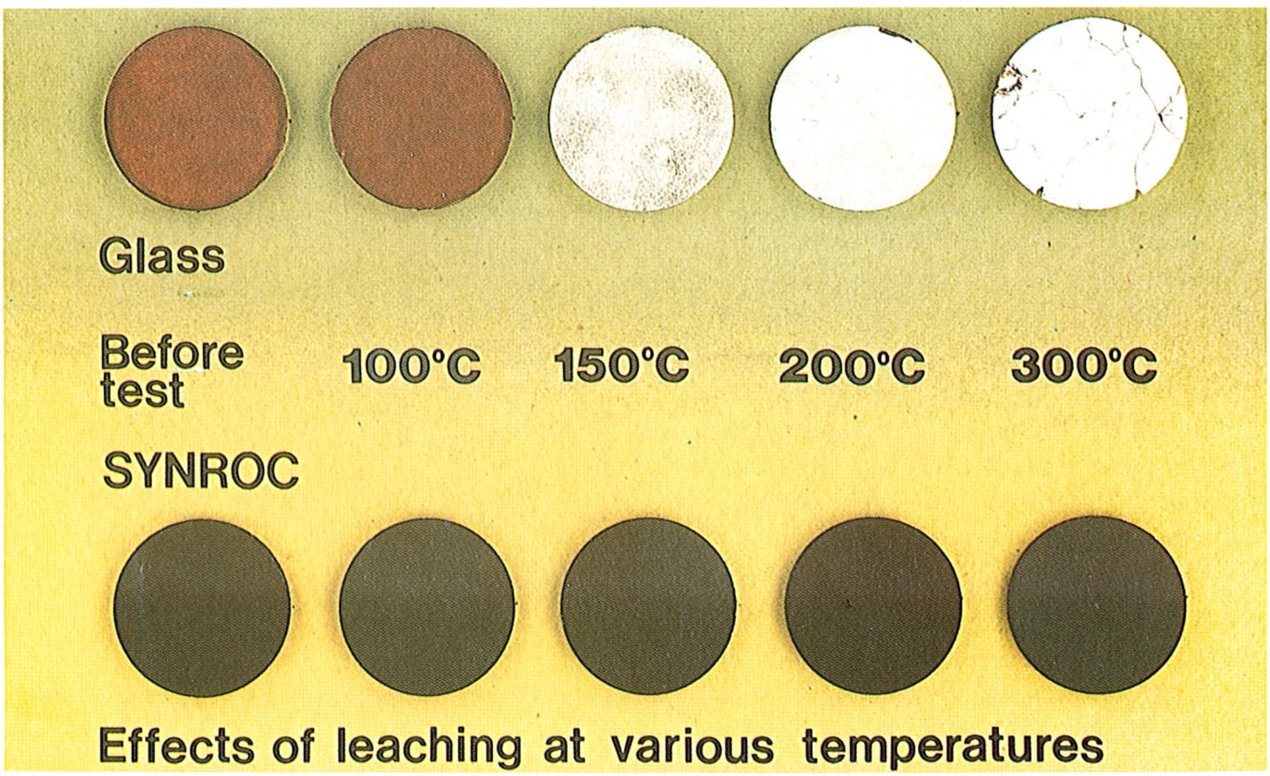
- SYNROC can accept higher loadings of heat-producing radioactive waste.
- SYNROC can survive underground for much longer and is therefore capable of immobilising the long-lived actinide elements.
- SYNROC can be buried in a greater variety of geological environments, e.g. in very deep (4 km) drill holes where underground temperatures are high.

How is SYNROC made?

SYNROC is made from five very simple chemicals - the oxides of titanium, zirconium, calcium, barium and aluminium - to which is added about 20 additional weight per cent of radioactive waste. (In the Australian program so far the waste is 'simulated' by means of non-radioactive chemicals of almost identical composition). The mixture is dried, calcined at 750 °C, then compressed at 1150-1200 °C. This produces a hard, dense black cylinder of solid SYNROC. The hot-pressing operation normally utilises a stainless steel container with a bellows-like configuration, which contracts and compresses around the SYNROC disc. This is the best type of process to scale up for eventual commercial operation and has been chosen for the AAEC/ANU SYNROC demonstration plant.

Will SYNROC be expensive to make?

All radioactive waste handling is expensive, but the fabrication of either radioactive borosilicate glass or radioactive SYNROC will amount to a very small proportion of the cost of electricity from a nuclear power plant. We believe that SYNROC will not be significantly more expensive, but operation of the demonstration plant will provide practical experience on which first estimates can be made of comparative costs of radioactive SYNROC and glass fabrication.



Comparative performance of SYROC and borosilicate glass

Does the radioactivity of the waste harm SYNROC?

This question is being answered by subjecting SYNROC to fast neutron irradiation in the Australian Atomic Energy Commission's research reactor HIFAR. Fast neutrons are much more effective than high level waste in producing damage to the SYNROC structure. In neutron irradiation tests lasting up to 6 months, SYNROC has withstood a simulated 100 000 years of high level waste containment without experiencing any significant physical damage or loss of resistance to leaching. Other verification comes from geological, crystallographic and isotopic studies of naturally-occurring SYNROC minerals, carried out at ANU. These minerals have been exposed to large cumulative doses of radiation over geological time. (The radiation is casued by the naturally-radioactive elements thorium and uranium which are part of the crystalline lattice structures of the minerals.) Despite radiation exposures much larger than SYNROC would experience over a million years, these minerals have quantitatively retained uranium, thorium and their decay products.

Has radioactive SYNROC ever been made?

SYNROC has not yet been fabricated with a full loading of radioactive high level waste. But scientists in Australia and other countries have prepared SYNROC (and SYNROC minerals) containing individual high level waste radionuclides, such as caesium, ruthenium, plutonium and other actinides. The ANU has prepared SYNROC containing radioactive uranium and technetium, and tested its properties. The AAEC has prepared and leach tested (with excellent results) radioactive SYNROC containing actinide elements and, separately, fission products which are a by-product from medical isotope production in the HIFAR reactor at Lucas Heights.

Collaboration with other countries

Australia is collaborating with the United Kingdom, Japan and Italy in its development and testing of SYNROC. In the UK small SYNROC specimens containing highly active UK reactor waste will be prepared in late 1986, and similar work is being planned in Japan for 1987-8. Collaborative arrangements with Italy cover the possible use of sol-gel technology in SYNROC fabrication.

SYNROC Demonstration Plant

Purpose

The purpose of the SYNROC Demonstration Plant Project is to show that SYNROC can be fabricated at or near full-scale on a non-radioactive basis in an engineered plant using process steps and an overall plant concept amenable to redesign for remote operation. Concurrently, operation of the plant will provide data for preliminary estimates of the cost of fabricating radioactive SYNROC. However, there is no commitment to build a radioactive SYNROC plant in Australia.

Plant Location

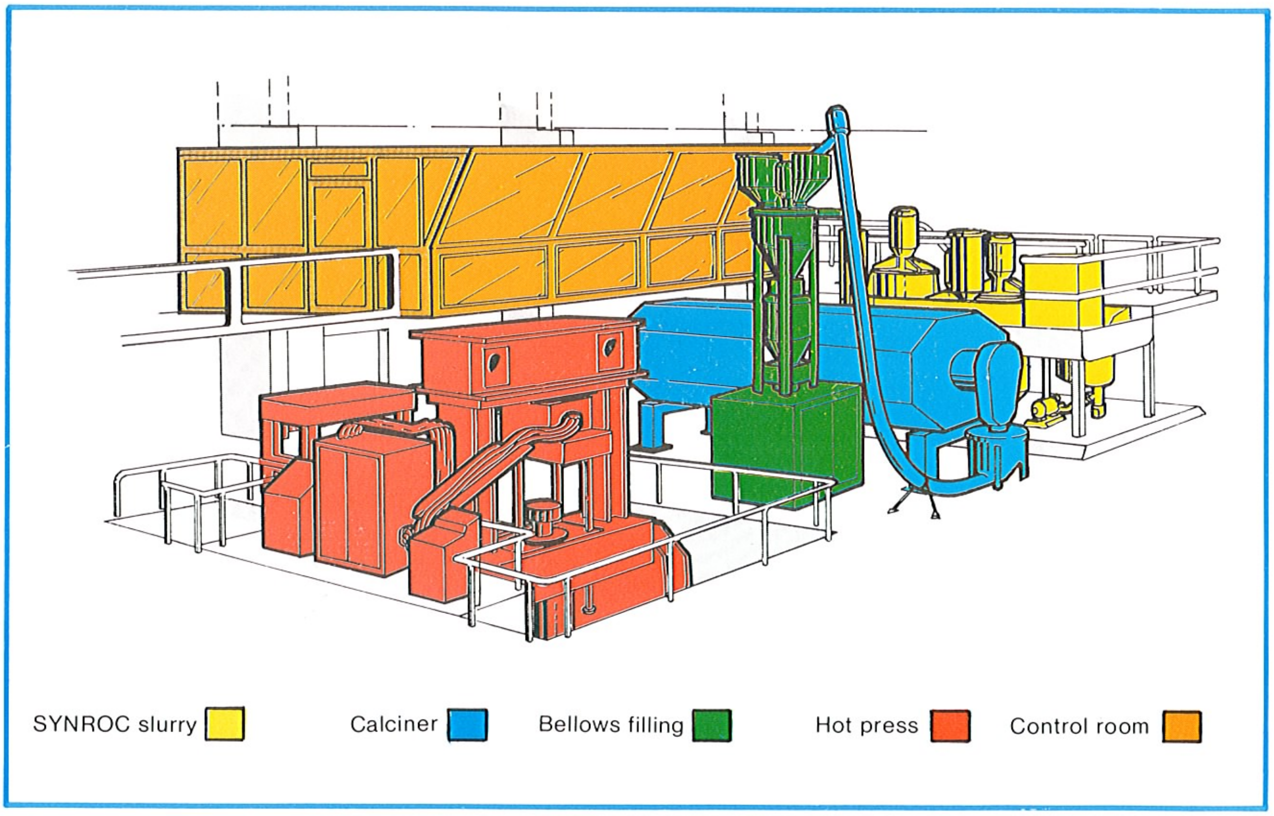
AAEC Research Establishment, Lucas Heights Research Laboratories, Sydney

Engineering Consultants

Davy McKee Pacific Pty Ltd, Chatswood, Sydney

Major Participants

The Australian Atomic Energy Commission (AAEC)
The Australian National University (ANU)



SYNROC demonstration plant schematic

Plant Scale and Time Scale

CANISTER SIZE

300-400 mm diameter

1-2 m long

CONTINUOUS PRODUCTION RATE

10 kg SYNROC per hour

(to be demonstrated in short campaigns)

PROJECT FUNDED FROM

July 1982

ENGINEERING CONSULTANTS APPOINTED

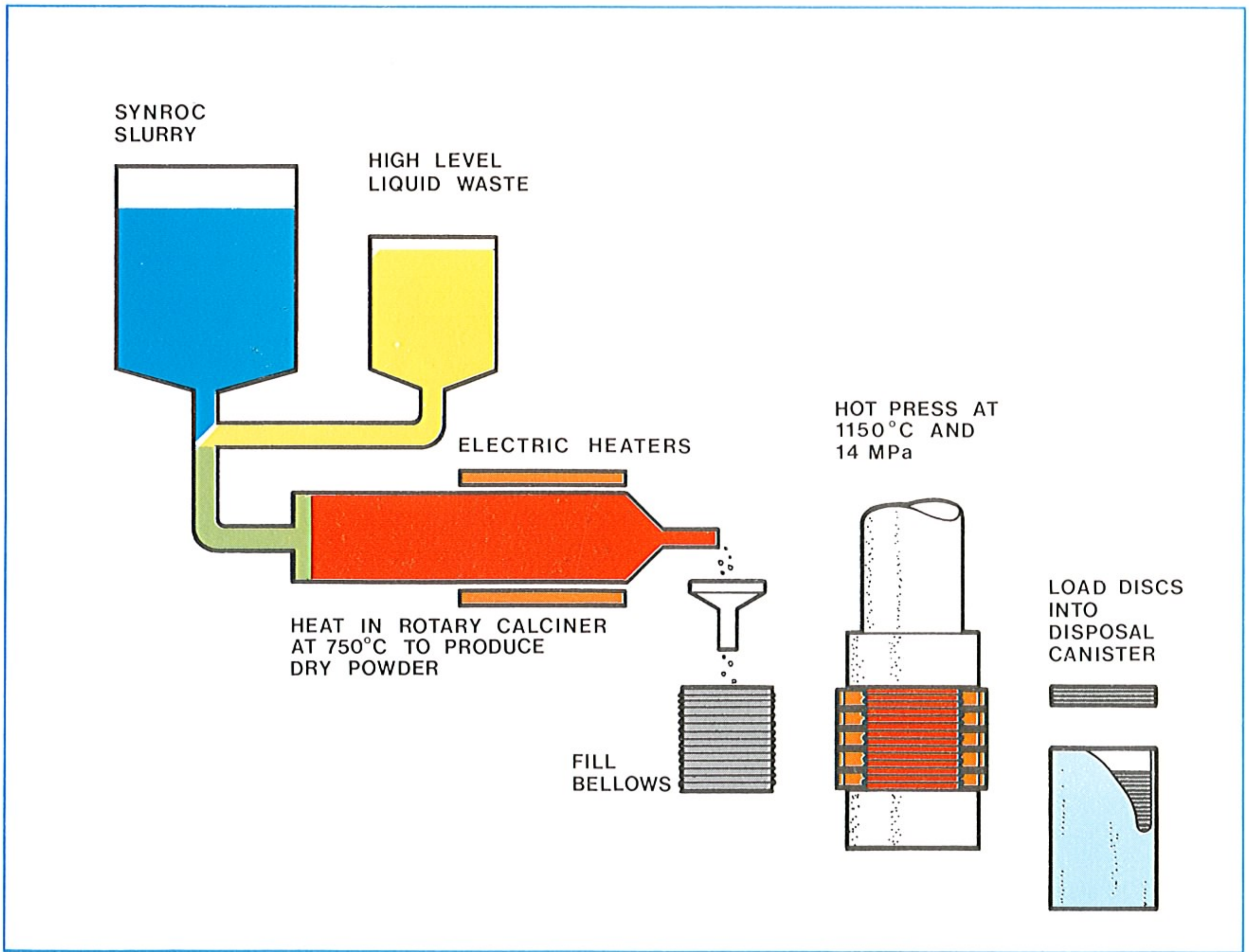
December 1982

DESIGN COMMENCED

January 1983

COMMISSIONING EXPECTED

1987



Schematic process flowsheet for the SYNROC demonstration plant

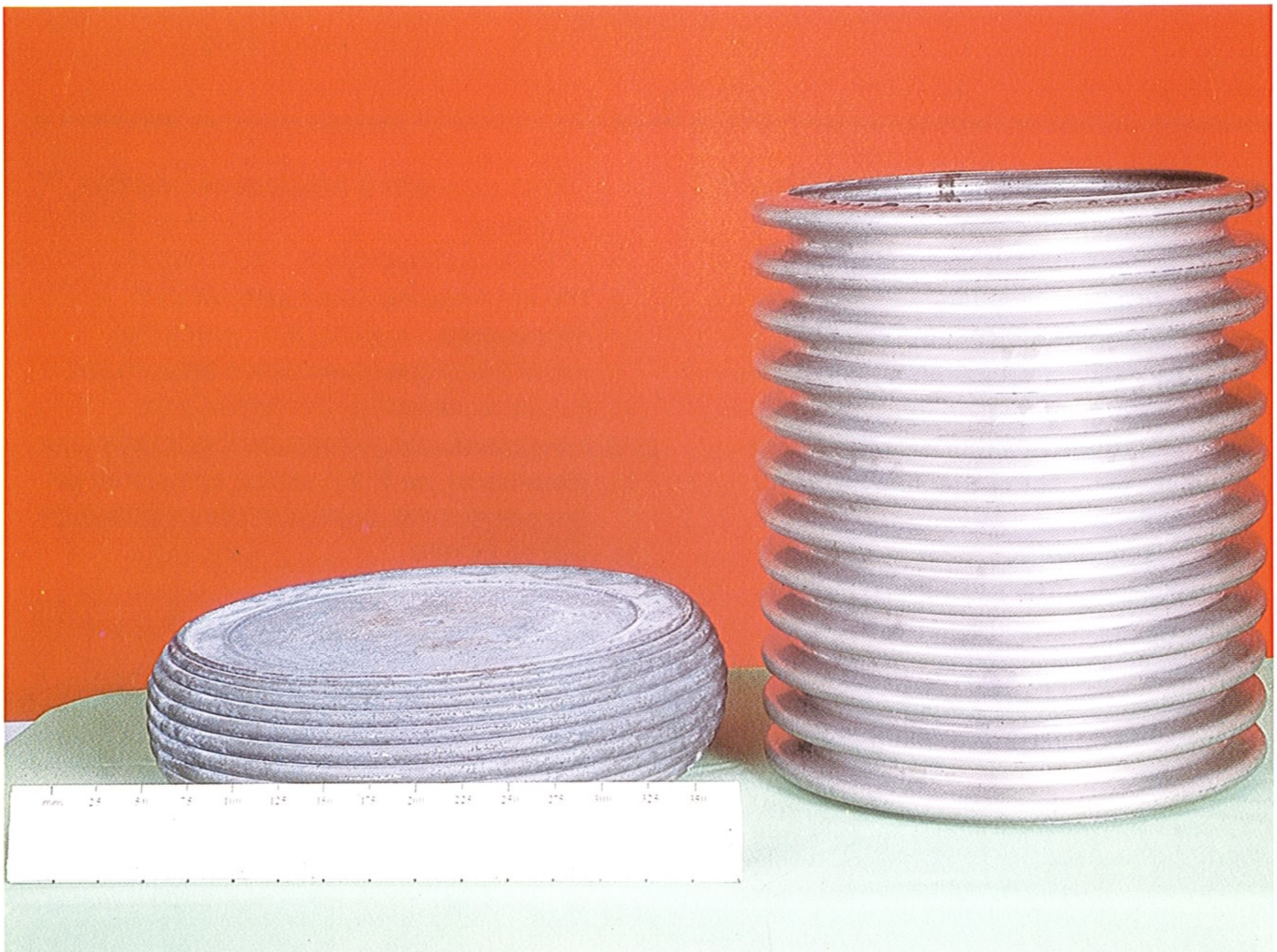
Status of SYNROC Demonstration Plant

1. The main process steps are:

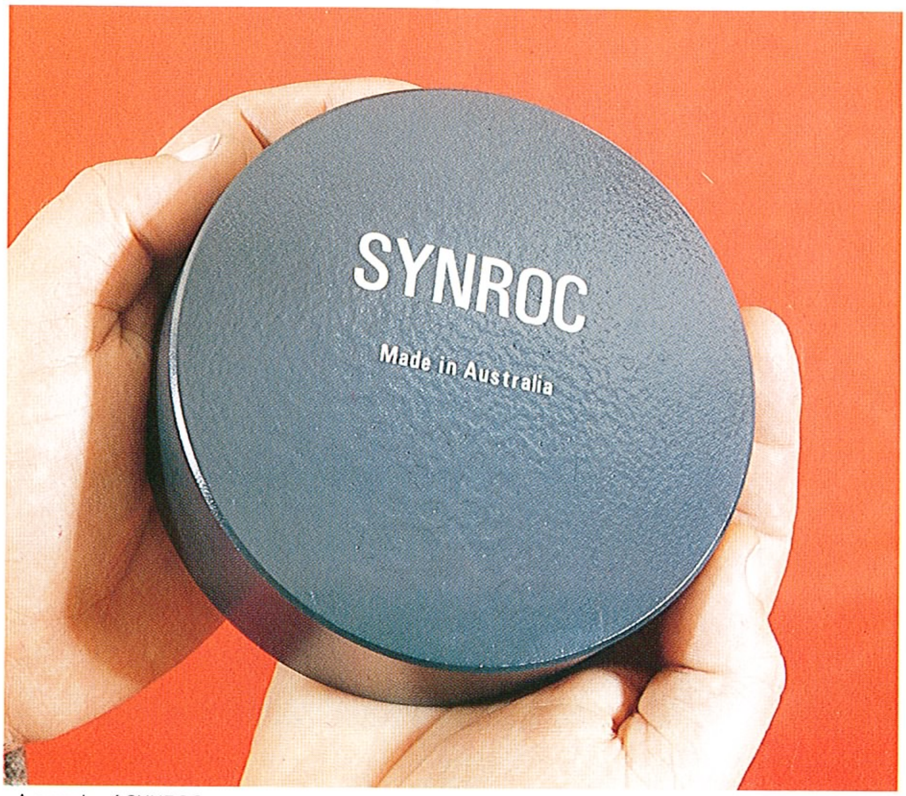
- Mixing of SYNROC slurry with simulated high level waste liquid.
- Injection of the resultant slurry into a rotary calciner operating at 750 °C under reducing gas atmosphere.
- Mixing of the resultant powder with 2 weight per cent of titanium metal for oxidation-reduction control.
- Filling of stainless steel or Inconel bellows with the SYNROC/Ti powder mixture.
- Uniaxially hot-pressing the filled bellows.
- Loading of the hot pressed bellows into a disposal canister and filling the interspace with a heat transfer medium, e.g. lead.
- Off-gas treatment.

2. The hot pressing step has been demonstrated successfully with a 250-tonne press combined with a 250-kilowatt induction heater.
3. Installation and commissioning of all major modules of the Plant will be completed by December 1986. Full Plant commissioning will be in 1987.

SYNROC filled bellows immediately after hot pressing at 1200 °C



The SYNROC powder fills the bellows on the right. A 'pancake' of densified SYNROC is fabricated when the bellows are compacted at high temperature



A sample of SYNROC

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