Comparison of [^{11}C]CH_3I yields from 2 in-house Methyl Iodide Production systems – Does size matter?

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The TRIUMF/PET Program is largely reliant on carbon-11 tracers for neurology studies. The reliability and high specific activity radiotracers are key components to the success of the program. Recently, we experienced low in-target [^{11}C]CH_4 yields which prevented us from synthesizing certain low radiochemical yield tracers. To circumvent the problem, a new module was constructed. We report our conversion yields obtained from 2 in-house built CH_3I modules and describe the changes made between the two systems.

[^{11}C]CH_4 is produced in a niobium target as previously described(1). The target contents and helium flushes (approximately 1.5 litres) are transported 50 metres in 3.2 mm stainless steel tubing to a hotcell in the radiochemistry lab that houses the CH_3I module. The target contents pass through phosphorous pentoxide to trap ammonia formed in target and are collected on 2 grams of Poropak N cooled at -196°C. Helium is used to flush nitrogen and hydrogen off the trap upon warming. After flushing, the recirculating pump is started and the [^{11}C]CH_4 is pumped through a 720°C quartz tube containing iodine vapour. An ascarite trap (9.5mm OD x 7mm ID x 12cm length) is placed between the quartz tube and CH_3I trap which is packed with 0.2 grams of Poropak N. Recirculation proceeds until the radiation level on the CH_3I detector levels off. The trap is heated to 180°C and helium elutes the [^{11}C]CH_3I into precursor solution or solvent for quantifying CH_3I.

Methyliodide Systems Description

The first TRIUMF gas phase recirculating [^{11}C]CH_3I system built in 1996 was based on works by Link and Larsen (2,3) with minor modifications. Our first system had a 19mm OD x 16.5mm ID x 30.5cm length quartz tube placed in a 15 cm horizontal oven. The I_2 vapour source was a heated side arm near the head of the quartz tube and temperature was varied from 50°C to 90°C to maintain a constant I_2 concentration. A copper coil with running water was placed at the end of the quartz tube to condense iodine and prevent migration through the system. System pressures during recirculation ranged from 2 to 4 psi and flows were 250-300ml/min for a period of 6 minutes. The [^{11}C]CH_4 trap was in the recirculation loop for this system. The conversion yields of [^{11}C]CH_3I averaged 20% decay corrected based on [^{11}C]CH_4 production. The system worked reliably and made enough dose for injection until we experienced target problems and low yields from our Niobium target. With high demand for scanning tracers to be shared with multiple scanners, the need for another CH_3I system was pushed forward.

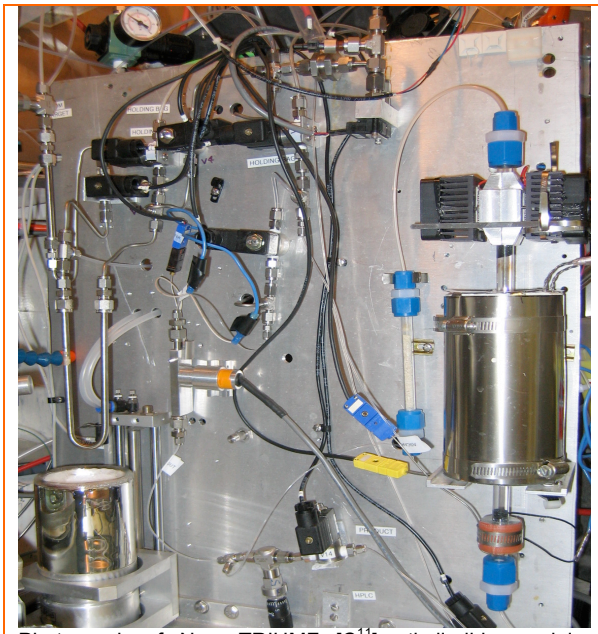
The new system was built with the same model oven rotated into a vertical orientation with a 12.75mm OD x 10.5mm ID x 38cm length quartz tube as the reactor and the flow upward through the tube. The I_2 is now inside a heated portion of the quartz tube (2.5 cm band heater set at 50°C) and sees the flow path directly. A Peltier cooler is used to condense and trap the I_2 vapor exiting the oven to prevent migration through the system. The relatively large volume diaphragm Cole Parmer pump from the original system was replaced with a micro diaphragm KNF pump as the recirculation pump. The system volume was further reduced by replacing the 3.2 mm stainless steel tubing to 1.6 mm teflon tubing where possible. Tubing from the outlet of the quartz tube to the ascarite trap was kept to 3.2 mm due to iodine plating out and causing high pressure and plugging of the system. Fittings were changed to PFA from stainless steel where possible to prevent corrosion in the system. The major difference between the two systems was the recirculation path. After CH_4 trapping, the trap contents were pressurized into the quartz tube. The CH_4 trap was isolated from the recirculation path and [^{11}C]CH_4 was recirculated for 3.5 minutes at a flow rate of 300 to 400ml/min. Pressures during recirculation ranged between 9 and 12 psi.

Results and discussion

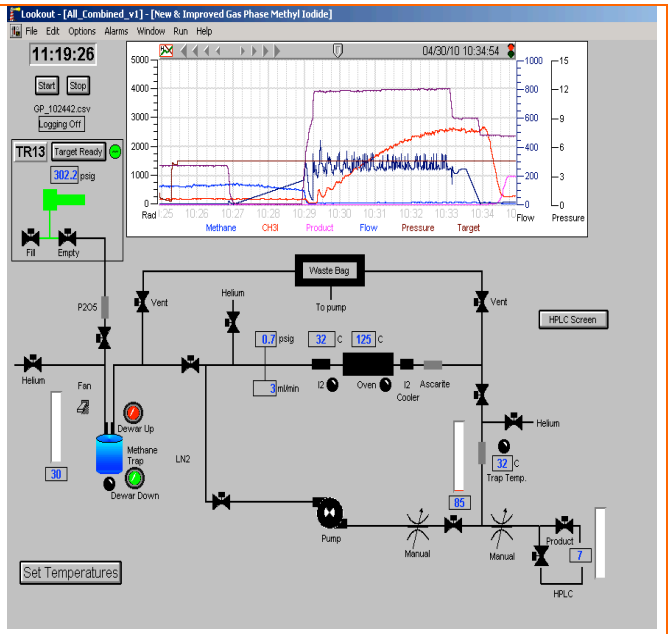
The original CH_3I system provided conversion yields averaging 20%. Due to poor trapping of I_2 after the oven, the ascarite trap was changed between every run, while the I_2 pot was topped up every 20 runs. The system was given a complete cleaning after 60 runs. Upon cleaning of traps, it was found that the CH_3I Poropak packing appeared light yellow in colour proving the breakthrough of iodine and preventing efficient $[^{11}C]CH_3I$ trapping. It was also noticed that the counts on the CH_4 trap radiation detector would rise during recirculation confirming breakthrough of the formed product. With routine maintenance of the system, high specific radioactivity was maintained and the mass of CH_3I produced ranged from 5 to 10 nmols.

With the new system we find the conversion yields increased close to 2 fold and averaged 40% with measured masses of CH_3I ranging between 15 and 25 nmols. We replace the ascarite trap at the beginning of each production day and can perform up to 6 batches with short turnaround time of 20 minutes. The iodine is scraped down the quartz tube for re-use periodically as the vapor concentration decreases thus avoiding the need to add fresh iodine. The system currently has operated with 100 runs without any intervention or I_2 filling.

A smaller recirculation volume allows for larger number of passes of $[^{11}C]CH_4$ through the reaction chamber over the same time period. The original system had a recirculation cycle time of 40 sec per pass providing approximately 10 to 12 passes for the given 6 to 8 minute recirculation time whereas the new system has a recirculation cycle time of 10 sec per pass providing approximately 18 to 24 passes in the 3 to 4 minute recirculation step. In addition, the removal of the CH_4 trap from the recirculation system avoids buildup, and therefore the loss, of any $[^{11}C]CH_3I$ not trapped or bled from the $[^{11}C]CH_3I$ trap. In conclusion, the changes made to the new system with smaller recirculation volume improved the conversion yield of the system.



Photograph of New TRIUMF [C[11]]methyliodide module. Note the vertically mounted quartz tube in the oven, band heater for iodine vaporization below and Peltier cooling unit for iodine trapping above.



Lookout Screen capture of new system. The graph trends target pressure, flow rate and pressure in recirculating loop, radiation detector values for methane trap, methyliodide trap and product.

References:

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