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Short Research Article

Radioactive waste recovery and enrichment[†]

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Introduction

The former Amersham plc, now GE Healthcare, has produced radiochemicals at its site near Cardiff (known as the Maynard Centre) since the early 1980s. The main isotopes being used on site are carbon-14 and tritium. The Maynard Centre manufactures a wide range of labelled biochemicals and drugs for ADME studies. The radiochemicals are produced either as one–off custom syntheses for customers or as a range of carbon-14 and tritium labelled compounds sold through a catalogue for use in life science research. These processes produce some waste materials which are a complex and constantly changing mixture.

The Maynard Centre is a nuclear licensed site and is regulated by a number of agencies:

- The Nuclear Installations Inspectorate (NII) who are part of the Health & Safety Executive issue the Site License.
- The Environment Agency (EA) issue Discharge Authorizations.
- The Office of Civil Nuclear Security (OCNS) is responsible for security of nuclear material.

Radioactive waste is discharged from the site, within authorized limits, by the gaseous route via the site stacks and via the liquid route to the local sewer into the Seven Estuary.

The site has a low radiological impact on the local environment. However, in the late 1990s, the company resolved to set up a programme to 'To radically reduce discharges to the environment from the Cardiff site.'

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The reasons for this decision were driven partly by the regulatory requirement under The Radioactive Substances Act 1993 to continually assess the Best Practical Means (BPM) to reduce discharges and partly as a move towards achieving the OSPAR objectives of reducing discharges to the North Atlantic marine environment to near zero. There was also a need to reduce long-term storage issues on site and to address local community and press interest.

Results and discussions

Figure 1 shows an overview of the project with the proposed reductions in discharges for the various waste streams. As can be seen, substantial reductions are proposed for all discharge streams as well as a large reduction in the quantity of waste put into store on site. As a result the amount of isotope purchased for use in site operations will be reduced.

The project set up to deliver these reductions was started in 1998 and an initial concept design was completed in 2001. The concept design was further refined and detailed engineering started in early 2004. The plant to process tritium waste is currently being constructed and will be commissioned during 2007.

Tritium waste treatment and enrichment plant

The waste produced by the tritium manufacturing operations is a complex mixture of organic molecules containing tritium together with simpler molecules such as tritiated water and solvents and tritium gas. Liquid waste will be collected in bottles and manually transferred to the waste treatment and enrichment (WTE) plant. Gaseous waste will be collected on molecular sieve traps, following oxidation if required, then manually transferred and purged into the WTE plant (see Figure 2).

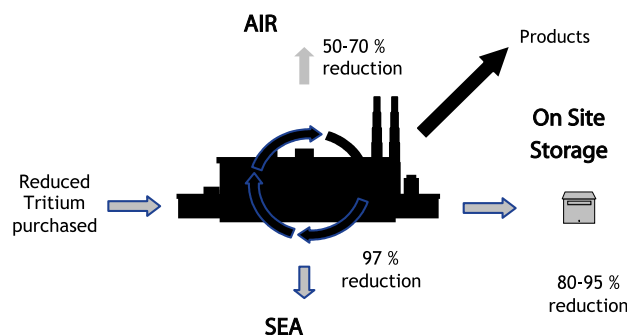WILEY InterScience®
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Figure 1   Project overview. Figure available in colour online at www.interscience.wiley.com
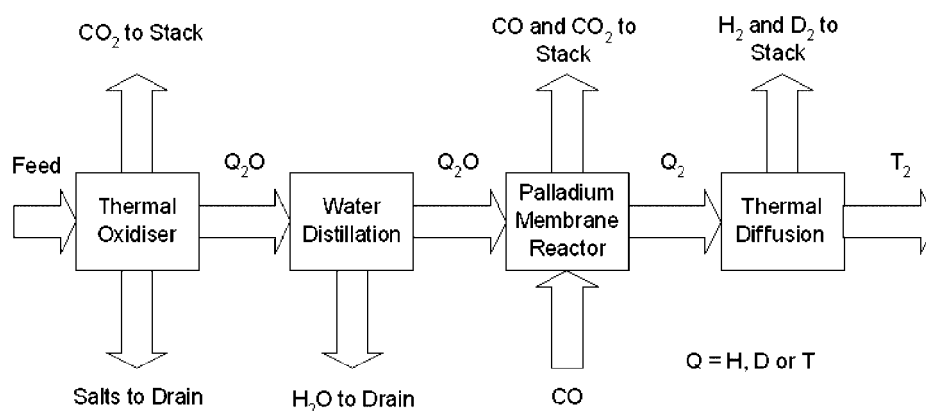

Figure 2   Process flow diagram.

The first stage of the process[1] is to reduce the number of complex chemicals in the waste by oxidation at a temperature above $1100°C$ in the presence of excess oxygen which will convert the complete mixture of organic materials into their most oxidized form, mainly carbon dioxide and water. The gas stream leaving the oxidiser, consisting largely of carbon dioxide, water vapour and excess oxygen, is quenched and the remaining gas stream (carbon dioxide and oxygen) is stripped of any remaining tritiated water, monitored and released. The water, containing about 3 ppm of tritium, is purified and taken on to the next stage.

The next step of the process uses water distillation to carry out the first stage of enrichment. The water is fed into the column and the liquid passes to the bottom of the column where it is boiled. The vapour passes up the column to a condenser where it is condensed and the liquid is allowed to flow back down the column. This counter-current flow of liquid and vapour leads to the tritium being concentrated at the bottom of the column. At the top of the column a small proportion of the condensed liquid, stripped of its tritium, is monitored and released to drain. At the bottom of the column the

water enriched in tritium is taken forward to the next stage. This process leads to an increase in the isotopic abundance of the tritiated water from about 3 to about 2000 ppm. At the same time the volume of water to be fed forward is reduced by a corresponding factor, reducing the scale of subsequent stages. To achieve the necessary separation in a reasonable size of equipment, the column is packed with a high-efficiency stainless steel mesh packing called Dixon rings.

It is now necessary to convert the tritiated water into a mixture of isotopes of hydrogen. This is achieved using a palladium membrane reactor, a device developed at the Los Alamos National Laboratory.[2] The water vapour from the bottom of the water distillation column is mixed with carbon monoxide and argon carrier gas. This is passed through a heated tube packed with a catalyst. Under these conditions a water gas shift reaction occurs forming a mixture of hydrogen (and isotopes) and carbon dioxide

$$H_2O(v) + CO(g) \rightarrow H_2(g) + CO_2(g)$$
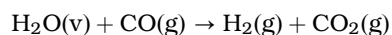
The walls of the reaction tube are made of a palladium alloy and when hot will allow the hydrogen isotopes to pass through and be pumped to the next stage in the

process. The residual gases (carbon dioxide and carrier argon) are monitored and discharged. The mixture of hydrogen isotopes has now to be enriched from ~2000 ppm tritium content to a level when it can be reused in GE Healthcare's manufacturing processes (> 99%). This is achieved using a thermal diffusion process.

The thermal diffusion plant consists of a column (split into two to keep the plant height reasonable) at the centre of which is a wire heated to 1100°C. The wall of the tube is cooled and the thermal gradient across the column causes the heavier isotope to migrate to the outer wall of the tube. Here, the lower temperature causes a downward convection flow whilst at the centre of the column there is a corresponding upward current. Thus, the tritium collects at the bottom of the column and the protium and deuterium move to the top where they are monitored and discharged. When the isotopic abundance of tritium at the bottom of the thermal diffusion column reaches 99%, as measured by an analyser, it is drawn off and absorbed onto tritium storage beds for reuse in the manufacture of radiochemicals.

Choice of technologies[3]

Large-scale facilities to extract tritium from heavy water moderator systems for nuclear reactors have been built in Canada, France and are currently in construction in South Korea.[4] Similar, although smaller, light water tritium recovery systems have been designed for fusion applications. All current large-scale tritium recovery systems employ a front-end process to transfer tritium from water to elemental hydrogen, followed by a cryogenic distillation cascade to perform all or most of the hydrogen isotope separation.

However, hydrogen isotope separation using a cryogenic distillation process has a number of drawbacks:

- Handling of liquid cryogens with associated hazards, such as high-pressure potential upon warm-up and evaporation, thermal stresses due to very low temperature process conditions and the requirement for a vacuum insulated cold box vessel to contain the cryogenic equipment.
- The potential for blockage of process lines due to freezing of impurities.
- Complex and costly process plant.
- Complex operation and maintenance leading to low availability.
- Non-modular process, making it difficult to upgrade and to keep equipment spares.

- Requires batch-operated dryers and a liquid nitrogen absorber to purify feed to the cryogenic distillation cascade.
- Such a process necessitates a large inventory of highly flammable hydrogen.

Water distillation as a method of tritium enrichment is attractive, as it is a robust technology which may be operated at reduced pressure, thus achieving additional safety benefits. The large separation factor between tritiated water and light water leads to modest sized plant.

However, the separation of tritiated water from light water is easier to achieve than the separation of tritiated water from deuterated water. The presence of natural deuterium at approximately 150 ppm means water distillation enrichment of tritium in light water is an efficient process only when the deuterium concentration is small,[5] which corresponds to a maximum practical enrichment in light water of about 1000 times. This degree of tritium enrichment is sufficient in many practical applications to reduce the tritium enriched product flow to a magnitude compatible with one or more downstream thermal diffusion columns, after conversion of water to an elemental hydrogen stream.

Thermal diffusion has been used successfully for small scale tritium separation, even up to ⩾99% tritium. The simplicity of the apparatus makes it attractive but cannot be easily scaled for large throughput. This is because thermal diffusion columns must operate in the laminar flow regime, and scale-up would move column operation into the turbulent flow regime.[6] The alternative of constructing many small thermal diffusion columns in parallel is unattractive when the throughput requirement is large. Thermal diffusion columns also have low thermodynamic efficiency, which while unimportant at small scale, becomes problematic at large scale. Using thermal diffusion for only the final stage of enrichment means that the scale of the plant and hence the inventory of hydrogen isotopes is minimized.

By combining the scalability of the water distillation column with one or more small throughput thermal diffusion columns, the overall process makes optimal use of water distillation and thermal diffusion. The combination of a high throughput initial process having a low radioactive inventory combined with final enrichment of a low chemical inventory of relatively benign tritium gas is inherently safe. Either process option on its own is either unattractive or impractical. Furthermore, the combined water distillation and thermal diffusion system is much simpler to operate than a conventional cryogenic distillation cascade.

There are no complex start-up, operation, or shutdown sequences. The process may be employed in continuous mode with no necessity for batch operations.

Conclusions

GE Healthcare's tritium waste treatment and enrichment project is now reaching fruition. The design of the plant has been completed. A building has been constructed to house the tritium waste treatment and enrichment plant. The interior fitting out has been completed and several of the process modules have assembled off site and delivered. A pre-commissioning safety report has been prepared and submitted to the Nuclear Installation Inspectorate. Commissioning activities have commenced and will continue during 2007.

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