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JRC Reference Laboratory for Radionuclide Metrology

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Working Group Report
Realization of the becquerel at the basic level

Motivation:

In order to prevent the potential loss of the realization of the unit becquerel through the international reference system (SIR) for at present more than 60 radionuclides – for example due to leakage of the SIR chamber – the concept of a reproducible ionisation chamber has been developed by D. Reher, IRMM, and M. Woods, NPL. Contrary to the existing SIR, the new system will be realized via a *set of definitions* allowing reproductions at any time and any place. It is the task of the working group to determine a design and operation criteria for a cylindrical IC system which should guarantee a reproducible output at the level of a few tenths of a percent for different chambers constructed according to these criteria.

At the previous CCRI(II) meeting in May 2005, the membership was revised following staff changes at IRMM and NPL, and further interested laboratories joined (BIPM, LNE-LNHB, PTB, CIEMAT, NIST, NMIJ).

Working group meeting September 2005:

Since the last CCRI(II) meeting in 2005, a working group meeting with 14 participants was held on occasion of the ICRM Conference in Oxford on 5 September 2005.

The concept of the project was recollected and the status of building a prototype chamber at IRMM was presented. Problems of the choice of material for the inner wall and collecting electrode and for the radionuclide ampoules were discussed. Whereas it was decided to stick to glass ampoules due to the lack of alternatives, the wall material needed further studies. Whereas one alternative, aluminium, requires extreme manufacturing tolerances on the wall thickness for reproducible ionisation currents from low-energy gamma-ray emitters, the other alternative, the polyimide Vespel®, requires metallisation, which was unsolved at the time. The working group decided to extend the energy range at the low end to include ^{125}I (30 keV) with good measurement accuracy. LNHB offered support in simulating the influence of different materials and operating conditions using PENELOPE, whereas IRMM would continue simulations with MCNP. The design of the electrometer was agreed in discussions between NPL and IRMM.

Development of project at IRMM:

Since then, the prototype of such a reproducible chamber has been realised and made fully operational at IRMM. First experimental tests with the chamber were performed end of 2005 by A. Švec, J. Paepen and J. Camps. The chamber is connected via a gas handling system to a RUSKA pressure balance, which allows

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gas pressure measurements traceable to the SI. The ionisation current readout is equipped with a Keithley 6517A electrometer. The chamber is not welded but can be dis- and re-assembled to test different configurations. The inner wall is interchangeable: 2 mm thick Al or 6 mm thick Vespel® (a polyimide) which is coated on the gas volume side with a thin layer of titanium.

Experiments with the prototype were done to gain experience with – and possibly optimise – the mechanical and gas handling systems, and to corroborate some findings of the simulations. In particular, the inner walls made of Vespel and Al could be compared. Good agreement between Monte Carlo simulations and experiments with the prototype were achieved. Furthermore, some aspects of simulation reliability were studied by comparison with simulations at LNE-LNHB using a different Monte Carlo code.

Monte Carlo simulations were further refined to better describe the required tolerances on machining (cf. Table 1) and to improve the design. The detailed results of the experimental and simulations work at IRMM during 2006 are documented in the report EUR 22609 EN[1)], which was made available to all working group members.

Table 1: Required machining tolerances for 0.1 % reproducibility in ionisation chamber efficiency at indicated energy. Values derived from simulations and partly experiments.

Item	**Value/statement**	**Required tolerance**
Total length	35-40 cm	1.6 mm (2 MeV)
Radius outer wall	9 - 9.5 cm	70 µm (2 MeV)
Radius inner wall	2.5 – 3 cm	55 µm (30 keV)
Radius electrode	5.5 cm	60 µm (20 keV)
Inner wall material	Aluminium/Vespel	
m (thickness inner wall)	2 mm Al / 6 mm Vespel	1 µm/15 µm (20 keV)
Electrode material	Aluminium	
m_e (thickness electrode)	3 mm	14 µm (30 keV)
Filling gas	Argon	
Gas pressure	2 MPa	2 kPa (2 MeV)
Source positioning	centre	0.9 mm (20 keV, Al)

Working group meeting May 2007:

With the experience gained from the described laboratory work, it was felt that the scope of the reproducible reference ionisation chamber could now be defined better. A meeting was held at BIPM on 22 May 2007 with 10 participants, where the results of the work achieved at IRMM (experimental and simulations), LNE-LNHB (simulations), and NPL (design and use of electrometer) were presented and discussed in very much detail.

1) J. Camps and J. Paepen, Development of an ionisation chamber for the establishment of the SI unit becquerel, Report EUR 22609 EN (2006), ISBN 92-79-04588-1.

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In several aspects of ion chamber operation, more than a single solution is possible, and for some of these, compromises between contradicting requirements had to be found and decided. Based on the decisions taken in the working group (cf. Table 2), more focused studies can be conducted on the remaining open questions such as the influence of glass ampoule edges, diameter of the inner tube, required mass or volume of source, choice of gas, improvement of the source holder. It should be noted that the decision against Vespel® as inner wall material was mainly taken due to doubts about long term availability of material with identical properties and questionable radiation resistance of this polyimide, in spite of much narrower machining tolerances required for the alternative wall material aluminium.

Table 2: Design decisions taken in WG meeting of 22 May 2007

energy range	(20,) 30 ... 2800 keV
which radionuclides ?	(^{109}Cd,) ^{125}I, ... ^{24}Na, also β^- and β^+ emitters
activity range	such that I is minimum 10 x I_{back}
mass of source	??
which solvents	??
material of inner wall	Vespel excluded → take thin Al optional with liner (for β^- and β^+)
diameter of inner wall	?? to be studied by MC simulations, beware ampoule edge effects
outer dimensions	about prototype chamber
welded or open system	?? gain more experience
gas	?? Ar, N, two chambers for both ?
gas pressure	2 MPa (1 MPa ?)
current measurement	implement NPL sampling system
replacement of ampoule	stick to glass ampoule, explore limits (of reproducible manufacturing and of measurement accuracy)
source holder	improve to minimize horizontal displacements
build an improved chamber	wait for further MC and experiments
next meeting	November 2007 ? (depending on progress made)

Provided sufficient progress can be made with the aforementioned studies until September 2007, the next meeting could be planned as early as November 2007. At that meeting the as yet open questions should be decided such that the detailed mechanical design (technical drawings) of an improved prototype can be started.

As working group coordinator I would like to thank all partcipants in the meeting (M.N. Amiot/LNE-LNHB, J. Camps/IRMM, Y. Hino and A. Yunoki/NMIJ, L. Karam/NIST, J.M. Los Arcos/CIEMAT, G. Ratel/BIPM, H. Schrader/PTB, and J. Sephton/NPL) for the open working atmosphere with lively discussions and in particular the representatives of IRMM, LNE-LNHB and NPL for their interesting presentations of recent work.

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