Research Reactor Center
University of Missouri-Columbia

1513 Research Park Drive
Columbia, MO 65211

Phone (573) 882-4211
Fax (573) 882-6360

APPENDIX 1

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Hydrogen Generation Analyses

The volume of hydrogen gas generation rate from radiolysis can be calculated from:

$$V_g = D \times G \times v / A_n \qquad \textbf{Eqn. 1}$$

Where: $\mathbf{V_g}$ is the volumetric hydrogen generation rate in cm^3/sec at standard pressure and temperature

D is the rate of energy absorbed in the liquid (MeV/sec)

G is the "g-value" for the production of molecules of hydrogen per unit of energy deposited (molecules/MeV)

v is the volume of 1 mole of gas at STP (2.24×10^4 cm^3/mole)

$\mathbf{A_n}$ is Avogadro's number (6.023×10^{23} molecules/mole)

D (the rate of energy absorbed) is calculated by knowing the activity of the I-131 and the energy deposited per disintegration or decay. I-131 decays emitting beta particles with an average energy of 0.19 MeV and gammas with an average energy of 0.364 MeV. It is assumed that betas deposit all of their energy in the vial. To determine the gamma-ray energy deposited, MCNP calculations were performed to determine the self-absorption of the gamma-rays in the vial. This fraction ranged from 0.035 for a vial one-half full to 0.045 for a full vial. Conservatively, the 0.045 fraction is used, resulting in 0.364 MeV * 0.045 = 0.016 MeV deposited per decay from gammas. Thus, the total energy deposited per disintegration is 0.19 MeV (betas) + 0.016 MeV (gammas) = 0.206 MeV.

The g-value for the production of hydrogen molecules was obtained from various sources. Elliot (Elliot, A. John, M.P. Chenier and D.C. Ouellette, "G-values for gamma-irradiated water as a function of temperature," *Can. J. Chem,* Vol 68, 1990, p 715, for alkaline nitrite solutions at 25^{o}C) measured 0.38 molecules/100 eV or 3800 molecules/MeV value and 4100 molecules/MeV for pure water at neutral pH. Other values found in the literature include 0.047 umole/J or 4530 molecules/MeV for neutral water (Choppin G. R., J.O. Liljenzin, J. Rydberg J., *Radiochemistry and Nuclear Chemistry,* 2nd Edition, pg 176, Butterworth-Heinemann, Oxford, UK, 2001). A conservative value of 4530 molecules/MeV will be used.

Assuming a maximum of 100 Ci of I-131, the initial dose rate in a vial containing 10 ml is 12.2 Gy/sec (1220 Rads/sec). Integrating this dose rate over a typical transit time of 3 days and including the decay of the I-131 over this time period yields a dose of 2.66 MGy (266 MRad). For the maximum envisioned transit time of 28 days the integrated dose is 8.49 MGy (849

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MRad). (Due to the 8 days half-life of I-131, the 28 day value is 96% of the infinitely long integrated dose and thus, 96% of the maximum amount of hydrogen that will be generated.)

Using **Equation 1** above, the integrated doses at 3 days and 28 days correspond to the production of 28 cm^3 and 89 cm^3 of hydrogen at STP, respectively.

Given that the minimum free air volume above the 10 ml of I-131 solution is 16.2 cm^3 in the vial, the resulting pressures are 25.4 and 81 psig, respectively. Note that any failure of the vial due to 28 days of hydrogen production would result in a release of only 12.5 Ci of I-131 due to its decay over this time period.

<u>Experimental Confirmation of Hydrogen Production</u>

To verify the calculations of hydrogen production given above, I-131 product vials received by International Isotopes, Inc. were tested for increased gas (hydrogen) pressure at time of receipt. A total of four measurements on four separate shipments were taken. Pressure was measured by the simple apparatus shown in the picture below, consisting of a pressure gauge attached to a needle for sampling the pressure in the vial prior to opening.

Picture of Pressure Testing Apparatus in Use at Idaho Isotopes Inc.

Since the volume of gas being measured will expand into the needle and pressure gauge, a detailed analysis of the volume of this apparatus was undertaken and estimated to be 0.997 cm^3, which has been rounded to 1 cm^3. The gas volume in the vials tested (used by a partner facility who receives liquid I-131 in a package approved by a foreign Competent Authority and approved

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by U.S. authorities) is on the order of 20 cm^3, so this correction to the total sample volume during the pressure measurement is on the order of 5%. Any errors in estimating the volume of the measuring system would cause little error in the final result.

The pressure gauge was calibrated using the MURR's dual calibration gauge test stand. This facility is calibrated biannually against a standard provided by KC Calibration Laboratory. All pressure gauges used in these studies were tested and were within ±2% of calibrated pressures.

Table 1 gives the parameters for the four experimental tests, including the time between vial fill time and test time, the volume of I-131 solution and its activity, the air gap volume in the vial above this solution (including the additional volume of the testing gauge), and the measured pressure. Based upon this pressure, the volume of hydrogen evolved during shipment can be calculated and compared with the volume of hydrogen predicted by equation 1) above. These two values are shown on the bottom two lines of the table.

Table 1 –Vial Pressure Test Measurements

Sample	Test 1	Test 2	Test 3	Test 4
Lot ID#	I062114	I062814	I070514	I071214
Test Date/Time	06/30/14 09:50 MT	07/02/14 10:00 MT	07/09/14 10:24 MT	07/17/14 9:30 MT
Fill Date/Time	06/21/14 09:25 SA	06/28/14 08:00 SA	07/05/14 07:12 SA	07/12/14 07:49 SA
Elapsed Time	9.35 days	4.42 days	4.45 days	5.40 days
Nominal Vial Volume	20 mL	20 mL	20 mL	20 mL
Actual Vial Volume	26.2 mL	26.2 mL	26.2 mL	26.2 mL
I-131 Liquid Volume	5.2 mL	5.45 mL	3.63 mL	3.63 mL
Air Gap in vial + test gauge volume of 1 cm^3	21.0 + 1 = 22 cm^3	20.75 + 1 = 21.75 cm^3	22.57 + 1 = 23.57 cm^3	22.57 + 1 = 23.57
Activity at Test Date/Time	26.9 Ci	42.9 Ci	26.9 Ci	29.7Ci
Activity at Fill Time	60.4 Ci	62.9 Ci	39.6 Ci	47.4 Ci
PSIG Measured	21 psig	14 psig	8 psig	12 psig
Measured H_2 Production	**31 cm^3**	**21 cm^3**	**13 cm^3**	**19 cm^3**
Calculated H_2 Production	**38 cm^3**	**24 cm^3**	**15 cm^3**	**21 cm^3**

As can be seen, there is agreement between experimental tests and calculations over a range of shipping times, I-131 solution volumes and I-131 activities.

In all cases the calculated volume is conservative compared to the measured by about 15%. This bias may be due to some recombination of hydrogen with other constituents in the vial or due to conservative assumptions given above and used in calculating the production of hydrogen. For example, reducing the g-value from the conservatively used 4530 molecules/MeV to Elliot's value of 3800 molecules/MeV would essentially eliminate this bias.

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Hydrogen Ignition

Under normal conditions of transport (NCT) all hydrogen will be trapped in the product container within the insert, and no source for ignition exists.

If somehow the product container fails, and the hydrogen escaped into the insert, and then the insert were to leak as well, into the containment vessel, and somehow ignition were to occur, the total energy release would be 966 Joules (231 calories).

The energy content of combustion of evolved hydrogen is negligible compared to the heating of the cask from the decay of I-131. For example, the decay heating rate of 200 Ci of I-131 was previously calculated to be 0.656 watts or 0.656 J/sec which would release 966 Joules of energy in less than one-half hour. Thus, the heating created by ignition of all of the hydrogen generated over 28 days would be negligible compared to the heating of the package by the decay of I-131. Additional perspective is gained by noting that the spontaneous combustion of all hydrogen produced over 28 days would heat a cup of water 4^{o}C.

These calculations and experiments indicate that hydrogen ignition in the case of I-131 liquid contents is not a credible source of risk to the public.