ICANS-XIII
13th Meeting of the International Collaboration on
Advanced Neutron Sources
October 11-14, 1995
Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

PROSPECTS FOR TARGETS AND METHANE MODERATORS AT ISIS

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ABSTRACT

This paper reviews the performance of targets and methane moderators at ISIS, the spallation neutron source at the Rutherford Appleton Laboratory. The operational experience is summarised and the plans for the future are outlined.

1. Introduction.

From the inception of the project to build ISIS it was recognised that the choice of depleted uranium for the target and methane as one of the moderators presented a considerable technical challenge. What little was known about the behaviour of these materials in the radiation field of a spallation source indicated that irradiation could cause serious deterioration in their physical properties. The justification for the choice made was the significant gain in neutron fluxes in the beamlines resulting from their use, compared to the alternatives.

Operational experience has confirmed the expected performance. Neutron fluxes from the Uranium target are twice that from the tantalum target, the increased background from neutron emitting nuclides produced in the uranium is, for most experiments, acceptable. The spectrum, intensity and pulse widths of the beams from the methane moderator have been a vital factor in the scientific success of ISIS.

However, the operational experience has shown that, if anything, the original assessment underestimated the effects of irradiation on the materials. This is best illustrated by the lifetimes achieved which is discussed below. A great deal of information has been collected during more than ten years of operation and the research and development programmes currently in progress are aimed at addressing the problems of limited lifetime so there is good reason to be optimistic for the future.

The operational experience is outlined below together with an assessment of the prospects of using uranium targets and methane moderators in the future.

Keywords: ISIS, Targets, Moderators

2. Targets

The design of the target has been reported at previous ICANS meetings [1] so only a brief outline is given here. A schematic diagram is shown in figure 1. The targets are constructed from 9 cm diameter disks held in stainless steel frames. The thickness of the disks varies from 7.7 mm at the front of the target where the proton beam enters to 26.2 mm at the back of the target. Spacers are arranged to give cooling channels 1.75 mm wide between the plates. These frames are welded into a module, fitted with manifolds to direct the water cooling and this assembly then welded into a pressure vessel.

The coolant is D_2O which flows through three main channels, each channel serving up to nine plate cooling gaps. The temperature at the centre of each plate is measured with a thermocouple. The flow and pressure drop of each of the three main cooling channels are measured. A quantity called the 'gap constant', which is flow/(pressure drop)^n is used to monitor the width of the gaps between plates. The index n is calibrated for each target and each of the three cooling channels and typically has a value close to 0.5. In the units used, the gap constant has a value of between 20 and 35 and a reduction by 1 unit means that one plate gap has become seriously blocked. A test rig has confirmed the sensitivity of this method of monitoring the target plate gaps.

The plates themselves are either depleted uranium clad in Zircaloy II or tantalum

2.1 Uranium Targets

To date nine uranium targets have been manufactured and eight installed. The mechanical design has been the same for all as described above.

The lifetimes of both the uranium and tantalum targets are shown in table 1.

Table 1. ISIS Target Performance Summary

Target	Gross Thermal Cycles	Integrated Current mAh	Typical Proton Current μA	Neutron Production mg
U#1	Not measured	92.4	30	75
U#2	40000	53.1	45	52
U#3	10389	174.9	65	163
U#4	4147	138.8	75	128
U#5	5074	295.6	90	273
U#6	2628	126.1	110	116
U#7	1805	107.2	125	99
U#8	Not	Used		
U#9	815	113.2	150	104
Ta#1	73378	1751.6	170	1037
Ta#2 *	21138	618.1	170	366

* Still in use

The lifetime of all the uranium targets has been disappointingly short. To be practical in the context of the ISIS operations a reliable lifetime of at least 400 mAh is required which is equivalent to three user cycles. In all cases the final manifestation of failure is a breach in the cladding releasing a small quantity of fission products into the cooling water. A gamma spectrometer is used to monitor for this.

Targets number 2 and 7 have been dismantled for examination. The results from the examination of target number 2 were reported at ICANS X [2]. The failure of this target was attributed at the time to cladding failure caused by thermal cycling growth of the uranium. Experience with uranium targets at IPNS [3] when combined with that at ISIS no longer supports thermal cycling growth as the primary mechanism for failure.

Temperature monitoring of the plates indicated that the failure in target 7 was associated with a problem in plate 1. The target was dismantled and examined in the Hot Cell facility at AEA Technology, Harwell and the condition of plate 1 is shown in figure 2. There is an obvious bulge in the plate and the cladding had failed. Much of the swelling can be attributed to chemical reaction between the water leaking through the crack in the cladding and the uranium producing a mixture of uranium hydride and oxide which occupies about three times the volume of the uranium. Also outside the immediate region of the central bulge the plate appears wrinkled. This wrinkling is present on all plates from 1 to 19 becoming progressively less pronounced with distance from the proton beam entrance window. The last three plates were not wrinkled. Figure 3 shows the condition of plate 3. With the exception of plate 1 there was no definite evidence of cladding failure although plates 4,7 and 9 had marks which could be interpreted as cracks in the clad.

A jig was constructed to measure the cross section profile of the plates. The aim was to distinguish between distortion with no volume increase and distortion with an associated volume increase. The jig had two direct measuring dial gauges with a resolution of 0.0001 inches. The plate being examined was mounted in the jig and scanned past the dial gauges which measured the profile of the plate on both faces simultaneously. Several plates were measured and a typical scan is shown in figure 4. The results of these measurements showed consistently that a bulge on one side of a plate was always associated with a depression on the other side. No evidence was found for an increase in volume of the plates. It was also clear that some plates had buckled are were no longer flat.

This data was interpreted as evidence for the dominant mechanism leading to failure as being irradiation growth. This is consistent with experience gained from neutron irradiation of uranium in reactors. Growth of individual uranium grains under irradiation is unavoidable but its bulk effects are crucially dependant on the size and orientation of the grains. The Zircaloy is bonded to the uranium using hot isostatic pressing, typically the conditions used are applying pressure to the plate and cladding with argon at 1850 bar pressure at a temperature of 850 C. There is a complex preheating and cooling cycle as part of this process and for the first eight targets manufactured the last part of the cycle was a slow anneal. This results in growth of the uranium grains. Figure 5 shows a micrograph of the uranium structure present in targets 1-8.

The grain size is ~ 1-2 mm. The wrinkling observed on the plates is consistent with growth of clusters of aligned grains of this size.

Development of the heating and cooling cycles used to produce the plates was undertaken to produce a cycle which would result in much smaller and randomly oriented uranium grains. This requires that the last process is a quench of the plate from high temperature. This operation puts substantial stress on the uranium/Zircaloy bond. A temperature cycle was developed which resulted in excellent bond integrity and small random grains as shown in figure 6.

The plates in target number 9 used this production method. The target instrumentation showed the time taken from good condition to failure was very similar to the previous targets [2], the increase in the temperature of plate 1 and the change in the associated gap constant taking about 12 hours. As can be seen in table 1 no increase in lifetime resulted. This implies that irradiation growth is not the dominate mechanism leading to failure as all experimental evidence would suggest that the use of small grains should produce a marked decrease in the bulging and wrinkling of the plates for the same dose.

It is now crucial to the future of uranium targets on ISIS that target number 9 is examined in detail to determine cause of failure. Irradiation in a spallation source has some important differences to that in a reactor and the behaviour of the small grain target was not what was expected on the basis of reactor experience. The main difference is that hydrogen and helium production is much greater in a spallation source than a reactor.

The effect of hydrogen in both Zircaloy and uranium is to form hydrides and thus cause embrittlement. This suggests two possibilities:

1. The embrittled Zircaloy cracked under the temperature stresses and allowed water to come into contact with the hot uranium. The resulting chemical reaction caused the plate to swell and this blocked the cooling channel causing the high temperature.

2. The embrittled uranium cracked under the temperature stresses and caused failure of the cladding. Detailed examination, at Argonne National Laboratory, of an IPNS target showed extensive cracking of the uranium.

Efforts are now underway to define a programme for such a detailed examination.

Finally, two used targets have now been disposed of to BNFL Sellafield. This has required a considerable investment both in a new transport flask and in the remote handling equipment used to load the target into the flask. Both transfers went without difficulty.

2.2 Tantalum Targets

Two tantalum targets have been used so far and the second is the current production target. Neither target has given any significant problems in operation.

The change in cooling characteristics reported previously [4] continued in that the time taken for a target plate to cool after a proton beam trip increased with total proton dose on the target. It is now clear that this is a direct effect of a change in the heat transfer properties of the target plates. The peak target temperature shows an increase with proton dose consistent with the change in cooling time. The peak temperature in target number 1 in its last running period was 320 C which is to be compared to the peak temperature in the second target of 180 C, identical to number 1 except for irradiation, in its first running period.

While for ISIS this effect has no significant effect on operations it is an important consideration when designing new targets. For example, the design of an optimised target for ISIS will have to take into account this change in the ability to remove heat as the total number of incident protons builds up. This will place a limit on the allowable plate thickness which will have to be somewhat thinner than would be the case for an unirradiated target.

The only problem encountered with the tantalum target occurred when it was removed from the target, moderator and reflector assembly. This was done several times in its life either to be replaced by a uranium target or to allow for moderator changes. In storage the target released significant quantities of tritium. The total inventory of tritium in target number 1 has been calculated to be 7 TBq and in storage there has been a steady release of about 500 MBq/day in the form of tritiated water vapour. This is the main source of tritium release from the ISIS target station. It may be affected by the temperature of the target in storage. The storage wells are not cooled and the heat from radioactive decay heat results in a temperature of about 150 C.

The main reason for removing the first tantalum target was to make it available for materials studies as part of the European Spallation Source studies. It is planned to dismantle this target and make detailed examinations of the condition of the tantalum.

2.3 The Future

There are two main options. It is clear that there is a case for redesigning the tantalum target. The current geometry has not been optimised for tantalum and a more efficient design could give and increase in flux of ~15%. The problems of containing the tritium releases from a stored tantalum target will also be addressed. At present there are no plans to change to tungsten.

The future for uranium depends on establishing, unambiguously, the detailed cause of failure. If it turns out that cracking of the uranium leading to cladding failure is the cause then there is no obvious practical solution. The uranium density in the alternative alloys and uranium compounds reduce the gain in neutron flux to the point where the costs of production and disposal of the targets is not longer justified. In this case it is almost certain that no more uranium targets will be produced. Were the cause to be cladding failure then there is a case for seeking an alternative to Zircaloy for cladding the uranium..

3. Methane Moderators

The overall design of the moderator is shown in figure 7. The practical effects of irradiation of methane are production of hydrogen gas and formation of solid deposits in the moderator can. This was described in detail in [5]. It is the production of solid deposits which limits the lifetime of the moderator when the blockage results in insufficient flow to maintain the cooling of the moderator.

3.1 Lifetime

To date six moderators have been used and a summary of their lifetime is shown in table 2. To take account of the factor of two difference in neutron production(and hence dose to the moderator) between uranium and tantalum targets the integrated proton current for tantalum target running has been scaled by 0.5 in this table.

In the original design the liquid methane entered the moderator at the bottom ad left at the top down the central tube. Reversing the flow direction cured an instability in the system caused by the build-up of bubbles of hydrogen gas in the top of the moderator can. However, as can be seen in table 2 this resulted in a substantial decrease in lifetime.

Table 2. The lifetime of the methane moderators at ISIS.

Moderator	Equivalent Integrated Proton Current mAh
#1 single foil	747
#2 single foil	575
#3 double foil reverse flow	198
#4 single foil reverse flow	164
#5 single foil reverse flow modified inlet pipe	142
#6 single foil reverse flow modified inlet pipe	61 * Still in use

(Integrated current is in units of equivalent mAh for 800 MeV protons on a Uranium target)

Moderator 3 was fitted with two poison foils to reduce the pulse width of the neutrons. The reduction in neutron intensity which resulted was unacceptable so the moderator was replaced.

The lifetime of moderator 4 was too low to be practical as the task of changing a moderator takes about 3 weeks. For the ISIS running schedule to be maintained there are only two shutdowns in the year when this can be done. This requires a minimum lifetime of about 400 mAh

The neutronic performance of moderator 4 was acceptable throughout its life. This contrast with moderators 1 and 2 where significant deterioration in neutron fluxes occurred towards the

end of their operation. Moderator 4 was dismantled. Figure 8 shows that the blockage in this moderator was local to the top of the outlet pipe in contrast to moderator 1 where the moderator can was found to be essentially filled with solid [5].

While a development programme, described below, was carried out the methane moderator was replaced by water.

3.2 Development Programme

The development programme concentrated on two main topics, the fluid flow regime in the moderator and the radiation chemistry of methane.

Detailed analysis of the flow in the moderator was carried out using finite element analysis and mock up experiments. It was found that the design for the moderator where methane entered at the bottom the original shape of the outlet pipe resulted in a region of recirculating flow just at the place where the solid deposits were found in the examination of moderator 4. A simple redesign of the pipe orifice removed this as illustrated in figure 9. Moderator number 5 was made with the new pipe design. It operated successfully with no change in pressure drop characteristic of a blockage. It was replaced due to leak in the vacuum insulation can. Moderator number 6 is currently in operation.

A model of the radiation chemistry has been made. Radiative bombardment of methane produces free radicals which react to give higher molecular weight hydrocarbons that form wax at low temperatures. The wax which is insoluble in liquid methane then forms a deposit in the moderator eventually leading to blockage. Irradiation of the wax releases hydrogen and the end product is carbon. The chemical basis of this process is:

$$CH_4 + \text{ENERGY} \rightarrow C^*H_3 + H_*$$

Followed by a reaction where the hydrogen radical reacts by abstracting a further atom from a methyl radical

$$C^*H_3 + H_* \rightarrow {}^*C_*H_2 + H_2$$

The di-radical would then be expected to react preferentially with other di-radicals and build up a chain of repeating methylene units $--(CH_2)_n--$ i.e. the wax.

An examination has been made of the possibilities of reducing the production rate of high mass hydrocarbons and thus hydrogen gas by chemical additives or catalysts. There is a substantial amount of published information on the effects of radiation on methane and on the chemistry of the recombination of free radicals in for example ethane-methane systems. This work has been with room temperature and cold systems. A survey of the literature indicates that the rate of formation of the hydrocarbons which lead to blockage could well be reduced by the addition of chemical agents. This is based on a hydrogen radical being the active species responsible for the formation of the wax. A typical example of this is ethylene ($CH_2=CH_2$) where the hydrogen will react with the double bond rather than a methyl radical. It also appears possible that a catalyst, such as nickel, could be used to cause the hydrogen to recombine with the methyl radicals Both of these processes could reduce concentration of hydrogen radicals in the system and thus limit the production rate of wax.

These ideas will be tested directly in irradiation experiments on methane. The equipment is complete and measurements should start soon.

3.3 Prospects for the Future

Operation of the current design of moderator is satisfactory but requires that the charge of methane is changed by the liquid replacement system [5] every few days. This is due to instability in the circuit caused by the build-up of hydrogen gas in the methane. The hope is that the experiments on the radiochemistry will identify an additive which will both inhibit the production of high mass hydrocarbons with a consequent reduction in hydrogen production. The aim is for a moderator which lasts a full year of ISIS operation.

5. References.

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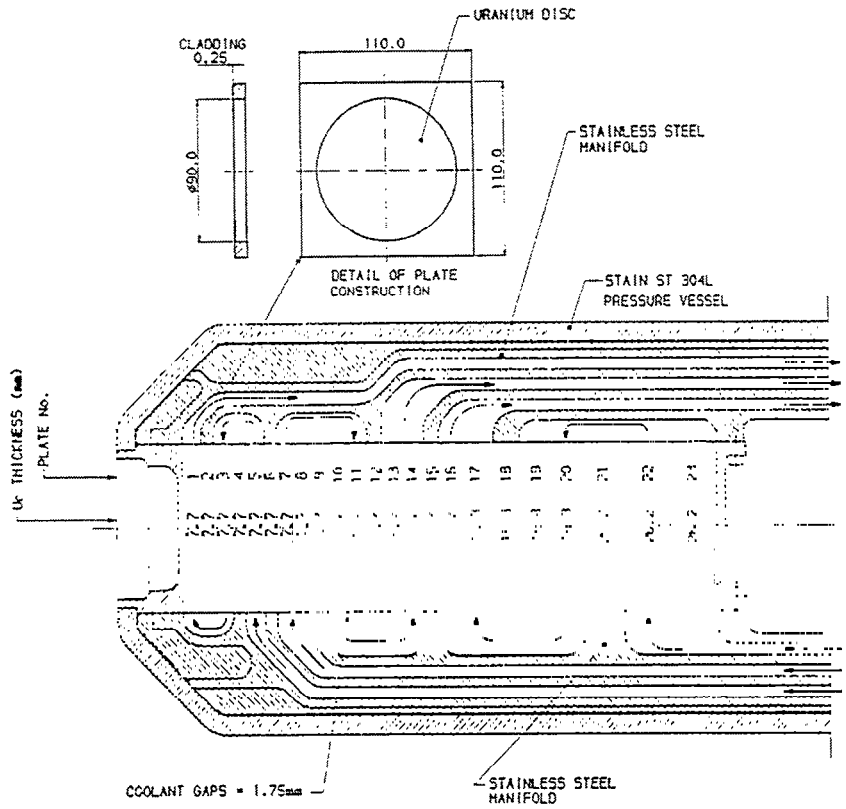
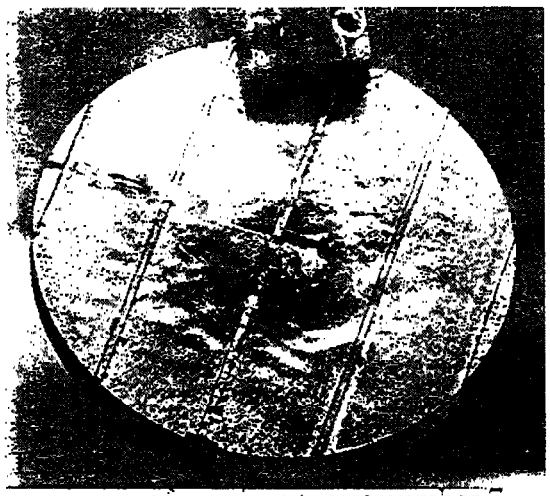
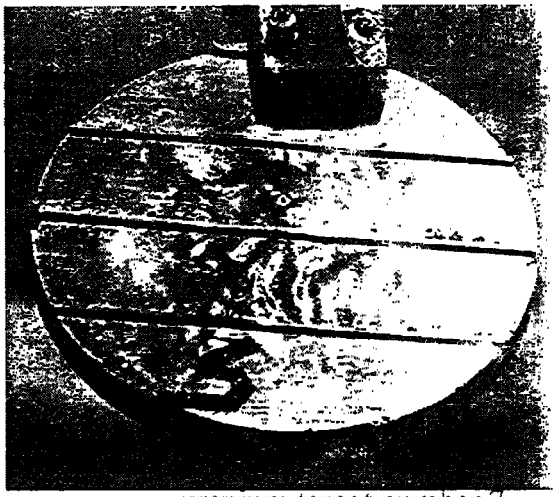


Figure 1. A Schematic layout of the ISIS target.



of uranium target number 7



uranium target number 7

Figure 4. Profile scan of plate number 4, uranium target number 7.

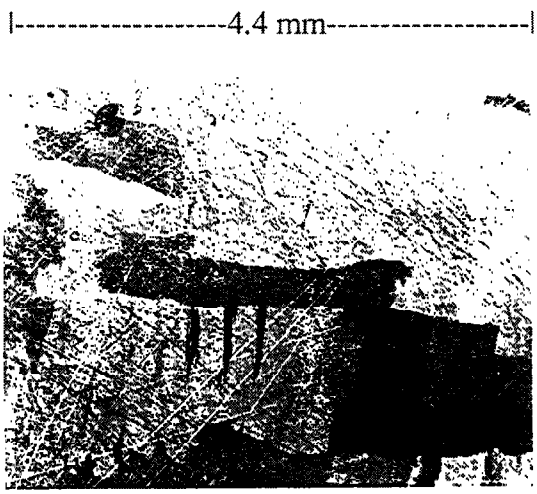


Figure 5. The grain size typical of uranium target numbers 1-8

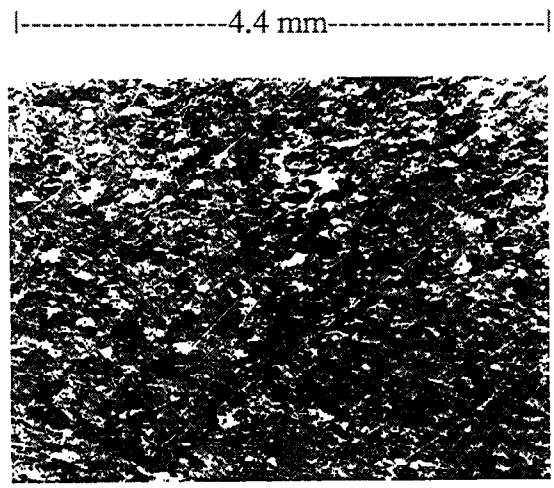


Figure 6. The grain size typical of uranium target number 9

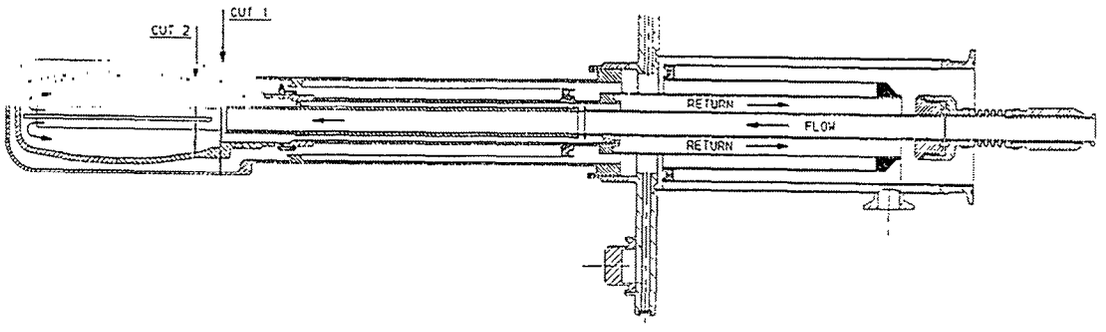


Figure 7. A schematic view of the ISIS methane moderator

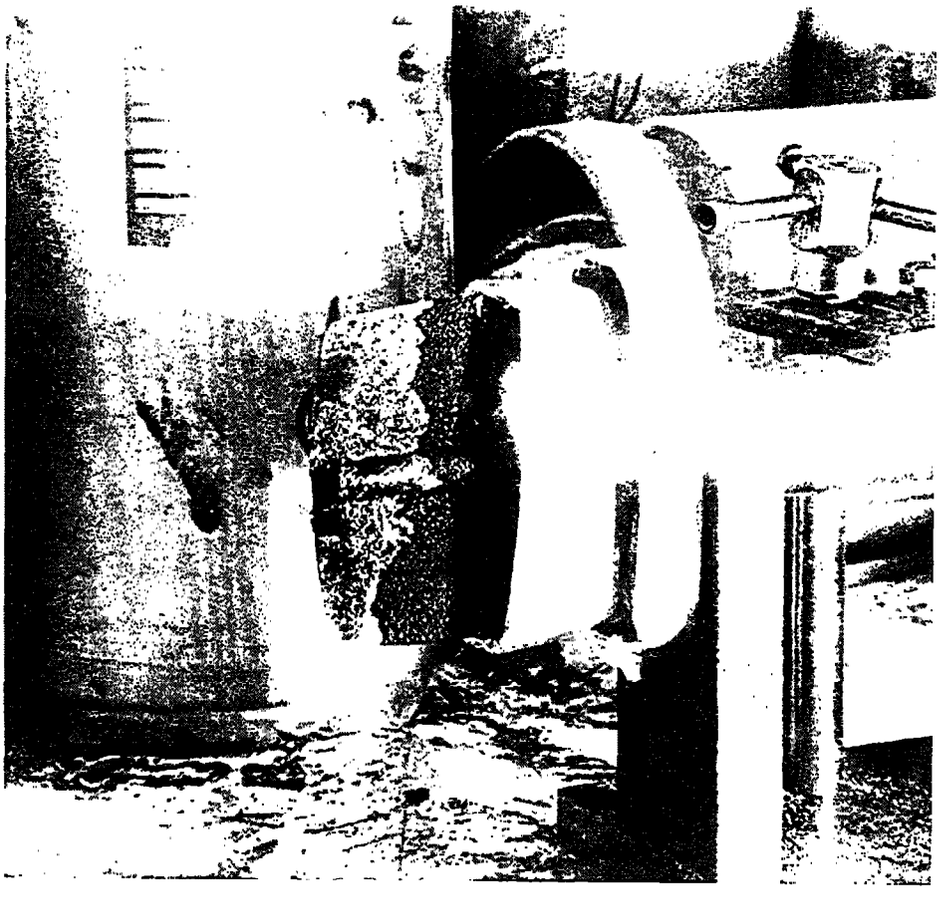


Figure 8. The carbon deposits in methane moderator number 4. Note the build-up at the end of the outlet tube.

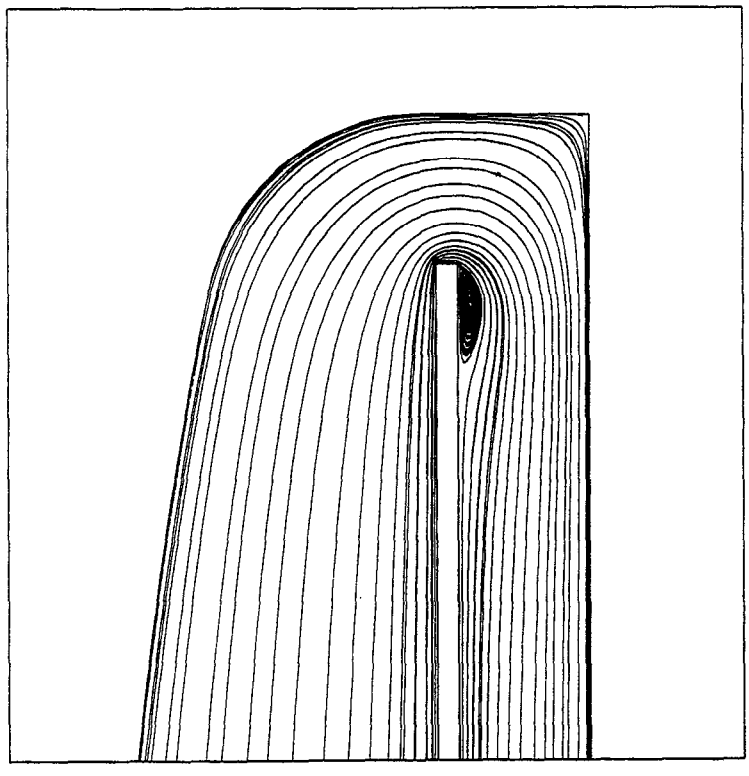


Figure 9a. The flow pattern in the top quarter of the moderator for the original design. Note the recirculation region at the end of the internal tube.

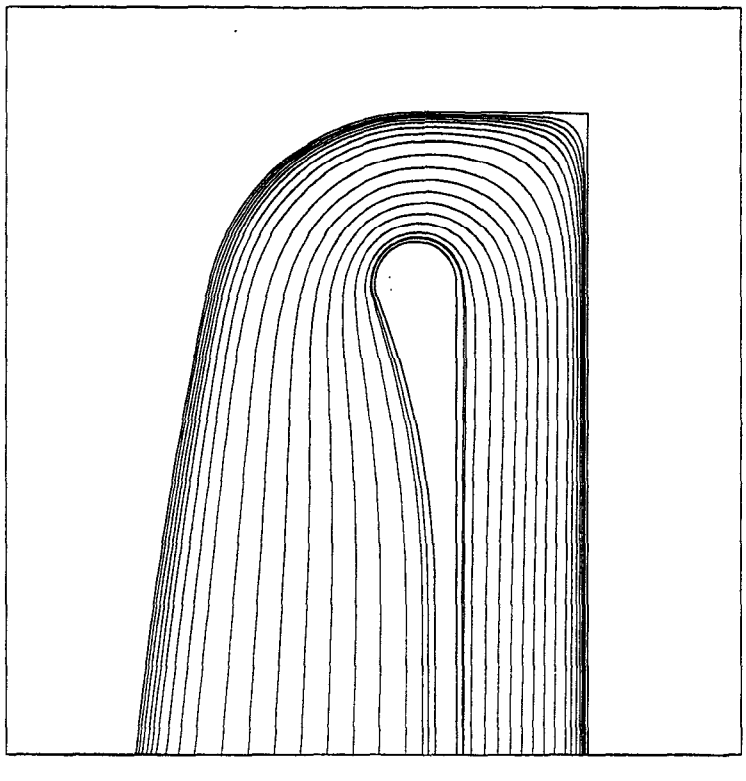


Figure 9b. The flow pattern in the top quarter of the moderator for the modified design with no recirculation region at the end of the internal tube.