Mono-isotopic Mercury and its Application

INTERNAL USE ONLY

***Abstract*— Methods for creating mono-isotopic mercury samples in the mg range are discussed, their applications evaluated and legal implications considered.**

***Index Terms*— mercury, thimerosal, thiomersal, isotopes, mass spectroscopy, D2O, heavy water, oral polio vaccine OPV**

I. Introduction

THIMEROSAL is the preservative of choice for vaccines. However, irrational consumer reactions hinder its former widespread use. The negative image of thimerosal is mostly due to contained mercury (Hg), which is abundant in toxic waste and thus particularly cheap. It is the common marketing hassle in the health system: The cheapest solution has lowest consumer acceptance. The answer is to find a way of use of thimerosal which goes undetected and thus avoids negative placebo-type reactions.

II. Principle of operation

THIMEROSAL content in vaccines is routinely checked by mass-spectroscopy. Whereas full mass-spectroscopy cannot be fooled, routine checks only investigate peaks in the spectrogram at preset positions. Under measurement conditions, thimerosal breaks up into Hg ions and various organic fractions. For convenience, only the Hg content is monitored by integrating counts around the atomic mass (a.m.) of Hg [1], [1]. Shifting the Hg peak by using isotopes of higher- or lower mass reduces counts within the default window and thus reduces derived Hg content (Fig. 1).

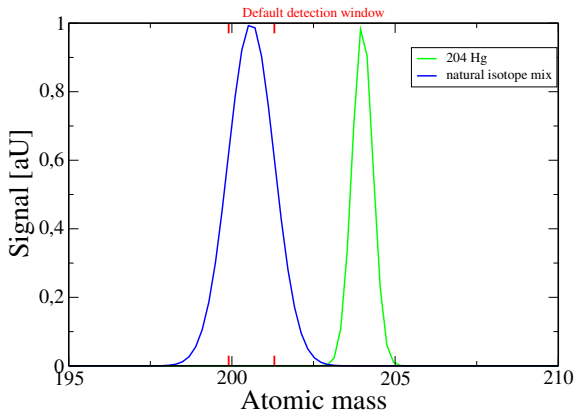

Fig. 1. Shifting the Hg peak in mass spectrometry by using ^{204} Hg

A quick look in the natural distribution of Hg isotopes (Tab. I) clearly reveals that ^{204}Hg is the best candidate for this purpose due to its relatively high abundance of almost 7% and large difference in atomic mass. The alternative ^{196}Hg has an abundancy of $< 1\%$ and is thus too scarce.

III. Preparation

IN principle, mono-isotopic mercury could be prepared by the same process used to enrich uranium. However, due to the military application of enriched uranium usage of such technology is closely monitored and thus not feasible without detection. Separation of isotopes by means of a magnetic field is an alternative, especially as only mg-quantities are needed anyway. Hereby, the same effects are used as in mass-spectroscopic measurement, and indeed it is possible and even easy to prepare mg samples of selected Hg-isotopes in the conventional mass spectrometer available in our laboratory anyway. The only modification required is the shielding of the detector with a target. The selected mercury isotopes will be implanted into the target and can be separated by standard chemical processes later. We managed to augment the build-in detector protection cap with an aluminum foil (Fig. 2). This foil can be changed via the sample door, thus not breaking the vacuum and allowing long up-times. The foil is irradiated with Hg after selecting an atomic mass of 203.97 and NOT removing the detector protection cap (See [2] for operator instructions). Afterwards, the foil is removed and dissolved. Run-times are long though, and over-night use is recommended. This also avoids exposure of the process to uninformed personel.

Fig. 2. Detector protection cap with- and without aluminum target foil.

IV. Results

^{204}Hg used for Thimerosal synthesis reduces the measured Hg content by about 99% (Fig. 3). Given that a Hg content below 0.3 μg Hg/ml is considered Hg-free by the NIH, up to 30 μg Hg/ml are acceptable, which is enough to serve its purpose.

V. Legal considerations

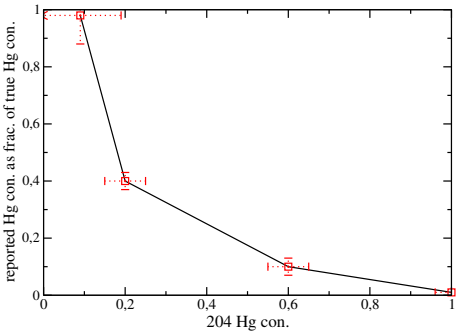

Fig. 3. Reported mercury content as function of ^{204}Hg content

TABLE I
NATURAL MERCURY ISOTOPES. NATURAL ABUNDANCE FLUCTUATES ABOUT .02 % DEPENDING ON MERCURY SOURCE. MERCURY HAS AN A.M. OF 200.59

Isotope	Natural Abundance %	Atomic Mass
196	.15	195.966
198	9.97	197.967
199	16.87	198.968
200	23.1	199.968
201	13.18	200.970
202	29.86	200.971
204	6.87	203.973

WHEREAS health risks of this procedure do not exist despite the minimally higher radioactivity of ^{204}Hg, legal consequences must be considered due to the unpredictability of the legal system. We suggest the following strategy of defense: Should a full mass-spectrographic scan detect the true Hg content, additional samples will be given. A batch of Hg-free sample must be kept ready at all times for this purpose. The intent is to delay a decision by authorities until samples already delivered are silently pulled back or used. Discussion with selected members of the legal department are ongoing, but suggest that legal risks are low.

VI. CONCLUSIONS

WE have found a practical way of using Hg, with all its undisputed advantages, but still avoiding the 'bad image' of Hg. Having considered all consequences, we can fully support usage of this exciting new technology.

Future work should focus on heavy water (D_2O) stabilized oral polio vaccine (OPV) [3], where similar irrational user concerns have to be overcome: OPV stabilized in this way does not require cooling and offers tremendous markets in african countries, where cooling chains cannot be guaranteed. However, H_2O/D_2O are usually detected as molecules, not atomes in mass spectroscopy. This means that their respective difference in a.m. is too low [1]. We consider usage of T_2O, which would yield an a.m. of 22 u.

[1] ~18 u vs. 20 u, resolution at lower a.m. is also lower

REFERENCES

[1] "Standard procedures for mass-spectroscopic monitoring of vaccines", Internal NIH report, file 1996-VZ-3141
[1] Anonymos European Pharmacopoiea 3rd ed. 1997; 0120: 1161
[2] "The Perkin-Elmer Mass-spectrometer: Operators guide", internal document, available at the lab
[3] http://perso.wanadoo.fr/simpson.karl/vaccines.html, a competitor to be monitored

MD5 v2 authenticity fingerprint
6849 2072 6c67 7561 7462 6120
6375 2068 6c61 656c 2073 2e2e