THE ISIS METHANE MODERATOR

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1. INTRODUCTION.

A liquid Methane moderator has been in use on ISIS since start-up. This paper reviews the operation of the three moderators which have been used and describes the steps which have been taken to deal with the radiation damage to the Methane. The results of the post irradiation examination of the first moderator are described.

The Methane moderator is one of the prime moderators on ISIS. Its location on the target moderator and reflector assembly is shown in figure 1. The moderator consists of an aluminium alloy can with a volume of about 0.5 litres. The first two moderators had a Gadolinium poison foil 0.05 mm thick positioned centrally through the vessel shown in figure 2. This gives pulse widths in the slowing down region of, typically, $35 \, \mu s$.

2. EXAMINATION OF MODERATOR NUMBER 1

In December 1991 after 4 years of operation at steadily increasing operating current the first moderator blocked.

From outset of ISIS operations there was clear evidence, as expected, of radiation damage to the Methane. When components of the circuit such as filters and circulators were removed for servicing they were found to be wet with 'oil'. Analysis of the oil showed it to be a mixture of hydrocarbons. The viscosity of the oil recovered from the circuit steadily increased with increasing irradiation. In the original design the cryogenic system was emptied by removing the pressure and allowing the Methane to boil off. It was recognised that this would result in the non volatile hydrocarbons remaining in the circuit. However, there was insufficient data available on their production rates to make a reliable estimate of the quantities of hydrocarbon which would be produced.

The neutronic performance of the moderator had shown a steady deterioration from mid 1989 as shown in figure 3. In December 1990 the pressure drop across the moderator became sufficiently large the flow rate of liquid Methane was insufficient to maintain cooling. The system was flushed with Methyl Ethyl Ketone. While a substantial amount of oil was removed the blockage was not cleared and the decision was taken to replace the moderator.

By the time of failure the moderator had been operated for 5 years during which time the integrated number of protons on target was 978 mAh. This had been at two proton energies and both Uranium and Tantalum targets had been used. The irradiation data is summarised in Table 1.

Proton energy	Uranium target	Tantalum Target
550 MeV	22.6	0
750 MeV	628.1	327.5

Table 1. The Irradiation History of Methane Moderator Number 1

The measured neutron flux from the moderator when operating with a Uranium target is two times that with a Tantalum target. Scaling also for effect of proton energy the effective dose to the moderator is calculated to be equivalent to 747 mAh of 800 MeV protons with a Uranium target.

The moderator was dismantled in the remote handling facilities of the Harwell Laboratory of AEA Technology. Figure 4 shows the cuts made through the moderator assembly. The condition of the moderator is shown in figure 5. The can was almost completely full of a solid black substance with some patches of brown discoloration. This deposit was restricted to the moderator can itself. The transfer lines and pipework feeding the moderator was found to be clean. The flushing with Methyl Ethyl Ketone had clearly been successful in cleaning all but the moderator can.

The composition of the deposit was examined by mounting a sample in the LAD spectrometer at ISIS. The structure factor S(Q) shows amorphous features with no Bragg peaks (Figure 6). The pattern is very similar to that of amorphous carbon hydrogen alloys produced by decomposition of hydrocarbon gases which had been a programme of study on LAD. From the shape and variation of S(Q) with scattering angle it can be deduced that there is still considerable hydrogen content.

3. THE LIQUID REPLACEMENT SYSTEM.

It was clear during 1989 that the lifetime of the Methane moderator could well be limited by the effects of radiation damage. Several options to deal with this problem were considered. These included improved filtering to remove the hydrocarbons which are solid at the normal operating temperature and various kinds of phase separators. Finally the decision was taken to redesign the cryogenic system to allow on line replacement of the irradiated methane with fresh liquid.

The assumption made was that the formation of high mass hydrocarbons was a many stage process and could be limited by removal of the damage products at an early stage. For the scheme to work required that the damage products were carried in the liquid flow.

Figure 7 shows a schematic of the new system. A condenser was added to the original circuit, cooled by the main liquid flow, to provide the fresh liquid Methane. Valves were added to make it possible to interrupt the main cooling loop and introduce the fresh liquid Methane which displaces the irradiated Methane into a dump tank. The main cooling loop is then reinstated. The irradiated liquid is then allowed to boil off and any residue can be collected from the dump tank.

The design and installation of this system were in progress when the first moderator blocked. Regular liquid replacement did not start until November 1991.

The system has worked as designed and considerable quantities of 'oil' have been collected from the dump tank. Between 100 ml and 200 ml is not uncommon after a 30 day run of ISIS.

4. THE SECOND MODERATOR.

The second moderator became operational in May 1991. Figure 8 shows the pressure drop across the moderator for the subsequent operational periods. As can be seen the pressure drop had risen significantly by the time routine liquid replacement started.

The moderator blocked early in 1993 after 814 mAh of 800 MeV protons. As with the first moderator the running was split between Uranium targets (335 mAh) and Tantalum (479 mAh) targets. This was equivalent to 575 mAh of 800 MeV protons on a Uranium target which was somewhat less than the integrated beam current on the first moderator when it blocked.

The lifetime of the second moderator was disappointing. There are several comments which can be made.

- The build-up of solid hydrocarbons in the moderator during the first year of operation may have been sufficiently well advanced that the use of the liquid replacement system was ineffective.
- Hydrocarbons form and stick to the surfaces of the moderator and a layer builds up, progressively, unaffected by the liquid flow and gradually blocking the moderator.
- The formation of insoluble, solid deposits is a direct process again building up to form a blockage.

At present there is insufficient evidence to provide a clear understanding of the phenomena involved.

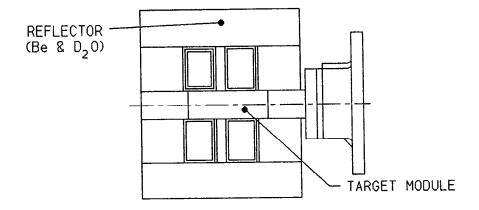
5. PROSPECTS FOR THE FUTURE.

A third moderator was fitted in 1993. This has two poison foils designed to produce pulse widths of about 25 μ s. In operation the pulse width turned out to be 18 μ s but the resulting penalty in neutron flux has proved to be unacceptable. This moderator will be replaced late in 1993 by a single poison foil moderator with the same design as the first two.

The double poison foil moderator will be dismantled after it has been removed and should provide valuable information. The liquid replacement system will have been used from the outset. The intention is to replace the liquid at least once per week. The

condition of the moderator will then indicate the effectiveness of this technique in inhibiting the blockage of the moderator.

If there has been a significant build-up of deposits in the moderator and from this point of view liquid replacement is ineffective there seem to be only two options. The first is to perform some tests to determine whether regular flushing with a solvent will clean the moderator before the deposits are converted to insoluble compounds. The second is simply to replace the moderator regularly. On the basis of the lifetime of the first two moderators this would require annual replacement which, while undesirable from the point of view of scheduling and cost, is not impractical. Investigations are continuing into chemical methods to limit the production of the higher mass hydrocarbons but the initial advice received is not encouraging.



SECTION THROUGH LOWER MODERATORS

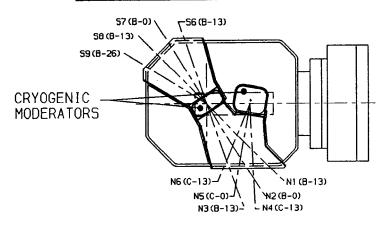


Figure 1. The ISIS Target Moderator Reflector Assembly

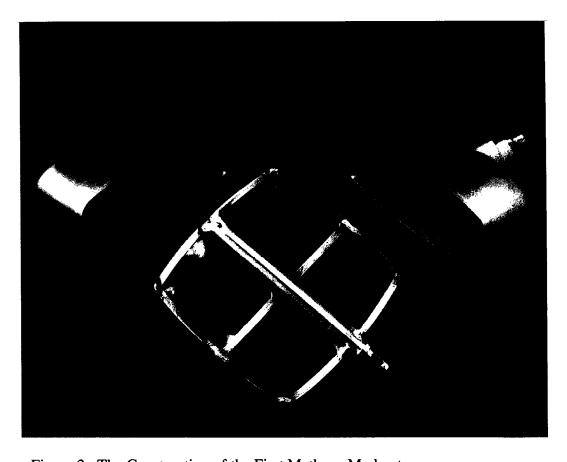


Figure 2. The Construction of the First Methane Moderator

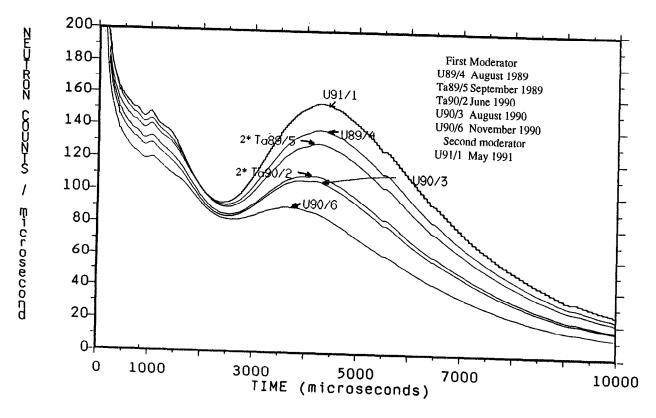


Figure 3. The Deterioration of the Moderator Performance with Time.

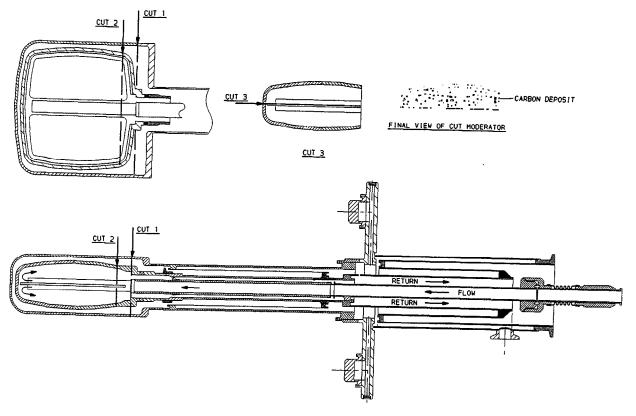


Figure 4. The First Moderator Showing the Position of the Cuts Made to Dismantle it.



Figure 5. The Build Up of Solid Deposits in the First Moderator.

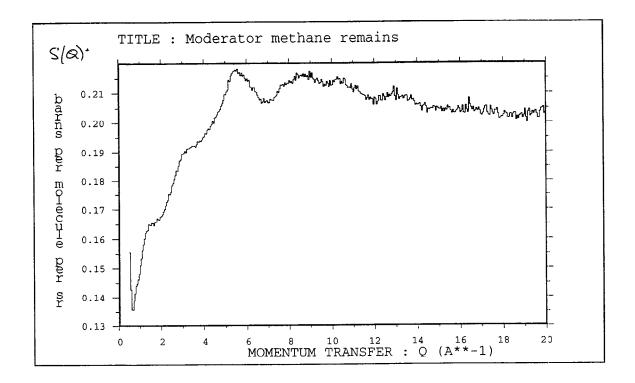


Figure 6. The S(Q) measured on the LAD Spectrometer for a Sample of the Solid Deposit from the First Moderator.

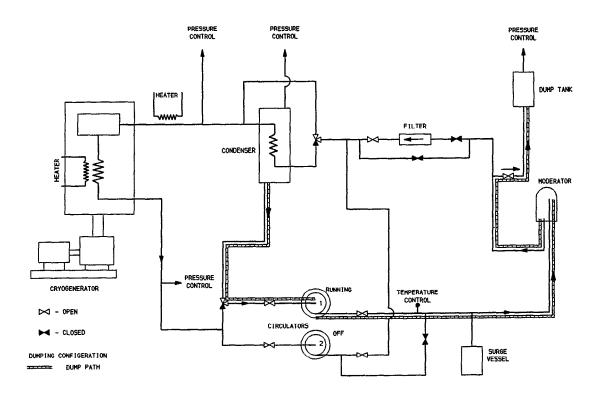


Figure 7. The Cryogenic Circuit for Liquid Replacement

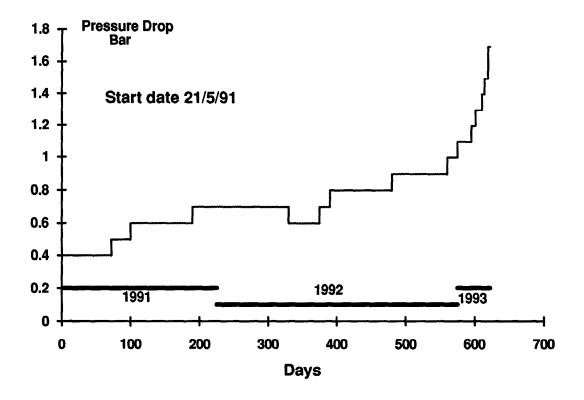


Figure 8. The Pressure drop Accross the Second Moderator as a Function of Time.