Gaseous $^{83m}$Kr generator of monoenergetic electrons based on $^{83}$Rb deposited in zeolite

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Abstract. The gaseous $^{83m}$Kr electron source is currently used in neutrino mass experiments KATRIN and Project 8, dark matter experiments XENON, LUX and DarkSide, and ALICE (CERN) experiment. The main attractive features of this radioactive noble gas are its monoenergetic conversion electrons with well known energies and a half-life of 1.8 h, which is short enough to avoid any long-lasting contamination of the system. The long half-life of the mother $^{83}$Rb isotope ($T_{1/2} = 86.2$ d) enables more time demanding measurement. Particularly, in the neutrino mass experiments with gaseous tritium in which the $^{83m}$Kr is applied in the same manner as the tritium, the K-32 conversion electrons with energy conveniently close to the beta spectrum endpoint represent an important test and calibration tool. Here, the design and characteristics of the gaseous $^{83m}$Kr generator, including the $^{83m}$Kr source itself, for KATRIN (KArlsruhe TRITium Neutrino) experiment are presented.

1. Introduction

The radioactive noble gas $^{83m}$Kr is currently used for an accurate energy calibration of detectors in neutrino and dark matter experiments. Due to the energy of its monoenergetic conversion electrons and ability to share space with source medium, it can be especially helpful for calibration and systematic measurements in tritium based experiments, such as KATRIN. KATRIN aims to achieve a sensitivity on the effective electron antineutrino mass of 200 meV/c² (90 % C.L.) [1].

2. The gaseous $^{83m}$Kr source

The gaseous $^{83}$Rb/$^{83m}$Kr source is based on the mother isotope $^{83}$Rb deposited into the zeolite beads. The zeolite cation-exchange properties makes them an ideal $^{83}$Rb ion trap [2].

2.1. Source production

The $^{83}$Rb is produced at the U-120M cyclotron of NPI of the CAS (Řež, Czech Republic) using a target filled with natural isotopic mixture of krypton [3]. Using 26.5 MeV protons at 15 μA beam and initial krypton pressure of 13 bar, more than 50 MBq of $^{83}$Rb per hour of irradiation is produced in reaction $^{83}$Kr(p,xn) $^{83}$Rb.

2.2. Gaseous $^{83m}$Kr release

In order to study $^{83m}$Kr release from such a source, a γ-ray spectroscopy technique using measurement of the 32.2 keV γ-line intensity was applied. This γ-line deexcites the isomer $^{83m}$Kr. In such a way, direct information about the amount of the $^{83m}$Kr atoms present in the source was obtained. It was concluded that the most suitable zeolite beads are those of type 5A. Newly prepared source of this type...
releases more than 80% of all the $^{83m}$Kr atoms produced in the $^{83}$Rb decay while the $^{83}$Rb is tightly bound in the beads [3, 4, 5]. If exposed to air with water vapours present, the release gradually falls down to 20%. However, the high release can be restored by heating the source. The $^{83m}$Kr release doesn't degrade if the source is kept in vacuum or exposed to dry gases such as H$_2$, He, N$_2$, O$_2$, even at various pressures. Moreover, preliminary measurements confirmed that the $^{83m}$Kr release is not influenced even in tritium environment.

3. The $^{83m}$Kr generator technical realization

Due to the nature of the KATRIN experiment, all the components are under the risk of tritium contamination. Therefore, they must be made of stainless steel or other suitable metal with a leakage rate bellow $10^{-9}$ mbar ls$^{-1}$. Thus, the $^{83m}$Kr generator body, see figure 1, mostly consists of original and customized Swagelok VCR components and the vacuum gauge.

![Figure 1: The $^{83m}$Kr generator.](image)

3.1. The shielding

The shielding, which is housing the $^{83m}$Kr source, consists of eight lead cylinders with a total weight of 150 kg. The cylinders allow easier manipulation during the transport, source insertion and exchange. Its robustness guarantees that the dose rate directly on its surface will not exceed 3 $\mu$Svh$^{-1}$ (or 1 mSv per year) for sources with activity of $^{83}$Rb up to 2.5 GBq.
3.2. *The ceramic oven*
Custom made ceramic oven is located in one of the lead cylinders. With height of 4 cm it enables partial insertion of the well with the $^{83m}$Kr source at the bottom. Thus the source can be heated directly in vacuum using DC. It requires power supply and controller.

3.3. *The filters and the U-tube*
These components serve as a barrier against $^{83}$Rb release. The sintered element filters with pore size of 0.5 $\mu$m help to stop the small zeolite abrasions and aerosols, which might contain traces of $^{83}$Rb, to spread further.

The U-tube, a tube in a form of the letter U, is supposed to serve as yet another $^{83}$Rb trap if needed. It is believed that Rb is efficiently trapped on cooled surfaces due to its chemical properties and the melting point of 39 °C. Therefore a combination of the dry ice and alcohol is seen as a U-tube coolant. It has to be manually put into the cooler, a Dewar flask around the U-tube, which is not showed on figure 1.

4. The $^{83m}$Kr generator characteristics
After the source is inserted into the well, pre-heated and cooled down in vacuum, another 18 hours are needed to reach the $^{83}$Rb/$^{83m}$Kr equilibrium. Then, the $^{83m}$Kr activity follows the $^{83}$Rb half-life of 86.2 day [6]. The $\gamma$-ray spectroscopy measurements show once again that more than 80 % of all $^{83m}$Kr atoms produced in $^{83}$Rb decay are available at the generator output if no part of it is cooled down.

The $^{83m}$Kr activity is lower by 7 % if the U-tube is sunked into the Dewar flask filled with mixture of dry ice and alcohol. Further investigation of the coolant influence on $^{83m}$Kr activity showed that if the same Dewar is filled with liquid argon instead of the dry ice, the activity falls to 60 %. In case of liquid nitrogen, it falls even further down to 20 %. In both cases it was also strongly dependant on the length of the cooled path.

Up to now, the $^{83}$Rb emanation at the generator output was measured only without the U-tube cooling. After 14.5 days of generator operation followed by 7 days of $^{83}$Rb activity measurement a promising result of 37 $\mu$Bq/h for 190 MBq $^{83}$Rb source was obtained as an upper limit for the $^{83}$Rb emanation.

5. Summary
The $^{83}$Rb/$^{83m}$Kr source emitting monoenergetic conversion electrons is useful in several experiments. The technical realization of the gaseous $^{83m}$Kr generator for KATRIN experiment prevents long-term contamination of the system, while having the advantage of being customizable without compromising radiation safety of the personnel even with sources 2.5 times stronger than KATRIN required.

Acknowledgments
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