GAS CHROMATOGRAPH ISOTOPE SEPARATION SYSTEM FOR KERNFORSCHUNGSZENTRUM KARLSRUHE GmbH

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INTRODUCTION

The Canadian Fusion Fuels Technology Project (CFFTP) is under contract to supply a tritium recovery system to the Tritium Laboratory (TLK) at the Kernforschungszentrum Karlsruhe (KfK), Germany. This patented recovery system has been designed and developed in Canada by Ontario Hydro Research, and is being fabricated by EG&G Labserco, AECL-SPEL and E.S. Fox Ltd. The system is scheduled to be installed in Germany later this year.

The hydrogen Isotope Separation System (ISS), which utilizes gas chromatography, will become an integral part of the service systems in the TLK. Its basic function will be to separate any mixture of tritium-bearing hydrogen isotopes that might result from various experiments in the laboratory, into distinct isotope product groups. The isotopes are retained on five uranium beds that can be regenerated to allow the tritium to be re-used in the TLK.

The ISS is designed to be apable of operating continuously in a fully automatic mode as a batch processor. The proposed operating cycle is four injections per week at up to 140 NL per injection, with weekly regenerations of all five uranium beds. It is estimated that it will take 130 hours to process the four injections and approximately 16 hours to regenerate the uranium beds. The design throughput of the system is 3 mol/day.

SYSTEM DESCRIPTION

A brief description of the ISS process flow through the following four main sub-systems

- The Feed Collection and Sample Injection Sub-system
- Isotope Separation Unit
- Isotope Storage and Recovery Sub-system
- Carrier Gas Loop

is given below.

The feed stream of mixed isotopes to the ISS is collected in the Feed Collection Tank, and from there it is compressed into the

Sample Injection Loop to form one sample batch. This sample is then injected into the helium carrier gas stream flowing through the Isotope Separation Unit comprising a set of Gas Chromatograph Columns (GCC) immersed in liquid nitrogen. As the injected plug of sample slowly moves through the columns, the isotope species separate and emerge from the GCC as six detectable groups. The carrier gas stream is directed alternately through five uranium getter beds to temporarily store the isotope species in the following five groups:

T_2
DT
HT
D_2 (with some tritium)
the inactive species H_2, HD, and D_2

After passing through one of the U-beds, the carrier gas stream is fed through a Zr/Fe scavenger bed to remove residual traces of hydrogen isotopes, and re-circulated through the ISS loop again via a continuously operating metal diaphragm compressor.

At some appointed time, typically once per week, the separation process is interrupted and the pure tritium is transferred back to the Tritium Storage System by regenerating the T_2 uranium bed. The inactive species are not retained for re-use and are vented from the ISS via the Central Tritium Removal System (ZTS). The HT, DT and D_2 streams are recycled through the ISS again for further refining of tritium arising from dissociation of HT and DT within the system.

The process control of the ISS is managed entirely by a dedicated programmable logic controller (PLC) housed in the local control console adjacent to the ISS glovebox. The control system also includes interfaces with the TLK Safety System that allows the ISS to be shut down independently should the normal process control system fail to do so. A display of ISS process conditions and limited control will also be available in the Central Control Room.

Although the primary circuit is designed to satisfy the specified low leakage rates of 10^{-8} mbar-l/s, the entire ISS (except the liquid nitrogen make-up tank, BD008) is enclosed in a glovebox to form a secondary containment barrier. The glovebox measures approximately 1.5W x 6B x 4H meters and is equipped with gloveports to allow access for maintenance of all serviceable components in the ISS. The pressure inside the glovebox is kept at 10 mbar below ambient pressure in the TLK so that leakage flow through this outer containment is inward into the glovebox.

UNIQUE FEATURES OF THE ISS

Gas Chromatograph Columns

The gas chromatograph column is at the core of the KfK Isotope Separation System. Designed and patented by Dr Chris Cheh, et al, the column is in two parts, each comprising of a number of series-connected subsections that are contained in two separate dewar vessels. Having the column in two stages improves the batch throughput by allowing two consecutive samples to be injected without waiting for the entire sample to elute from the column.

The column subsections are identical in design and are in the form of two concentric stainless steel tubes, 75 mm and 25 mm in diameter respectively, with 5A molecular sieve powder packed in the annular space. Sintered stainless steel frits at top and bottom of the annular spaces keep the fine column packing material in place while still allowing gas to flow through. Shaped stainless steel end-fittings seal the top and bottom of each sub-assembly and facilitate a smooth flow of carrier gas into and out of the annular column packing. The end-fittings also provide an opening for the liquid nitrogen, in which the column is submerged, to flow up the inside of the inner tube so that the radial temperature gradient in the column packing is minimized.

The column subsections are connected in series by 10 mm tubing to form one subassembly for each of the two dewar vessels. The subassemblies are held together by two radial support plates and the whole assembly suspended in the liquid nitrogen from the underside of the dewar flange.

Temperature programming of the GC column

Raising the temperature of the column has a major impact on the elution rate and hence the throughput capacity of the system. The temperatures of the column assemblies are controlled by turning on and off heater elements in the liquid nitrogen and regulating the pressure of the liquid nitrogen, thereby ensuring that the liquid nitrogen is at its boiling state. This design feature allows a fast and even temperature change in the containers, as boiling nitrogen has a much larger heat capacity than in the vapour state, and the heat transfer between the liquid phase and the column is better than vapour.

The liquid nitrogen dewars that contain the GC column are pressure vessels able to withstand 35 bar, thus allowing the operating temperature of the columns to be regulated over the range 77 to 120°K.

Uranium Beds and Scavenger Bed

Another unique feature of the ISS is the design of the Uranium Beds used in the ISS. The ISS has five double-containment U-beds, each having a storage capacity of 15.75 moles of any hydrogen isotope at 50 % stoichiometric capacity, to provide temporary storage of the eluted hydrogen species in the ISS.

These U-beds are "flow-through" beds, differnt from the more usual getter beds, with the full carrier gas flow of 1 NL/s passing through the beds during normal operation. They have been designed by Dr Walter Shmayda of OHRD, and fabricated by AECL SPEL.

The in-flowing carrier gas stream enters the U-bed through a central tube and flows up through the uranium powder dispersed on the several interior shelves. Gas exits from the top of the U-bed assembly. A manual valve welded to the top of each vessel allows for the isolation of that vessel as a unit, independent of connecting tubing and valves.

To retrieve the isotopes adsorbed in a U-bed, the uranium is heated from room temperature to 350°C by means of the internal heater to bring about desorption from the uranium. The heaters in each uranium bed are connected to controllers to regulate the temperature during the recovery process.

During the isotope recovery process, the annulus between the two containers is evacuated to provide a thermal barrier for minimizing heat transfer to the glovebox environment . The annulus also provides a means of containing any tritium which may permeate through the primary vessel.

Downstream of the Uranium beds, a flow-through scavenger bed is included in the circuit to remove any remaining hydrogen isotopes from the carrier gas after leaving either of the uranium beds and before returning to the rest of the carrier gas circuit. The scavenger bed, built by E.S Fox, employs a Zr/Fe alloy getter material enclosed within a stainless steel vessel. It includes heaters and thermocouples to regulate its normal operation, which is at approximately 350°C. For regeneration, the operating temperature is raised to 700°C.

PERFORMANCE RESULTS TO DATE

Following the development of a pilot scale GC system in 1986, a large scale demonstration system for a throughput of 3 mol/day equimolar mixture was designed. This demonstration system was jointly funded by KfK, CFFTP and Ontario Hydro and built by Labserco. The performance results obtained from this demonstration system led to the award of the present contract between CFFTP and KfK.

Many experiments were carried out on the demonstration system to study the performance capability of the system to separate hydrogen isotopes at high throughput. Various temperature programming schemes were tested, heart-cutting operations were evaluated and very large (up to 138 NL/injection) or high tritium (up to 740 Ci) samples were separated in the system. Due to the great flexibility of the system and the time limitation of the project, the operation of the system was not optimized, but the results did indicate that the column performed well and good separation could be achieved even with the large sample.

The system being supplied to KfK has not yet been completed, so at present there is no information on its general performance characteristics regarding tritium separation, control system response, etc. So far, the only results that have been obtained are a few chromatograms of dry air samples done at room temperature. The fact that these chromatograms compare favourably with those of similar tests performed on the prototype columns, indicates that system being supplied to KfK should meet the required performance specifications. Installation and acceptance testing is schedules for the Fall of this year.

BLOCK DIAGRAM FOR THE KfK ISOTOPE SEPARATION SYSTEM