Process Filtration of Liquid Methane Radiation Products Using Centrifugal Separation

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Abstract. The radiation chemistry of methane is relatively well known and the creation of hydrogen and long chain hydrocarbons (in the form of oils, waxes and even solid carbon-like deposits) in the presence of a neutron source has presented a challenge to the ISIS liquid methane moderator operation since first start up. The rapid build-up of gaseous hydrogen leads to pressure fluctuation and circulation failure as the pipework becomes gas-locked. Current operational practice sees us replace the methane charge every single day which, while proving effective at removing the built up hydrogen gas, leads to temperature instability and periods where the system is unavailable to the users. Additionally, this approach does very little to curtail hydrocarbon build-up; previous attempts to remove the hydrocarbons using mesh filters have also proved ineffective and instead the system must be regularly flushed with solvent, while the moderator vessel requires complete replacement after only 2 or 3 operational cycles.

We intend to investigate the possibility of using centrifugal separation in order to constantly remove heavy hydrocarbons and hydrogen gas from the process stream of liquid methane. If successful this could realise great benefits in terms of system stability, maintenance costs and beam time available to users. This paper will provide details of the proposed changes to be incorporated as part of the Target Station 1 (TS1) upgrade project.

1. Introduction
The ISIS Target Station 1 uses liquid methane as a moderator material, reducing the energy of fast neutrons created at the target down to thermal neutron levels which are useful to the installed instruments. Within the moderator vessel, the intense neutron radiation causes methane molecules to break apart into atomic hydrogen and free radicals of methane. These then recombine, forming molecular hydrogen (H₂) and a variety of hydrocarbons. As the material is recirculated through the moderator, the hydrocarbon chains progressively increase in length, forming oils and waxes (referred to as ‘polymer’) and eventually resulting in solid carbon-like deposits. Figure 1 shows a moderator can which has become completely blocked as a result of this hydrocarbon formation.

The molecular hydrogen, which is gaseous at operating conditions, builds up very rapidly within the system. After only a matter of hours of operation, the methane system experiences significant pressure and temperature excursions, resulting in a loss of availability to users. It is believed that this is caused by H₂ concentration reaching a level where the performance of the circulation pump is impaired. No such instability is experienced if the system operates with no beam on the target, and...
outside of these momentary excursions the control system is comfortably able to maintain steady state conditions with varying beam current.

2. Previous Work

Although the potential issues with methane radiolysis products were known when the system was first designed, the scale of the problem became apparent only when beam energy was increased after several years’ operation [1].

A theoretical analysis [2] was carried out in order to estimate the rate of build-up of the contaminants, and this work was backed up by examination of irradiated samples [3]. The study estimated that, at current beam energies, approximately 2 liters (at operating conditions) of hydrogen and 0.5 grams of polymer are created within the moderator per day.

The system was originally fitted with two 70µm filters, but these were not found to be effective – the filters did not produce any operational benefit but did create a large amount of radioactive waste. When removed, the filters were found to be saturated with oil, but it is suspected that this only occurs as the system warms up and the oils become liquid.

In addition, a small swirl tank was incorporated into the Cryogenerator head with the hope that the resulting region of low methane flow would allow the hydrogen to naturally bubble out of the liquid. The efficacy of this feature is unknown but operational experience suggests it offers no meaningful improvement.

A major modification was undertaken to the coldbox system in order to allow the full methane charge to be replaced at operational temperature. Gaseous methane is liquefied using excess refrigeration from the process stream over the course of a day, and this is then released into the system, pushing the existing charge into a dump tank from where it can be vented in a controlled manner.

In current operations, the methane charge is replaced every single day in order to prevent hydrogen build-up from reaching a level where the system becomes gas-locking. This represents a significant cost to the organization and the charge change itself inevitably creates large temperature fluctuations in the methane, resulting in periods where the system is unavailable to users. Additionally, the methane moderator vessel requires replacement every 2 to 3 cycles, while the transfer lines linking the moderator and supply system are periodically flushed with the solvent MEK to prevent hydrocarbon build up from blocking the system completely.

3. Continuous Filtration

It is therefore hoped that a means can be found to continuously remove contaminants from the fluid as it circulates, as this could allow the methane moderator system to run for a greatly extended period with much improved pressure and temperature stability. It is proposed that, since H₂ will be in a gaseous state and oils are likely to be solid at the operating conditions, centrifugation of the process fluid could utilise the differences in the relative density in order to separate contaminants from the liquid methane. The replacement of the moderator supply system as part of the TS1 Project represents an opportunity to incorporate new equipment, and two options are currently being investigated, a motor-driven centrifugal separator, and a hydrocyclone separator.
3.1. Centrifugal Separator

A motor-driven centrifuge, in which the methane fluid enters a drum rotating at very high angular velocity, could create large forces, and thus a high degree of separation, in the fluid components. Figure 2 shows how such a unit might be arranged; methane returning from the moderator enters through a central vertical inlet pipe which directs the flow against the wall of the drum. The drum, spinning at very high angular velocity, induces rotation in the fluid and thus subjects it to centrifugal force.

The drum is shaped so that the heavier oils are forced to move outwards and downwards, collecting at the base, while the less dense hydrogen moves towards the top and center. A vent hole allows the gas to collect in a void above the centrifuge, from where it can be periodically released. Clean methane then exits, through outlets at the base, to the Cryogenerator Unit where it is re-cooled to operating temperature.

The design of such a device would share much in common with the pumps currently installed to circulate the methane around the system. Indeed, by angling the outlets of the centrifuge drum, a degree of pumping action could be provided, mitigating the pressure drop created.

A potential shortcoming for this design would be that the centrifuge drum would need to be regularly removed from the coldbox for cleaning in order to remove accumulated oils, unless a novel solution could be found to clean the drum during operation. Given the radioactive nature of this material, this would represent a hazardous operation.

3.2. Hydrocyclone

A hydrocyclone consists of a conical vessel with outlets at the top and bottom. Fluid enters tangentially into the top of the unit and the geometry induces a vortex, causing heavier components to move towards the outer wall and exit through the apex at the base of the unit. At the same time, the flow pattern sets up a secondary vortex moving vertically upwards through the center of the unit. As lighter components are drawn to the middle, this secondary vortex transport them away, exiting through the overflow pipe at the top of the separator.

A combination of two hydrocyclone units could be employed in order firstly remove less dense H₂ from the process stream, followed by the higher density long chain hydrocarbons.

This solution has the advantage that it uses no moving parts and so is essentially maintenance free, hydrogen gas can
be vented through the normal vent system and accumulated oils can be collected and drained during planned shutdown periods.

Hydrocyclonic separators are well established and widely used in industrial processes. However, it is unclear at present whether the low flowrate in the methane system will be sufficient for separation to occur, or if the particle size would be sufficient to be effectively captured using this type of device.

4. Future Work

A prototype hydrocyclone unit has been produced using 3D printing and a test program, initially using water as the flow medium, will be carried out in order to ascertain suitability of this type of unit at low flow rates and to gauge the likely effect on pressure drop in the system.

Further investigation into material properties of hydrocarbons at cryogenic temperatures is needed. Although hydrocarbon samples are readily available – approximately 100ml of oil is drained from the system after every cycle – the highly radioactive nature of this substance prevents any analysis being carried out. This means that the actual composition of the oil cannot be easily determined and so material properties at cryogenic temperatures are unknown. A representative mixture based on theoretical radiolysis models could provide a starting point to address this requirement.

A motor-driven centrifuge will require significant development work and an industrial or university partner is being sought to collaborate in this work.

5. Conclusion

Cryogenic liquid methane has excellent properties as a neutron moderator, but the operational difficulties associated with its use mean that it is not a favored option for this purpose - ISIS is currently the only spallation source using a liquid methane moderator.

The TS1 Project represents an opportunity to address the long-standing operational challenges inherent in liquid methane and it is hoped that, with some development, a centrifugal separator could provide a successful solution to these problems, providing great benefits in terms of system stability, maintenance costs and beam time available to users.

References
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