Nuclear Instruments and Methods in Physics Research B 271 (2012) 48–54

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Dismantling and decommissioning: The interest of passive neutron measurement to control and characterise radioactive wastes containing uranium

F. Jallu [a,*], A. Reneleau [b], P. Soyer [a], J. Loridon [a]

[a] Commissariat à l'Energie Atomique, CEA Cadarache, France
[b] AREVA NC, UPA/EMP, BP16, 26701 Pierrelatte cedex, France

ARTICLE INFO

Article history:
Received 1 July 2011
Received in revised form 16 September 2011
Available online 7 October 2011

Keywords:
Waste
Passive measurement
Neutron
NDA
Uranium
Dismantling

ABSTRACT

This paper deals with the evacuation of the wastes resulting from the dismantling of an ^{235}U enrichment facility. Gamma-ray spectrometry is usually used to quantify the ^{235}U residual mass and the activity of those wastes. In the present case, the waste internal content made of aluminium prevents gamma-rays from reaching the detector. We show here how neutron passive measurement has been used as an alternative to gamma-ray spectrometry. The controlled objects are about 1000 compressors of masses up to 2000 kg employed in a gaseous diffusion enrichment process. A dedicated neutron-based setup has been designed using Monte Carlo modelling, and a Detection Limit Mass (DLM) of about 4–6 g of ^{235}U is reached. For one of the compressors, the ^{235}U measured mass has been compared to the same mass determined by destructive methods. The comparison shows a very good agreement.

1. Introduction

Non-destructive nuclear measurement methods [1–5] are often used to control and characterise radioactive waste packages, which constitute the final step of the nuclear fuel cycle (Fig. 1). Nowadays, more than before, the dismantling and the decommissioning of nuclear facilities are entirely integrated into this cycle, which leads to a great volume of technological radioactive wastes. Those wastes need to be characterised in order to be sent to the adequate final disposal or interim storage.

The dismantling of the ^{235}U enrichment facility for military purposes located in Pierrelatte, France, results in the characterisation of big volume, metal compressors, among other wastes. Due to their complex, internal metal composition, gamma-ray spectrometry, which is usually used in that field, can not be applied to that special waste. The application of passive neutron measurement was then studied as a gamma-ray spectrometry alternative, and led to the development of a setup aiming to control the ^{235}U fissile mass.

The design basis of a setup, according to performances and detection limit criteria, was made using Monte Carlo calculations. The setup was then built and used to preliminary, performance tests. This setup is now installed in the dismantled facility, in order to be routinely employed. More than half of the compressors have currently been measured.

Section 2 presents the context of the study, and the principle of the method. The modelled design basis setup is presented in Section 3, while Section 4 shows the difficulties that have occurred during the preliminary experimental tests. Then, the 300 first industrial measurement results are presented together with the result uncertainties and a comparison between a measurement result and the same result obtained by using destructive methods demonstrates the accuracy of the method.

2. Context and physical data

Due to the nuclear test ban and the comprehensive test ban treaty (CTBT) signature, the French gaseous diffusion ^{235}U enrichment facility built in the 60's years was stopped in 1996 and is currently dismantled. Its components are deconstructed, decontaminated and/or characterised in order to be sent to the appropriate waste evacuation way (final disposal or interim storage). Some of these components are three kinds of compressors (called UB, UM, UH) having great masses and big volumes (Fig. 2). About one thousand compressors need to be measured.

The compressor design prevents gamma-ray spectrometry from accurate results: due to their 3 cm thick, surrounding aluminium layer, and to their aluminium internal content, the attenuation coefficient is difficult to determine and the low measured signal can not be easily analysed.

The uranium content is made of ^{234}U, ^{235}U and ^{238}U. The chemical form is assumed to be UO_2F_2 resulting from the UF_6 oxidation. The total neutron emission (TNE) from such chemical molecules is

* Corresponding author. Address: CEA Cadarache, DTN/SMTM/LMN Bât. 224, F-13108 Saint Paul lez Durance, France. Tel.: +33 442253011; fax: +33 442252367.
E-mail address: fanny.jallu@cea.fr (F. Jallu).

0168-583X/$ - see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.nimb.2011.09.018

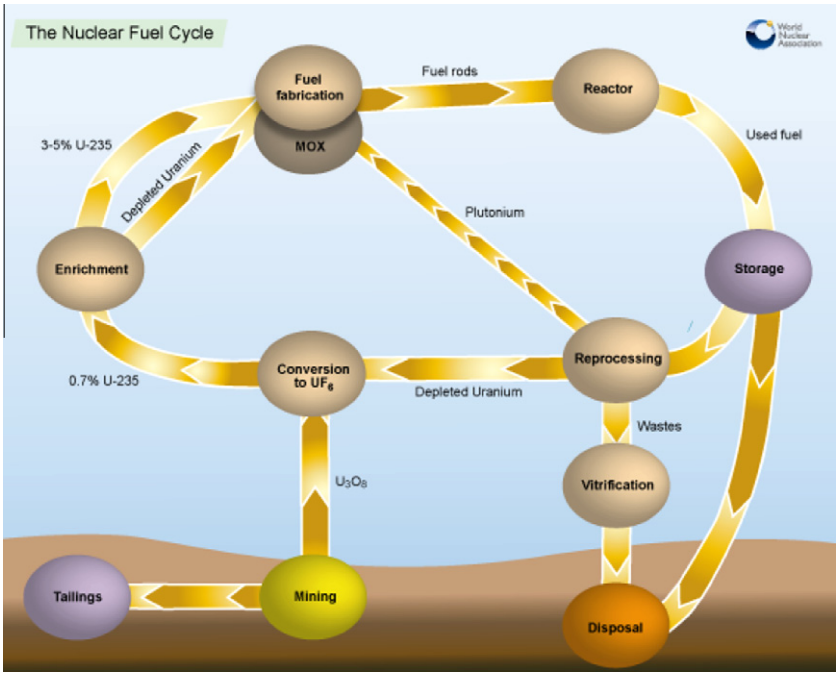

Fig. 1. The nuclear fuel cycle.

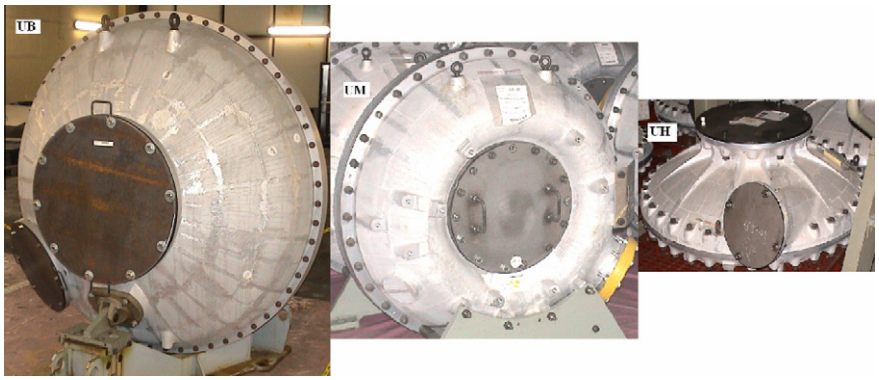

Fig. 2. Photographies of the three types of compressors to be controlled (UB, UM, UH).

due to spontaneous fission (SF) and $^{17}O(\alpha,n)$, $^{18}O(\alpha,n)$ and $^{19}F(\alpha,n)$ reactions. The main α emitter is ^{234}U. Fig. 3 show the variation of the $(\alpha,n)/[(\alpha,n)+SF]$ ratio and the TNE(^{234}U)/[TNE(^{234}U)+TNE(^{235}U) +TNE(^{238}U)] ratio as a function of the ^{235}U enrichment. Data are calculated using the SOURCES4C calculation code [6].

In case of an enrichment greater than 1% ^{235}U, Fig. 3a shows that most of the neutron emission is due to (α,n) reactions, and Fig. 3b shows that the main α emitter is ^{234}U. A passive neutron signal would then be mainly due to the $^{234}U(\alpha,n)$ nuclear process. Furthermore, the gaseous diffusion enrichment industrial process leads the ^{234}U isotopic abundance to increase or decrease as the ^{235}U enrichment would do, approximately.

As a consequence, Fig. 4 presents the variation of the neutron emission linking the ^{235}U mass to the measured signal according to (Eq. (1)):

$$M(^{235}U)(g) = \frac{\text{net signal}\,(s^{-1})}{\text{neutron emission}\,(s^{-1}.g^{-1}(^{235}U)) \times \text{detection efficiency}} \quad (1)$$

For example, neutron emission = 4.76, 3.79 and 3.49 $n^{-1}\,s^{-1}\,g^{-1}$ (^{235}U) at an enrichment of 1%(^{235}U), 3%(^{235}U) and 8%(^{235}U), respectively.

The greater variation appears to be in the [0.2% ^{235}U–1% ^{235}U] UB range, while the neutron emission less varies in the [1% ^{235}U–4% ^{235}U] UM and [4% ^{235}U–12% ^{235}U] UH ranges. In the signal analyse process of each compressor type, a "mean" neutron emission coefficient is considered, while the difference between the neutron emission at the up and down limits of each range is taken into account in the uncertainty evaluations.

The determination of calibration coefficients is needed to analyse the measurement signal. Due to the lack of experimental UO_2F_2 neutron sources, the calibration process is numerical, and is made using the SOURCES4C code and the MCNP4C2 Monte Carlo code [7].

3. Design basis

The measurement setup has been designed using the calculation codes mentioned above, and considering the two following criteria: 1. a Detection Limit Mass (DLM) of less than a few grams of ^{235}U; 2. a Detection Limit Activity (DLA) of less than 100 Bq per gram of raw waste.

The general form is a $L \times l \times H = 1800\,mm \times 2100\,mm \times 2200\,mm$ (internal dimensions) parallelepiped composed of