



Ultra-high resolution mass separator—Application to detection of nuclear weapons tests

K. Peräjärvi^{a,*}, T. Eronen^b, V.-V. Elomaa^b, J. Hakala^b, A. Jokinen^b, H. Kettunen^b, V.S. Kolhinen^b, M. Laitinen^b, I.D. Moore^b, H. Penttilä^b, J. Rissanen^b, A. Saastamoinen^b, H. Toivonen^a, J. Turunen^a, J. Äystö^b

^a STUK-Radiation and Nuclear Safety Authority, P.O. Box 14, FI-00881 Helsinki, Finland

^b Department of Physics, University of Jyväskylä, P.O. Box 35 (YFL), FI-40014 University of Jyväskylä, Finland

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ABSTRACT

A Penning trap-based purification process having a resolution of about 1 ppm is reported. In this context, we present for the first time a production method for the most complicated and crucially important nuclear weapons test signature, ^{133m}Xe. These pure xenon samples are required by the Comprehensive Nuclear-Test-Ban Treaty Organization to standardize and calibrate the worldwide network of xenon detectors.

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1. Introduction

Radioactive noble gases produced in nuclear weapons tests offer a sensitive probe for the verification of the compliance with the Comprehensive Nuclear-Test-Ban Treaty (Bowyer et al., 2002). Even in the case of underground detonation it is probable that noble gases are released into the atmosphere (New Scientist, 2007; Ringbom et al., 2009). In practice, the detection effort is focused on ^{131m}Xe ($T_{1/2}=11.84$ d), ^{133m}Xe ($T_{1/2}=2.19$ d), ¹³³Xe ($T_{1/2}=5.243$ d) and ¹³⁵Xe ($T_{1/2}=9.14$ h) (Wernsperger and Schlosser, 2004). Their relative amounts in a sample, in particular the ratio of ^{133m}Xe to ¹³³Xe, are key indicators (Reeder and Bowyer, 1998) to reveal the origin of the sample, i.e., to attribute the source to a nuclear test or to releases from nuclear reactors or isotope production plants. The above mentioned xenon isotopes and isomers are ideal for the verification of the Treaty compliance because of their large production cross sections in fission and long enough half-lives (Bowyer et al., 2002). Below we present a technique for producing pure samples of these isotopes and isomers using the most challenging case, ^{133m}Xe, as an example.

In mass, the isomeric and the ground state of ¹³³Xe differ only by about 2 ppm. Such a small mass difference coupled with the identical chemical behavior make the production of pure ^{133m}Xe samples very challenging. None of the presently available

techniques allow the straightforward production of high-purity (> 90%) samples of ^{133m}Xe. For example, the beta decay of ^{133I} is not a useful method of producing highly enriched ^{133m}Xe samples since it mainly (97.1%) feeds the ground state of ¹³³Xe. Neutron-induced fission coupled to rapid chemical manipulation on the other hand is governed by the direct xenon production cross sections. For example, in case of the neutron-induced fission of ²³⁵U the initial purity (^{133m}Xe/[^{133m}Xe+¹³³Xe]) is around 73% yet ¹³⁵Xe is simultaneously produced with a yield about 19 × higher in comparison to ^{133m}Xe (Bowyer et al., 2002). Due to its shorter half-life, ¹³⁵Xe can be removed from the sample via radioactive decay. Extended decay time, however, also significantly worsens the ^{133m}Xe/[^{133m}Xe+¹³³Xe] ratio.

Laser-based separation techniques do not work well either because the first excited atomic state of xenon is at too high energy to be reached with existing lasers and DC- or RF-discharge sources used to populate metastable states from which lasers can be utilized are rather inefficient. Storage rings do not possess high enough mass resolving power to separate the isomeric and ground states of ¹³³Xe. A Penning trap is an existing device which has the highest potential for reaching the required resolving power for purification (Brown and Gabrielse, 1986). Using well established trapping techniques in a gas-filled Penning trap (Savard et al., 1991), a mass resolution of 20 ppm is routinely reached. A more accurate cleaning method has been recently demonstrated in the Accelerator Laboratory of the University of Jyväskylä, Finland, reaching 4 ppm for ⁵⁰Mn and ⁵⁴Co (Eronen et al., 2008a). For ^{133m}Xe preparation, the resolution had to be

* Corresponding author. Tel.: +358 9 759 88 705; fax: +358 9 759 88 433.
E-mail address: kari.perajarvi@stuk.fi (K. Peräjärvi).

improved by about a factor of two, without sacrificing the throughput.

2. Experimental techniques and results

The desired $^{133\text{m}}\text{Xe}$ nuclides were synthesized using proton-induced fission reactions in a natural uranium target (thickness 15 mg/cm^2). The proton beam energy was 25 MeV and intensity $5\text{ }\mu\text{A}$. The synthesized nuclides recoiling out from the uranium target were thermalized in flowing helium gas which subsequently transported them out from the target chamber. A fraction of the recoils exit the target chamber as ions. These ions were separated from the neutral buffer gas and accelerated to 30 keV of kinetic energy by using electric fields and differential pumping. After acceleration, the ion beam was mass separated using a dipole magnet. In the present work the magnet was tuned to transmit only species with a mass-over-charge ratio of 133 prior to further manipulation. For more information about the used Ion Guide Isotope Separator On-Line technique, see [Äystö \(2001\)](#).

The mass-separated ion beam was transferred to a buffer-gas filled radiofrequency quadrupole structure, which was used to cool and accumulate the incoming ions ([Nieminen et al., 2001](#)). Following this the ions were extracted as short bunches with excellent ion optical properties for the injection into the double Penning trap system JYFLTRAP ([Kolhinen et al., 2004](#)), which was used to separate $^{133\text{m}}\text{Xe}$ in a two-step cleaning process. The first mass-selective cleaning step was performed in the helium gas-filled purification trap. This cleaning step, with a mass resolution of about 30 ppm, could remove all other ions except ^{133}Xe , $^{133\text{m}}\text{Xe}$, $^{133\text{m}}\text{I}$ and ^{133}I . Additionally, some traces of ^{133}Te were observed after this sideband cleaning process, described in detail in [Savard et al. \(1991\)](#). About 250 ms was needed to accomplish the first cleaning step.

The second cleaning cycle was performed in the gas-free precision Penning trap using ion motion excitation with a dipolar RF-electric field using Ramsey's method of time-separated oscillatory fields ([Eronen et al., 2008b](#)). The contaminating ions are excited to large radial orbits while the ion species of interest remain close to the center of the trap. The ions are then extracted back towards the gas-filled purification trap whereby the contaminants are completely removed as they hit a narrow 2 mm diaphragm located between the two Penning traps. In this work, a Ramsey ion motion excitation time pattern of 20–40–20 ms (On–Off–On) was used, providing a mass resolution of about 1 ppm, enough to separate the different nuclear states of ^{133}Xe as shown in [Fig. 1](#). Once the bunch is re-cooled in the purification trap, the isomerically clean sample is finally released for implantation and analysis. This second purification step also needed about 250 ms to complete. Approximately 30 ions per released bunch appeared to be the maximum achievable throughput.

A quadrupole frequency scan was performed in the precision trap to diagnose the peaks of [Fig. 1](#). This scan was made only after completion of both purification steps. When the excitation frequency matches the cyclotron frequency $\nu_c = 1/(2\pi) B q/m$ of the ions, the ions arrive at the detector in a shorter time. In the equation the symbol B represents the magnetic field and q and m correspond to the charge and the mass of the particle. The time-of-flight ion-cyclotron resonances ([König et al., 1995](#); [Gräff et al., 1980](#)) for different cleaning frequencies [A–C] are shown in [Fig. 2](#). An excitation time of 400 ms was used, resulting in a FWHM of about 2 Hz, enough to clearly identify the two states of ^{133}Xe . It is apparent from the time-of-flight resonance curves shown in [Fig. 2](#) that the different ion species are fully separated, i.e., only a single equally deep time-of-flight minimum is observed in each case.

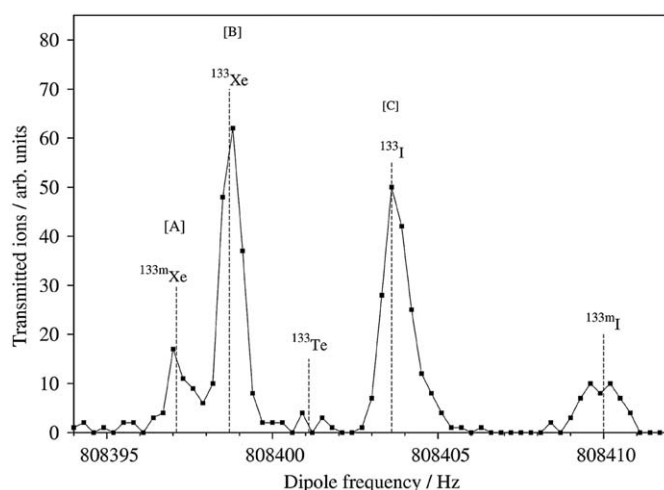


Fig. 1. Frequency scan in dipole cleaning. The number of transmitted ions is plotted as a function of the dipole frequency. The different ion species are marked. The frequencies marked with [A], [B] and [C] correspond to the transmission of $^{133\text{m}}\text{Xe}$, ^{133}Xe and ^{133}I ions, respectively.

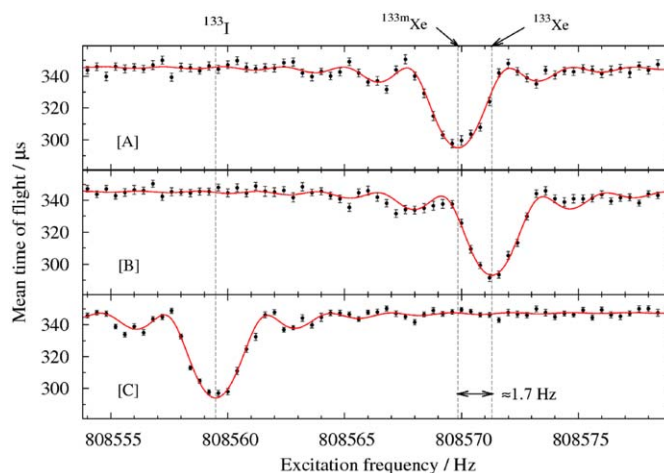


Fig. 2. Time-of-flight ion-cyclotron resonance curves for ion species that are transmitted when the dipole frequency is set to [A], [B] or [C] shown in [Fig. 1](#). The cyclotron resonance frequencies of the three species are marked with dashed vertical lines. The relative mass difference of the states in xenon is only 2 ppm, corresponding to a cyclotron frequency difference of about 1.7 Hz.

The positions of these minima provide unambiguous identification of the two ^{133}Xe and ^{133}I peaks of [Fig. 1](#). Based on the data presented above, the $^{133\text{m}}\text{Xe}$ (or ^{133}Xe) beam purity after the trap is 95%–100%.

The extracted $^{133\text{m}}\text{Xe}$ ions were reaccelerated to 30 keV of energy and implanted into a thin aluminum foil (thickness 3.2 mg/cm^2 , purity 99.5%). [Peräjärvi et al. \(2008\)](#) demonstrates that the xenon does not rapidly escape from the aluminum foil while using such implantation settings. The typical $^{133\text{m}}\text{Xe}$ implantation rate was approximately 60 ions/s. With the current system performance, a sample of 10^6 atoms can be prepared in less than 5 h. A sample size of 10^5 $^{133\text{m}}\text{Xe}$ atoms is large enough for the calibration of the noble gas sampling instruments ([Bowyer et al., 2002](#); [Fontaine et al., 2004](#); [Stocki et al., 2004](#)). This corresponds to 0.37 Bq initial activity. Altogether four samples containing from 80 000 to 6.9×10^5 $^{133\text{m}}\text{Xe}$ atoms were successfully fabricated during two accelerator beam-time periods. The

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