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The Canadian Fusion Fuels Technology Project represents part of Canada's overall effort in fusion development. The focus for CFFTP is tritium and tritium technology. The project is funded by the governments of Canada and Ontario, and by Ontario Hydro.

The Project is managed by Ontario Hydro.

CFFTP will sponsor research, development and studies to extend existing experience and capability gained in handling tritium as part of the CANDU fission program. It is planned that this work will be in full collaboration and serve the needs of international fusion programs.

ATOMIC ENERGY OF CANADA LIMITED

THE FUSION BLANKET PROGRAM AT CHALK RIVER

I.J. Hastings

ABSTRACT

Work on the Fusion Blanket Program commenced at Chalk River in 1984 June. Co-funded by Canadian Fusion Fuels Technology Project and Atomic Energy of Canada Limited, the Program utilizes Chalk River expertise in instrumented irradiation testing, ceramics, tritium technology, materials testing and compound chemistry. This paper gives highlights of studies to date on lithium-based ceramics, leading contenders for the fusion blanket.

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FUSION BLANKET PROGRAM AT CHALK RIVER

by I.J. Hastings, Chalk River

Atomic Energy of Canada Limited (AECL) is in the process of assuming a lead role in Canada's fusion effort, as a result of recent federal government decisions calling for the staged transfer of fusion responsibility from the National Research Council to AECL. Transfer of the program, which is funded under the Energy, Mines and Resources' Panel on Energy R&D, should be completed by 1987 April 01. Major components involved are the Tokamak de Varennes and the Canadian Fusion Fuels Technology Project (CFFTP). AECL has had a fusion program at Chalk River Nuclear Laboratories since 1983, co-funded by CFFTP and concentrating on the fusion blanket, tritium recovery and storage, monitoring and biological effects. The Fusion Blanket Program utilizes CRNL experience in instrumented irradiation testing, ceramics, tritium technology, materials testing, and compound chemistry.

Dr. Ian Hastings, Manager of the Fusion Blanket Program at CRNL, sends us this report for "Perspective".

Lithium ceramics are favoured by many to make up the blanket that will surround the plasma in an operating fusion reactor. The blanket material will be irradiated externally by fusion neutrons from the D-T reaction and internally by fast ^{3}He and ^{3}H (tritium) ions, formed in $^{6}Li + n$ and $^{7}Li + n$ capture reactions. Heat generated in the blanket will be removed for conventional electricity production; tritium will be swept from the blanket by a carrier gas, reprocessed, and re-introduced into the plasma as fuel. It is impractical to test all possible candidate materials for fusion reactors by irradiating with fusion-energy neutrons because of the cost in building high flux, 14 MeV neutron facilities. An alternate approach is to examine fundamental and engineering properties of candidate materials by irradiating with neutrons in fission reactors, or with particle beams in accelerators. These techniques allow scoping tests on blanket materials, and form the basis of the Fusion Blanket Program at Chalk River.

The major irradiation testing facility at CRNL is the NRU research reactor. NRU has the capability for two types of irradiation tests to evaluate solid breeder ceramics. The unvented capsule tests, designated CREATE (Chalk River Experiment to Assess Tritium Emission), are those that do not have tritium release instrumentation (thermocouples and flux detectors can be included). Tritium release information is obtained after the irradiation is complete, and the capsule is removed from the reactor. These tests can measure the interaction between the ceramic and the cladding material; also pellet swelling, cracking, and grain size and pore size changes can be observed, as well as the amount of tritium remaining in the ceramic. Information is obtained on the form of tritium released, as a function of capsule material and sweep gas. Maximum sample size is 2 cm diameter and 15 cm long, but typically, samples weighing 50-100 mg are cut from sintered pellets of the ceramic for irradiation. Each sample is vacuum-annealed in a quartz tube, and sealed in the tube for irradiation without further exposure to air, then irradiated for 48 h at an estimated temperature of less than 370 K. The maximum flux available is 4×10^{18} $n \cdot m^2 \cdot s^{-1}$ (thermal) and 7×10^{17} $n^2 \cdot m^2 \cdot s^{-1}$ (> 1 MeV). The free tritium recovered at room temperature is measured, as well as the isothermal tritium release at the post-irradiation test temperature; both tritiated water and reduced tritium are determined. The tritiated water may include T_2O and HTO, and the reduced tritium T_2 and HT. Both He and He-1% H_2 are used as sweep gases at a flow of 0.5 L/min, with oxygen and moisture contents of the purified gas less than 1 μL/L and extraction vessels constructed from quartz, stainless

steel, Inconel-600 and nickel are available. All post-irradiation tests are performed at 873 K for 4 h; tritium remaining in the ceramic after annealing is determined by liquid scintillation counting. Four tests on $LiAlO_2$ and Li_2O in the CREATE series have confirmed that, under reducing conditions ($He-H_2$ sweep), most of the tritium (> 70%) is released as HT or T_2; the balance as HTO or T_2O while residual tritium is very small, less than 0.02%. Varying the sweep gas composition has a dramatic effect on the form of tritium released. With a quartz extraction tube during post-irradiation heating, a He sweep gas results in 10-30% release as HT or T_2. CRNL-fabricated $LiAlO_2$ behaved similarly to that from ANL in these tests. Further CREATE testing has been performed using $LiAlO_2$ from France and Li_2O from Japan, under the BEATRIX international breeder exchange matrix; data analysis is underway.

The vented capsule tests, designated CRITIC (Chalk River In-Reactor Tritium Instrumented Capsule) permit continuous in situ monitoring of the tritium release from the ceramic during the irradiation, by passing a sweep gas around or through the ceramic and into an analysis train. The form of tritium is also determined. Since fusion reactors will use a sweep gas to recover the tritium in the same way, the experiment attempts to model a miniature segment of a blanket. Figure 1 shows a diagram of the CRITIC assembly. A sample size 4 cm diameter by about 10 cm long is possible in the current capsule, typically in the form of sintered pellets, with about 20% porosity. The capsule provides approximately uniform ceramic temperatures to facilitate analysis of release data; a small radial temperature gradient of about 50 K enables calculation of thermal conductivity of the

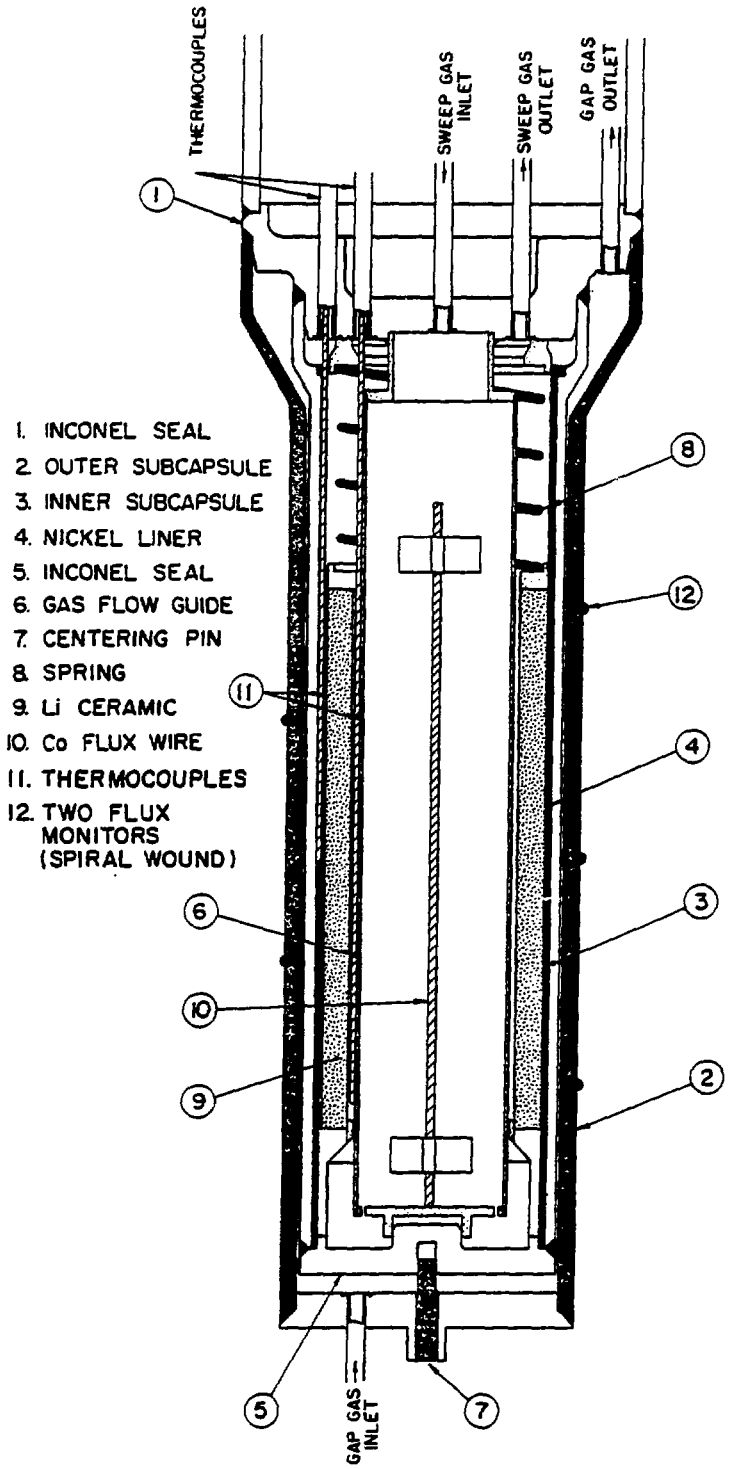

Figure 1 CRITIC - I vented capsule assembly.

ceramic. The temperature is adjustable between 400 K and 1200 K, which covers the range of expected operation in a commercial reactor, by varying the composition of an insulating gas layer (gap gas). In addition to on-line tritium analysis, gamma spectroscopy monitors the release rate of trace quantities of other radioactive species. These are expected both from neutron activation and from fission of uranium impurities in the ceramic. A moveable spectrometer will be located at the glove box containing the tritium analysis system, and a portable spectrometer will be available adjacent to the gas line exit ports from the reactor. Other instrumentation will include thermocouples, on-line flux monitors, and integrated flux monitors. Analysis of the gap gas also permits measurement of the permeation rate of tritium through the Inconel capsule wall.

The first vented test at Chalk River, CRITIC-I, will examine ANL-fabricated Li_2O, 0.3 wt% ^6Li, 30 mm ID, 40 mm OD annular pellets, in a six-month irradiation at 700-1200 K, varying the sweep gas, with on-line HT/HTO measurement. Burnup will be 0.3%. Li_2O conductivity and tritium permeation will also be measured. Start-up is scheduled for mid- 1986 for this BEATRIX test. CRITIC-II is scheduled to test CRNL-fabricated $LiAlO_2$ spherepac in 1987; French $LiAlO_2$ will be irradiated under BEATRIX in CRITIC-III (1988).

Most neutronics effort has been in support of the CRITIC experiments in NRU, and blanket modelling has also been completed. In NRU tests, tritium production will occur almost exclusively from thermal neutron captures by ^6Li. In a fusion reactor, tritium will be generated from captures, by both ^6Li and ^7Li, of neutrons within a wide energy range, 0-14 MeV. In both

cases, helium will also be produced, and the stoichiometry of the ceramic will change. The average tritium production rate per gram of ceramic in a blanket will not be far different from the rate in NRU. However, in a breeder material in a fusion reactor, displacement damage from energetic neutrons will be larger than in NRU. Near the first wall of a fusion reactor, the displacement damage rate will be larger by up to two orders of magnitude, near the blanket rear, it will be comparable. A transport calculation (P_3S_8) using the 1-D XSDRNPM code was performed on a helium-cooled Li_2O blanket; natural ^6Li enrichment (7.5 at%) was assumed for the breeder blanket. The blanket tritium breeding ratio and energy multiplication were calculated to be 1.19 and 1.3, respectively. These values are in close agreement with reported values for this particular blanket.

Chalk River has wide prior experience in nuclear ceramics. For example, the concept of high density UO_2 fuel for CANDU reactors was developed at CRNL, before being transferred to Canadian industry. Of the potential solid lithium breeder candidates, three have been selected for further consideration at CRNL: the oxide, the beryllate, and the aluminate. Lithium oxide has the highest lithium density and so is the most desirable from a breeding standpoint. The main problem with the oxide is its high affinity for water vapour; any processing or handling must be performed in a glovebox with good atmospheric control. The beryllate is attractive because of its potential for neutron multiplication, but a potential technological drawback is its low melting point. Perhaps the biggest challenge of the beryllate from a fabrication development standpoint is its toxicity, which makes glovebox operations a necessity.

Lithium aluminate is probably the most preferred compound next to the oxide at the present time. It does not have as high a lithium density but is easy to work with in a normal laboratory environment. Effort was focused on this compound because it was important to acquire as much experience in as short a time as possible on the behaviour of lithium ceramics, and to make rapid progress in the development of certain novel fabrication concepts.

Most solid breeder materials have been made in the form of pellets pressed from powders. In the case of the aluminate, the powder is commonly made by decomposing the carbonate in the presence of alumina. Recently, advantages have been cited for fuel in the form of microspheres (spherepac), and several approaches by other investigators have been pursued in that direction. At CRNL we are investigating powder approaches for fabricating pellets, and both powder and sol-gel approaches for making microspheres. There is significant prior experience in this area from our work on fission fuels. The pellets from commercially-available powder have been delivered to CEN-Saclay as part of the CRNL contribution to the BEATRIX program. The pellets have also been irradiated at CRNL.

Rotary agglomeration is a promising novel method of preparing microspheres from powders. With this method a suitable powder is agglomerated into microspheres by tumbling; microspheres about 2 mm in diameter have been produced. Sintered densities are 65-70% of theoretical. This approach to making microspheres is unique within the breeder blanket fabrication community; preliminary results have been encouraging.

A major problem with using commercial powders sold only for their chemical purity is that physical properties such as particle size, particle morphology, and surface area are not controlled or even consistent. These properties determine the properties of the product. Thus it is important to be able to synthesize the aluminate such that the powder has the physical properties desired, and work has been progressing in this direction.

The CRNL approach to the sol-gel formation of microspheres is to use the lithium in a form that is soluble in an aqueous medium but not in an organic one such as alcohol. The aluminum component is hydrolyzed to a sol and then gelled into the microsphere form in alcohol. This is accompanied by the simultaneous precipitation of the lithium component contained with the gelled alumina structure. At present microspheres of pure alumina have been prepared. This work has provided valuable information on sol formation and microsphere gelation characteristics. Work has started to introduce the lithium component, with the oxalate and formate having been selected as candidates for the starting lithium compound.

In lithium compound chemistry, to evaluate the practicality of various chemical exchange systems for lithium isotope separation, one must be able to measure two key experimental parameters, K_{dist} and α. K_{dist} is the distribution coefficient of a given lithium salt between two immiscible liquid phases, and is calculated from the equilibrium concentrations of the salt in the two phases. The isotopic separation factor, α, gives the extent of isotope fractionation between the same phases for a single stage; it is determined from the equilibrium $^6Li/^7Li$ ratio for the salt in each phase.

Lithium concentrations are usually obtained for solutions by atomic emission or atomic absorption spectroscopy, and lithium isotope ratios by mass spectrometry. Alternatives to these methods have been examined, in an attempt to find more efficient means of analysis. A simple procedure based on a commercially available cation-specific electrode and pH/mV meter has been developed for measurement of lithium ion concentration in aqueous solution. A good calibration curve is achieved over the $[Li^+]$ range 1×10^{-5} to 1 mol/L; sample analysis time is less than five minutes, and accuracy/precision are comparable to or better than ICP-AES over this concentration range (± 1-2%). Because lithium-specific electrodes are not yet available, other cations like Na^+ or K^+ are interferences. Therefore this procedure is only applicable to lithium solutions containing negligible concentrations of other cations. Only a few lithium salts are suitable for analysis of the $^6Li/^7Li$ ratio by mass spectrometry. At CRNL all lithium samples must be converted to LiF, and interfering organic contaminants must be removed by chemical oxidation. In the search for a more convenient method we have been looking at nuclear magnetic resonance (NMR) spectroscopy. Both ^6Li and ^7Li produce a signal, so that coaxial solutions containing an isotopic standard and the unknown sample, respectively, can provide a ^6Li and a ^7Li spectrum. Current tests are being done to determine the limiting precision and accuracy obtainable by this technique.

In fundamental material studies, the three main accelerator facilities at CRNL that have been used for implanting heavy ions are a 70 kV mass separator, a 2.5 MV mass separator and a 2.5 MV Van de Graaff accelerator. Using various heating and cooling stages, the sample temperature can be

varied from 35-770 K. Facilities are also available for sweeping the ion beams over the target, thus resulting in uniform implantation over areas up to ~ 1 cm^2.

In one test series, HT^+ and DT^+ ions have been implanted into polycrystalline sintered samples of Li_2O and $LiAlO_2$ and into single crystal Li_2O. Ion energies varied from 10 keV to 50 keV and ion doses were in the range 10^{19} - 5 x 10^{20} ions/m^2. Elastic recoil detection, ERD (using 2 MeV α-particles), and thermonuclear reaction analysis, TRA ($T(d,\alpha)n$ reaction at E_d = 0.25 MeV) were used to obtain tritium depth profiles. The ERD technique was suitable for profiling tritium at small depths (0.1-0.2 μm) below the surface, while the TRA technique was best for deeper profiling (0.5-1.0 μm); in both cases the sensitivity was ~ 10^{19} T atoms/m^2. The diffusion of implanted tritium was followed as a function of temperature up to 800 K and the data obtained are compared with other tritium retention studies. In addition, the ERD technique was applied to study depth profiles of H and D upon exposure to H_2O and D_2O, after prolonged ion bombardment, and as a function of annealing time and temperature.

A major effort has been associated with the Tandem Accelerator Superconducting Cyclotron (TASCC). A cryogenic target chamber for light ion irradiation (proton, deuteron, He) has been constructed and incorporated into the beam line of the TASCC and an energy range of 15-22 MeV has been chosen because it best simulates the energy spectrum of the primary knock-on atoms produced by 14 MeV neutrons. The most common problem associated with

ion irradiation, especially at cryogenic temperatures, is beam heating. To minimize this, the specimens are immersed in a low pressure helium cooling gas (~ 0.1 Pa). The ion beam is swept over a length of 2.5 cm using a sweeping magnet upstream. Radiation damage is presently being determined by resistivity changes, but in future it will be monitored by measuring simultaneously resistivity and the length of the specimen, using a scanning laser; a computerized data acquisition system is used during all stages of the experiments. A series of experiments has been performed without an ion beam to confirm the thermal characteristics of the cryostat and one preliminary test has been conducted with a 15 MeV proton beam. The helium cooling gas performed well, resulting in a temperature rise of less than 1 K at a beam current density of 10^6 nA/m^2 and a resistivity of an annealed zirconium specimen increased from 0.1 μΩ•cm to 0.5 μΩ•cm in the 4 x 10^4 s of irradiation time. Design and construction of a new target chamber is underway, to allow use of a scanning laser, sensitive to 2.54 x 10^{-6} m, for dimensional measurements during irradiation. This will be applied to the thermomechanical behaviour of the blanket.

Additionally, irradiation with spallation neutrons at IPNS (ANL) has shown damage production, measured as a change in electrical resistivity, as a function of solute additions at 4 K. The addition of Ti, Sn, Au, and Dy in Zr all enhance damage production. Post-irradiation annealing results show all four solutes suppress recovery of Stages I and II, suggesting interstitial trapping. Zr with 1000 ppm Ti, Sn, and Au (each in separate samples) has also been irradiated with 14.1 MeV "fusion" neutrons in RTNS-II at Lawrence Livermore National Laboratory. Damage is opposite that

from IPNS. The difference is tentatively attributed to the difference in neutron spectrum; further tests are planned.

A blanket systems element of the program has a number of tasks. There is a liquid metal breeder "watching brief", and a major review of the European program has been completed. Work has progressed on the novel RPI/Grumman/CFFTP aqueous, self-cooled blanket design, and studies of enhanced tritium production in CANDU reactors, and of organic coolant applications to fusion, have been made.

The strategy of the first three-year program has accomplished the basic objective of establishing a Canadian presence on the world scene, and the solid breeder materials route has proved most effective. The challenge of the next five-year period (1987-92) will be to steer the program into a broader range of activities, including liquid metal blankets, while preserving the basis built in the first three years. It is encouraging that many of the topics identified by the FINESSE study on directions of fusion technology are already components of the Chalk River program: advanced tritium recovery experiments; effects of burnup, material, and sweep gas on tritium behaviour; assessment of novel materials (e.g. beryllates); and fabrication development. The long-range goal for the program, with the expertise acquired in other CFFTP programs, is to put in place the Canadian capability for a total fusion fuel cycle package.