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BREEDING RATE MEASUREMENTS IN SOLID FUSION BLANKETS WITH METALLIC LITHIUM SAMPLES*

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ABSTRACT

Measurement of the local breeding rate in a large assembly of fusion blanket candidate materials, irradiated by a fusion neutron source, serves the dual purpose of blanket design support and, perhaps more importantly, calculational method and cross section library testing. In this report, we present technical details of a measurement scheme based on deployment of encapsulated lithium metal samples and subsequent thermal digestion of the samples in a metered carrier hydrogen stream, conversion to THO and LS-counting. A comparison of the scheme to other means of tritium production rate (TPR) measurement with respect to accuracy and other characteristics indicates that its potential accuracy exceeds that of wet-chemistry tritium extraction from lithium salt pellets or TLD deployment and is comparable to the best accuracy of lithium-glass traversing schemes.

The sample fabrication and tritium extraction techniques that will be described evolved from well-tested equipment that was previously used in critical (fission) reactor work and cross section measurements, but needed some modification to increase the throughput and thus allow processing the large number of samples required in blanket assay. The applicability of this scheme to measurements at arbitrarily high neutron flux and higher temperatures will be briefly commented on.

I. INTRODUCTION

The quantitative assay of the amount of tritium generated in neutron-irradiated lithium samples has found a number of diverse applications in both basic and applied physics.

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These include the measurement of lithium reaction cross sections, capture-to-fission ratio measurements in critical (fission) reactor assemblies and fusion breeding blanket evaluation. Encapsulated metallic lithium samples were first used in cross section measurements that still rate as the most accurate available.¹⁻³ The technique of "isotopic dilution" by means of which one can accurately determine the extraction yield of each sample was then adapted to use in fast-reactor critical assembly experiments relying on a ^6Li "reference absorber".⁴⁻⁷ In most of these applications, the carrier hydrogen-tritium mixture is converted to THO and mixed with liquid scintillator for LS-counting, currently the most effective method. Yet another evaluation scheme is based on mass-spectrometric measurement of the ^3He produced in $^6\text{Li}(n,\alpha)t$ reactions as a result of tritium beta decay.⁸

Meanwhile, the somewhat different requirements of fusion blanket breeding assays, emphasizing a large number of individual samples (and accepting, at least initially, a higher margin of error), led to the development of schemes based on pressed-powder pellets of lithium salts, from which tritium is extracted by wet-chemistry methods.⁹⁻¹¹ Other sampling techniques that have been considered or tested in fusion blanket work include lithium fluoride TLD's¹² and lithium-track foil sandwiches.¹³

This, not necessarily comprehensive, survey of sampling schemes seems to illustrate the maxim that the accuracy of results tends to be commensurate with the amount of trouble it takes to obtain them. LS-counting and mass-spectrometric analysis of tritium extracted from encapsulated-metal samples are clearly the most accurate yet also the most difficult, entailing sample fabrication and encapsulation as well as a time-consuming extraction process. Lithium salt pellets are com-

mercially available while wet-chemistry extraction is relatively fast, yet extraction yield and surface loss of tritium can only be estimated, rendering these schemes less accurate. TLD devices are also commercially available, small (hence, readily emplaced) and easily read, but, at present, yield the least accurate result. Track foil sandwiches are especially easy to emplace, yet difficult to read; moreover, while applicable in a thermal neutron spectrum, they are defeated by a large background of proton recoil tracks in a fast-neutron spectrum.

Accuracy and convenience apart, one may usefully consider the potential of different schemes for possible future applications in an ITER blanket module, or a similar deployment at temperatures well above ambient and arbitrary fluence levels. This will be briefly commented on after presenting a description of the Argonne Metallic Lithium Isotopic Dilution (ARMLID) scheme in its present configuration.

The ARMLID system was already fully developed and calibrated¹⁴ when it was first applied in a fusion blanket breeding measurement, in the framework of a series of joint U.S.-Japan investigations whose earlier results have been widely reported; some recent results are being presented at this session. As that work developed, it became apparent that fabrication and processing of up to 60 samples twice per year, in addition to other tasks associated with the experiments, was almost *ultra vires* at the processing speed of 1-2 samples/day provided by the existing hardware. System modifications that were gradually introduced, after thorough testing, allowed the processing rate to be increased to 4-5/day; other modifications speeded up and simplified sample fabrication and encapsulation. The hardware configurations and procedures that resulted from this process will now be briefly described; a more comprehensive description is in preparation.¹⁵

II. SAMPLE FABRICATION

Fabrication takes place entirely within a glovebox of standard design, equipped with port covers, an interlock and a large door. The box is pumped down and backfilled with U.S. grade 8 argon. Lithium metal (natural, depleted and enriched in ^6Li), normally kept in desiccators, is liquefied and poured into an extrusion die whose parts are shown in Fig. 1. A lithium metal ribbon is extruded by

means of a hydraulic ram. Individual foils of about 0.9 cm diameter are cut from the ribbon with a simple "cookie cutter" (also shown in Fig. 1) and weighed to 0.1 mg or, about 1 part in 1500-6000 (for 0.5 mm thick and 2 mm thick foils, respectively). Foils are assembled with 2-S grade, machined and vacuum-annealed aluminum capsule halves. One by one, foil-cover packages are inserted into a cold-welding fixture that again makes use of the hydraulic ram and support cradle, as shown in Fig. 2. After cold-welding at maximum pressure, each package is weighed immediately, and reweighed just before use to verify hermeticity. Lithium stock isotopic composition is routinely verified for a sample taken from each extruded ribbon.

III. TRITIUM EXTRACTION AND COUNTRY

The extraction system, shown in Fig. 3, provides four calibrated storage flasks, filled together but used individually, for U.S. grade 8 carrier hydrogen. Irradiated samples are cleaned and inserted into a stainless steel liner that is in turn inserted into the melting furnace, while the elevator-mounted oven is pre-heated to about 300°C. After pumping down and back-filling the furnace with hydrogen from one of the storage flasks, the oven is raised, rapidly heating the furnace. A record of oven and furnace temperatures and manifold pressure, presented in Fig. 4, shows an initial pressure rise followed by a steep decline as the capsule broaches and lithium hydride forms. Hydrogen as well as tritium is released again when the temperature rises further, to about 750°. As pressure equilibrium is reached, the gas is allowed to pass into a conversion furnace filled with copper oxide; steam formed in that furnace passes on into the back manifold and is frozen onto the wall of a large cold-trap. Thus, back manifold pressure is maintained below 10 micron and a steady draft through the conversion furnace is kept up and regulated by means of a needle valve. The final step in the process is vacuum distillation of the collected liquid directly into a counting vial, attached to the back manifold by means of a special fixture. For typical runs, the yield of this procedure (ratio of collected to carrier equivalent water-weight) comes to .990-.995 at a carrier equivalent weight of about 1.6 g. After adding 18 ml of high-efficiency scintillator, vials are sealed and counted.

For these samples, the LS counting system in current use provides a counting efficiency

Figure 1. Extrusion die parts and foil punch.

Figure 2. Cold-welding equipment: cradle, welding die, and housing.

Figure 3. Extraction system, ARMLID process.

Figure 4. Typical extraction, furnace temperature and pressure record.

of 0.42-0.46 at 4-5 cpm background, allowing even weak samples to be counted with negligible statistical error. Counting efficiency is determined from comparison to diluted NIS tritiated water standard, with a typical error of 0.2-0.5% (including uncertainty quoted by NIS). The combined experimental error attached to the number of tritons per gram sample, calculated at the time of the irradiation, thus comes to about 1-1.5% (excluding accidental factors such as, e.g., contamination of low-counting Li samples by ambient tritium or contamination of the extraction system).

IV. DATA PROCESSING

To take full advantage of this potential accuracy, sample location within the assembly must be determined with commensurate accuracy while calculations would ideally require explicit modelling of the immediate environment of each sample, especially so for samples emplaced in a steep flux gradient (e.g., at boundaries between different materials). However, such fully detailed calculations were not found practical in the mentioned series of investigations. Instead, calculations were limited to the "TPR" or tritium production rate per target neutron and per atom of ^6Li and ^7Li , respectively, in an undisturbed assembly. To calculate that number from the measurements requires two samples of different isotopic composition, exposed to what may be judged about the same neutron environment (with an error of perhaps 2%). A further requirement is a correction for the effect of the sample on the local neutron population that should, in principle, employ the theoretical angular and energy distribution of the flux but in practice has had to make do with the calculated energy distribution and an assumed angular distribution. The error margin of the result thus includes errors attached to the modelling assumptions made by the correction formula as well as the uncertainty attached to the calculated information this formula makes use of. In the above mentioned series of experiments, agreement between pure calculation and measurement-calculation of the local TPR steadily improved from one experiment to the next as both cross section libraries and calculation techniques improved; however, TPR numbers near boundaries still disagreed by factors well beyond the purely experimental error and thus must be ascribed in part to the other errors mentioned, and in part to the calculated corrections.

For many of the experiments, an alternative measurement by a lithium-glass scintillator traversing scheme¹⁶⁻¹⁸ was available. This scheme does require a traversing channel (of which several had been provided in the experimental assemblies) but offers some important advantages: data can be produced immediately after the measurements are done; adequate count rates obtain at low neutron fluences and the equipment is not vulnerable to contamination; accuracy is comparable to that of the ARMLID system.

V. CONCLUSION

In conclusion, we offer some observations on the suitability of ARMLID and other TPR measurement schemes to possible future measurements at higher temperatures and/or under very heavy neutron irradiation. Unencapsulated lithium salt samples are at a disadvantage here as surface tritium loss becomes both large and difficult to estimate. The temperature sensitivity of traversing schemes can perhaps be dealt with through a fiber-optic link between scintillator and PMT,¹⁸ however, supportable countrates would be exceeded unless neutron irradiation can be controlled at low levels. So far as the ARMLID scheme is concerned, metallic lithium is limited to about 150°C and aluminum encapsulant to 350°C. For temperatures between these limits, tests with aluminum-clad lithium oxide have yielded encouraging results.¹⁹ Between 350 and 950°C, copper encapsulant can be used; a background from (n,t) reactions would have to be separately measured and subtracted. Thus, with certain modifications, the scheme appears to have a strong potential applicability in measurements that, whether in the near or more distant future, will be ultimately required.

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