

DETERMINATION OF IN-CORE POWER IN THE LFR 30 kW ARGONAUT REACTOR BY MEASUREMENT OF ^{16}N AND ^{18}F IN THE PRIMARY COOLANT

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As most reactors incorporate out-of-core neutron detectors an accurate relationship between this indicated power and actual core thermal power is required. In relatively high power research reactors this relationship can usually be determined by measuring the thermal capacity of the core using a calorimetric method. However, this may not be possible in low power research reactors and therefore only relative measurements of reactor power might be used on a routine basis.

The present study combines the measurement of ^{16}N , as a relative method, with the measurement of ^{18}F , as an absolute method, in the primary coolant of the LFR 30 kW Argonaut reactor at NRG, Petten, the Netherlands. The measurement of ^{16}N , which is formed by the $^{16}\text{O}(n,p)^{16}\text{N}$ reaction in the primary water of the reactor, has become a common method of measuring relative reactor power by detection of the 6.1 MeV gamma-ray photon. However, the measurement of ^{18}F , which is formed by the $^{18}\text{O}(p,n)^{18}\text{F}$ reaction in the primary water, has been less widely applied to the measurement of reactor power even though its presence in the reactor primary coolant was reported as long ago as 1968. The lack of utilization of the ^{18}F method may be due to the fact that it is a positron emitter and thus its measurement, through detection of the 511 keV gamma-ray photon requires radiochemical separation of the ^{18}F from the primary water. Never-the-less, the ^{18}F method provides a viable method of direct measurement in low power reactors and an alternative for calorimetric measurements in high power research reactors.

This paper will detail the methodology employed to determine actual reactor power, present an improved and reliable radiochemical method for separation of ^{18}F from the primary water, propose that the ^{18}F derived power can be used to calibrate an on-line ^{16}N monitor and suggest that the ^{18}F measurements could be used for power measurements during routine reactor maintenance schedules.

1. Introduction

For reactors operating at significant thermal powers (MW), calorimetric ($\Delta T/dt$) measurements can be used to determine the actual heat flux, and hence thermal power. Where the reactor design, or thermal power constraints preclude this, there is often no easy means of measuring actual thermal power. Accurate power measurement is crucial to the safe operation of any reactor and error analysis techniques are often applied to the nucleonic indicated power, with the indicators calibrated to read power at the upper extreme of the determined error band. This results in a situation where the indicated power may always be higher than the actual thermal power by as much as several percent, and whilst this may be perfectly acceptable for normal operation, the experimental verification of actual power is desirable from a safety justification point.

One common technique for tracking the linearity of indicated power relative to actual power is the measurement of ^{16}N . Produced by the reaction $^{16}\text{O}(n,p)^{16}\text{N}$ with a threshold energy of approximately 10 MeV, this nuclide β decays with a 7.4 s half life to ^{16}O accompanied by the emission of a 6.1 MeV γ photon. Despite the high threshold energy, the abundance of ^{16}O and the energetic γ photon result in a significant contribution to operating gamma dose rates. The 6.1 MeV photons can be detected with HPGE and NaI detectors, but the superior efficiency of Bismuth Germanate (BGO) detectors for high

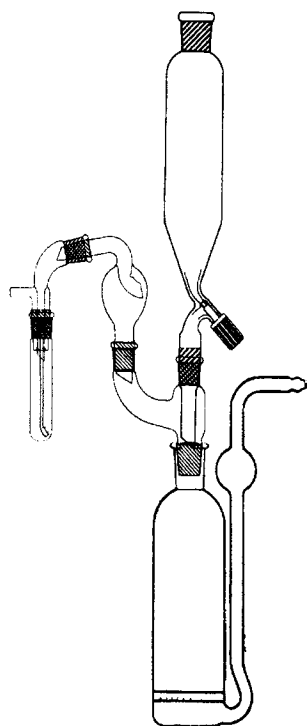


Figure 1
Apparatus used for the ^{18}F
separation

energy photons results in BGO often being the preferred choice. The monitoring of the relative power of nuclear reactors by measurement of ^{16}N in the primary coolant is now a well established [2,5] technique.

Also found in the coolant of water cooled reactors is ^{18}F , with a half life of 109.8 minutes. Fluorine 18 is produced by the reaction $^{18}\text{O}(p,n)^{18}\text{F}$, with a proton energy threshold of about 2.5MeV. Despite the low (0.205%) abundance of ^{18}O , the correlation between the threshold energy of the scattered proton, the peak in thermal fission neutron spectrum and significant (n,p) scattering cross section results in measurable ^{18}F activity even at very low powers. The predominant disadvantage being the positron decay route of the ^{18}F , resulting in difficulty in counting directly due to interference from ^{13}N (among many other positron emitters) and large background count rates resulting from Compton scattering of higher energy emitters than the 0.511 MeV annihilation photon. As detailed below, it is however possible to separate the ^{18}F from the coolant water matrix with an excellent chemical yield, enabling the activity per unit volume, and hence the total induced activity in the coolant water to be determined. From this total activity the fast ($> \sim 2.5\text{MeV}$) flux can be inferred. With a knowledge of the reactor geometry and neutron spectrum (enabling Φ_f / Φ_{th} to be determined) the in-core thermal flux (power) of the reactor can be derived. This paper represents the extension of work originally carried out on the JASON reactor at the Royal Naval College Greenwich[1]. Safety limitations imposed upon the JASON reactor resulted in an operating power constraint of 10kW, whereas the LFR reactor, although very similar in design is able to operate routinely at an indicated power of 30kW. The main objectives of the work were to further improve the chemistry technique, to ascertain the

chemical yield with increased confidence, and to determine the mathematical compensation required for the small design changes between the two plants.

2. Experimental

Separation of the ^{18}F from the coolant matrix exploits the reaction:



The produced trimethylfluorosilane is a volatile, hydrophobic acidic compound, that is separated from the bulk solution by gas stripping and recovered by dissolution in alcoholic sodium hydroxide solution. Figure 1 shows a diagrammatic representation of the chemistry apparatus required for the separation. The separation was performed by drawing air through the equipment at a rate of about $50\text{cm}^3/\text{min}$, the suction being provided by a HyFlo type pump. To effect the separation, the conditions and volumes employed were as follows.

To 200cm^3 of coolant water sample were added; 100cm^3 of 70% w/v Perchloric acid; $150\mu\text{g}$ of fluoride ion, as $75\mu\text{gml}^{-1}\text{F}$ as NaF and 15cm^3 of 6M hydrochloric acid which had been previously saturated with hexamethyldisiloxane by prolonged shaking in a separating funnel. The collection trap was filled with 15cm^3 of 5%(w/v) NaOH in industrial methylated spirit (90%), and air drawn through the apparatus for twenty minutes.

The chemical yield of the method had previously been quoted[1,2] with considerable variation (75-99%), often arrived at by radiochemical means, with the major contribution to the error being the uncertainty of the supplied ^{18}F . Work was therefore undertaken to identify a method suitable for the determination of chemical yield in this matrix at the sensitivity levels required. Various techniques exist for the determination of fluoride ion at low levels, but the approach found most suitable was a variation on the sodium alizarin-sulphonate : zirconium colorimetric method [3]. For this use, a dilute

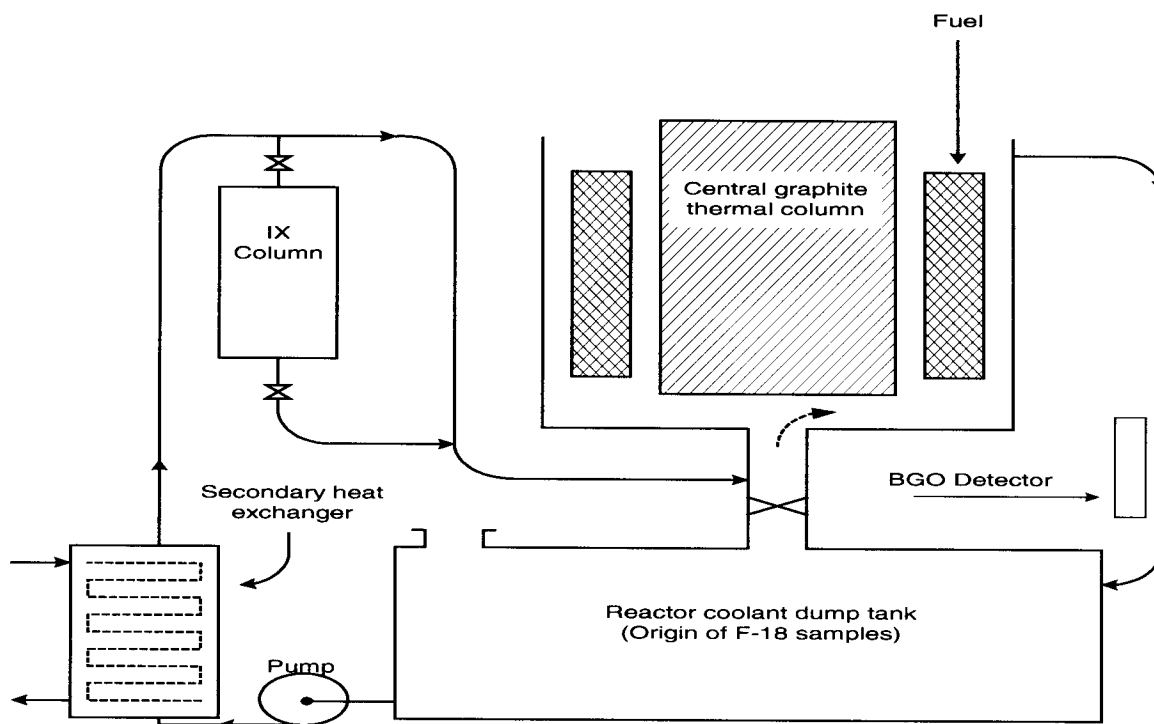


Figure 2.
Basic coolant circuit with sampling positions shown

solution of sodium alizarin-sulphonate was acidified with sulphuric acid before titration with zirconium chloride solution to a colour density considerably short of the obtainable maxima. This solution was then used to produce sets of standards and perform analyses on the contents of the absorber tube. Most variations of this technique utilize the bleaching of colour by the fluoride ion, but by ensuring that the reagent was far from saturated with zirconium, excellent sensitivity was obtained for the low levels required, and no matrix interference effects were observed. Five blank and five inert sample determinations were performed using a Perkin Elmer Lambda5 uv-visible spectrometer measuring the absorbance at 524nm. No fluoride was detected in the blank determinations, with indicated recoveries of 144, 151, 147, 153 and 142 μg in the inert samples. Basic analysis of these figures reveal a yield of 146.5 μg \pm 4.27 μg , or 98% \pm 3% (1 σ), a chemical yield of 98% was then assumed for all sample separations, which were performed in duplicate.

The primary coolant circuit of the Argonaut reactor consists of a nominal 200 gallons (909 litres) of water, divided between a dump tank, the reactor annulus and the associated pumps and pipework. Whilst operating, coolant is continually being recirculated from the dump tank to the reactor annulus, from where it overflows a weir and returns to the dump tank. The heat exchanger on the LFR is normally operated in a fully automatic configuration, such that the temperature of the coolant being recirculated into the reactor is maintained within prescribed limits. Flow over the weir is a constant nominal 14 litres / minute. The time to achieve a mixing factor of 0.99 was estimated using the saturation equation to be 4.5 hours. All ^{18}F measurements were then performed after the reactor had operated at constant indicated power for *at least* this time. Shown in Figure 2 is a diagrammatic representation of the coolant system, it should be noted that the ion exchange column was isolated for the duration of this work, to avoid a requirement to correct for IX purification.

The ^{18}F activity measurements were made using a NaI(Tl) (5 \times 10cm) and an EG&G *Nomad* portable spectrometry system. The efficiency of the system had been determined using a standard prepared from a certified reference solution of ^{22}Na .

The saturated activity of ^{18}F is proportional to the abundance of ^{18}O and the portion of the recoil proton flux with sufficient energy to initiate the reaction. The ^{18}O abundance is known to be 0.205 percent, so from the saturated ^{18}F activity the proton flux can be inferred, and thus the portion of the neutron flux

capable of imparting sufficient energy upon a recoiled proton to initiate the reaction. Previously[4], the LWR WIMS code has been used to estimate the thermal to fast neutron flux (Φ_f / Φ_{th}) enabling the calculation of the thermal flux, and hence the fission rate. With knowledge of the core volume and rate of fission, the thermal power can be calculated :

$$P(\text{watts}) = \frac{\Sigma_f \Phi_{th} V_c}{3.1 \times 10^{10}} \quad (\text{Eqn. 1})$$

Where,

- Σ_f = the macroscopic fission cross section
- Φ_{th} = the in-core thermal flux
- V_c = the fissionable volume of the core

The effective thermal flux was derived from the saturated ^{18}F activity according to the equations:

$$\Phi_p = \frac{A_{^{18}\text{F}}}{N_{^{18}\text{O}} \sigma_{p,n}}$$

(Eqn. 2)

$$\Phi_f = \frac{N_p}{n_p \sigma_{mean}}$$

(Eqn. 3)

$$\Phi_{th} = \Phi_f \frac{\Phi_{th}}{\Phi_f}$$

(Eqn. 4)

Where,

- $A_{^{18}\text{F}}$ = the saturated ^{18}F activity /unit volume
- $N_{^{18}\text{O}}$ = the ^{18}O atom density
- N_p =the proton production rate /unit volume
- n_p =the hydrogen atom density
- Φ_p = the proton flux used to derive N_p
- Φ_f =the fast flux
- Φ_{th} =the thermal flux
- $\sigma_{p,n}$ =cross section for the $^{18}\text{O}(p,n)^{18}\text{F}$ reaction
- σ_{mean} =Mean proton scattering cross section

Power determinations were made on three consecutive days, two at an indicated power of 30 kW and finally one at an indicated power of 10 kW. For the two measurements at 30 kW three spectra from

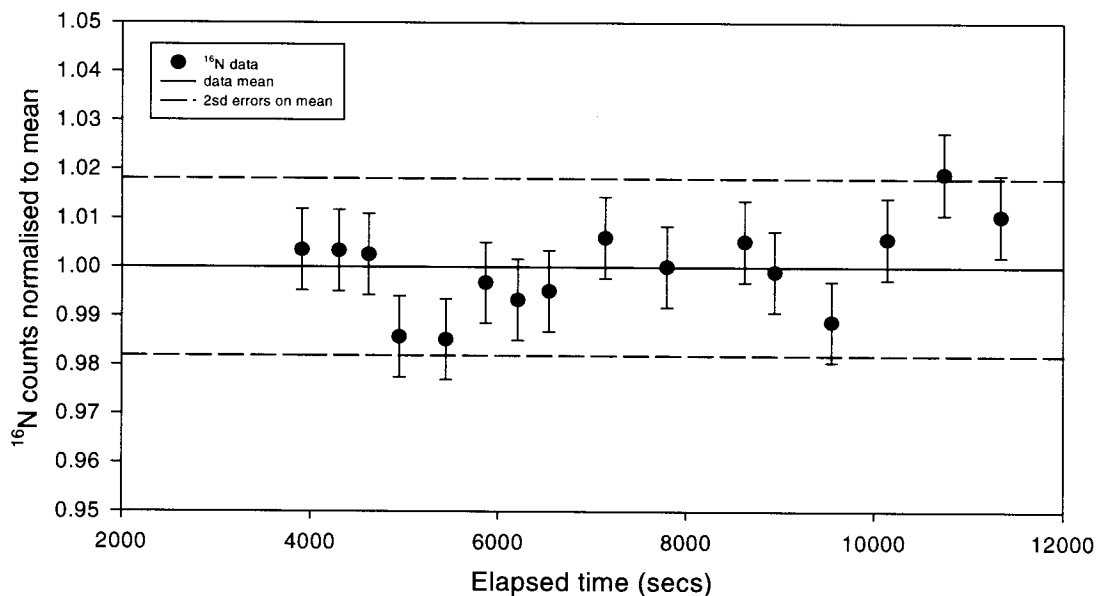


Figure 3.
 ^{16}N linearity data

each of the two replicants were acquired. It was not possible to collect three spectra from each of the 10 kW samples as considerably longer counting times were required to achieve acceptable statistics. The saturated ^{18}F activity per unit volume of coolant was then calculated for each sample set and the ^{18}F indicated power determined using equations 1 to 4. At an indicated 30 kW and 10 kW the mean ^{18}F indicated powers were 32.0 ± 1.7 kW and 8.4 ± 0.9 kW respectively. Two detector systems were used to record the relative variations in ^{16}N activity in the coolant, the NaI(Tl) system described previously and a small BGO (3×2 cm) detector coupled to a counter – scaler. To avoid excessive dead time the NaI detector was positioned approximately 1.5m above the coolant pump machinery pit. A series of 21 spectra for a live time of 200 seconds at approximately six-minute intervals were recorded with the reactor operating at a constant indicated power of 30 kW. These data are presented in Figure 3. The BGO detector was positioned within the machinery pit close to the return pipe carrying coolant from the reactor tank. To confirm that ^{24}Na activity did not cause any significant interference the half-life was determined. When the reactor was shut down the entire coolant volume was rapidly (<2 seconds) dumped into a large dump tank in the machinery pit, the half-life was then determined from a series of repeated 1 second counts. This process was repeated twice and a mean half-life of (5.9 ± 1.2) seconds was found, this was considered acceptable compared with the JEFF2.2 library value of 7.12 ± 0.02 seconds, as allowance must be made here for equipment constraints (difficulties encountered in setting a proper discriminator threshold), and the manual timing of the start of each count.

At an indicated power of 30 kW five 1 second counts were taken, power was then reduced and re-balanced at 20, 10, 5, 1 and 0.5 kW and these counts repeated (counting time increased to 10 seconds at 1 and 0.5 kW). At each new power approximately 2 minutes were allowed to elapse prior to any measurements. Subsequently, power was raised and the measurements repeated at the same powers. These results are shown below in Figure 4.

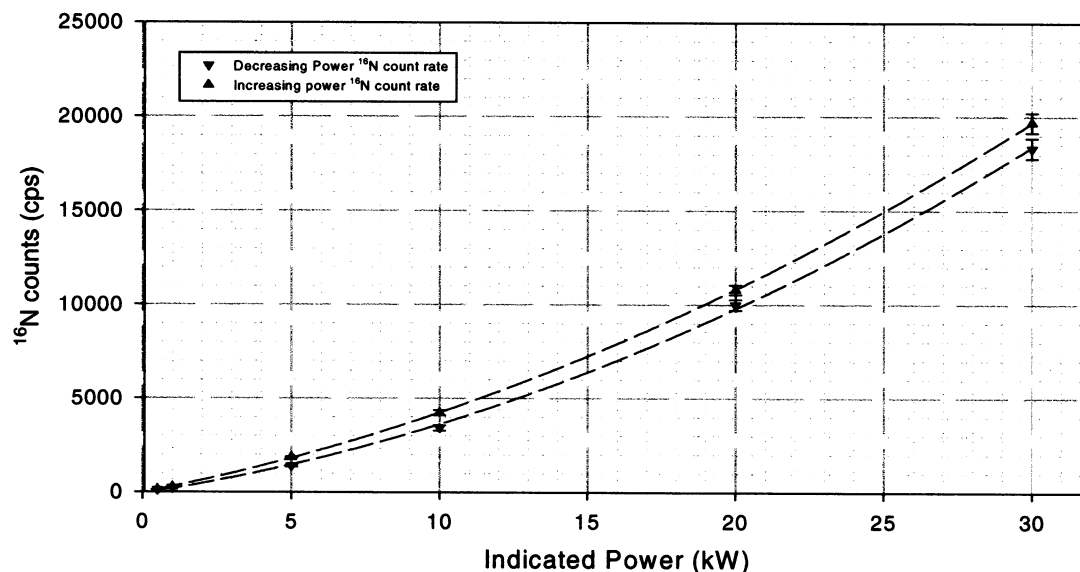


Figure 4.
BGO ^{16}N data - increasing and decreasing power

3. Conclusions and Recommendations

The ^{18}F data obtained are in good agreement with the indicated thermal power, at 30kW the ^{18}F power was determined at 32.0 ± 1.7 kW (106% \pm 6% of indicated). The 10kW data suggests 8.4 ± 0.9 kW (84% \pm 9% of indicated) but allowance must be made here for an error in the linear power indicator that

is understood and normally compensated for by LFR staff. At a nominal 15kW, there is a requirement to switch ranges on the linear channel, and it is observed that the value indicated on the linear power meter of 4.0 (arbitrary scale) reduces to 0.36 on switching (16%down). If this offset were to be considered linear over that entire switched range, the *actual* indicated power would be 8.6kW (1/1.16 of 10kW), a figure that converges well with the ^{18}F calculated power. This calibration offset was also reflected clearly in the power linearity data (see Figure 3) The BGO power linearity data fully supports both the NaI linearity and ^{18}F 10kW data, confirming the indication that actual power is lower at this point. The NaI linearity and ^{18}F 30kW data also confirming that indicated power is slightly less than actual power at the full power calibration point. Based upon the data obtained from this work, it is recommended that the full-range calibration of the linear power channel at the LFR be investigated with a certified current source to ascertain the true response of the channel. Further confirmation of the calibration set points of 10kW and 30kW should be undertaken using carefully prepared foils, and it is anticipated that this work will reflect the results presented here.

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References.

1. JE Moorman, PA Beeley and JP Highton, A review of four methods for the independent monitoring of core power in the Jason Argonaut reactor, IAEA-SM-360/50P, Proceedings of the Research Reactor Utilization, Safety and Management Symposium, Lisbon, Portugal, 6-10 September 1999
2. PA Beeley, JM Brushwood, MG Henesy, MW Collins and CA Haywood, Determination of in-core power in low energy research reactors by measurement of N-16 and F-18 in the primary coolant. Journal of Radioanalytical and Nuclear Chemistry, volume 215, No.1 (1997) 135-139
3. LC Thomas and GJ Chamberlain, Colorimetric chemical methods, 9th edition., 1980, Fluoride method 1, 187-188.
4. M. Robinson, Unpublished DNST Internal report, DNST/ NAC/35/04, 1994.
5. Rowan, NA and Riley, GA., On line measurement of nitrogen 16 in the primary loop of the JASON reactor. Unpublished DNST internal report, DNST/NRC/71, 1994.