The chemistry of high temperature gas phase production of methyliodide

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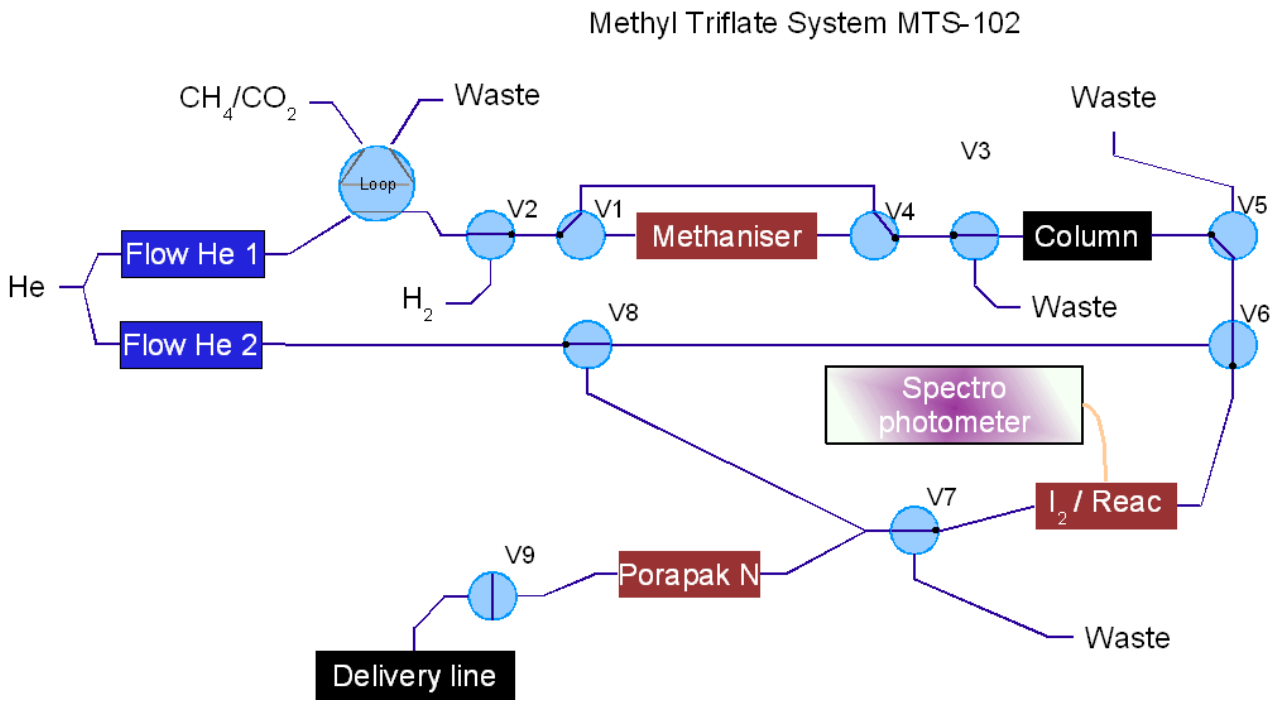
A methyliodide system was set up to react iodine and methane at high temperature in the gasphase (Larsen).

$CH_4 \uparrow + I_2 \uparrow \rightarrow CH_3I \uparrow$

The apparatus consists of an iodine vaporizer, a high temperature (about 700º C) reactor and a Porapak-N methyliodide trap. The lenght of the tube which is heated to the high temperature can be varied.
A known quantity of methane is added from an injection loop or from a methaniser which is fed with carbon dioxide from the injection loop. The methane is transported by a controlled flow of helium through a carbosphere column, which is needed to remove hydrogen from the methane (which is present when starting with methane from a cyclotron and after methanisation). Behind the iodine oven a UV spectrometer is positioned to measure the absorbance in the glastube and the iodine absorbance is used as feedback to regulate the temperature of the vaporizer and thus control the iodine concentration (Link, Clark).

Scheme:



This way all relevant parameters are under control and known quantitatively. The initial amount of methane was choosen as 9 µl, which is the amount of carbon delivered from a cyclotron when producing carbon-11 of moderate specific activity.

The relation between the iodine concentration and the absorbance was calibrated, by collecting the iodine at a stable absorbance during a defined time and weighing the absorbed iodine.

The MeI is collected in methanol (> 90 % is known to be trapped in the first bottle) and analysed by HPLC over an ACE 5 C18 column (15 x 4.6 mm, particle size 5 µm) eluting with methanol / water 60/40 (v.v.) and UV detection (240 nm). A standard solution containing Methyliodide (MeI) and diiodomethane (MeI_2) was used for calibration.

Results

The results given here are preliminary and have to be more precisely calibrated

Transport flow (He)-flow) dependence:

The MeI yield decreases at high and low transport flow. Over a broad flow range, the variation in yield was not significant.

Various flows with a I2 abs of 0.10					
Flow [ml/min]	15	23	30	38	45
Peak area	0.38	0.61	0.39	0.50	0.38
MeI [uMol]	0.026	0.042	0.027	0.035	0.026
Yield [%]	7	10	7	8	7

Iodine concentration dependence:

The MeI yield increases with increasing iodine gas concentration, the maximum concentration still has to be determined:

Various flows with and I2 concentrations resulted in the following yields			
	0.10 I2abs	0.15 I2 abs	0.20 I2 abs
23 ml/min	10	13	17
30 ml/min	7	11	16
38 ml/min	8	13	16

References

Larsen P., Ulin J. and Dahlstrom K. (1995) A new method for production of ^{11}C-labelled methyliodide from ^{11}C-methane. *J. Lab. Comp. Radiopharm.* **37**, 76-78
Linl, J.M., Krohn K.A., Clark J.C. (1997) Production of $[^{11}C]CH_3I$ by single pass reaction of $[^{11}C]CH_4$ with I_2. *Nucl. Med. Biol.* **24**, 93-97