IAEA-TECDOC-1051

Management of radioactive waste from ^{99}Mo production

INTERNATIONAL ATOMIC ENERGY AGENCY IAEA

November 1998

The IAEA does not normally maintain stocks of reports in this series. However, microfiche copies of these reports can be obtained from

INIS Clearinghouse
International Atomic Energy Agency
Wagramerstrasse 5
P.O. Box 100
A-1400 Vienna, Austria

Orders should be accompanied by prepayment of Austrian Schillings 100,– in the form of a cheque or in the form of IAEA microfiche service coupons which may be ordered separately from the INIS Clearinghouse.

The originating Section of this publication in the IAEA was:

Waste Technology Section
International Atomic Energy Agency
Wagramer Strasse 5
P.O. Box 100
A-1400 Vienna, Austria

MANAGEMENT OF RADIOACTIVE WASTE FROM ^{99}Mo PRODUCTION
IAEA, VIENNA, 1998
IAEA-TECDOC-1051
ISSN 1011-4289

Printed by the IAEA in Austria
November 1998

FOREWORD

^{99m}Tc used for labelling different pharmaceuticals is the most important radionuclide in nuclear medicine practice, and probably will continue to play this important role for the foreseeable future. The prominent position of this radionuclide on the market has been due to its near ideal nuclear properties, the ready availability in the form of convenient ^{99}Mo/^{99m}Tc generator systems and the rapid progress made in recent years in the development of a variety of ^{99m}Tc radiopharmaceuticals for application in oncology, cardiology and other fields. ^{99m}Tc is the short lived daughter product of the parent ^{99}Mo, which is mainly produced by the nuclear fission of ^{235}U. Small amounts of ^{99}Mo are produced also by the neutron activation method. However, current applications of ^{99}Mo/^{99m}Tc generators almost completely depend on the fission production of ^{99}Mo.

Management of radioactive waste is an integral and very important part of ^{99}Mo production, especially for fission ^{99}Mo, and should be given high priority during production process development and operation. To ensure a continued reliable and cost-effective supply of ^{99}Mo, the management of associated waste must be carried out in accordance with internationally accepted safety criteria and related national regulations.

Recognizing the importance of the waste management issue associated with ^{99}Mo production the IAEA initiated preparation of this report to provide Member States and existing and potential producers of ^{99}Mo with practical approaches and the available information on the subject. Because of the commercial character of ^{99}Mo production in some countries the information on the processes and on management of related waste might be not comprehensive. In this context the objective of the report was to emphasize the main scale of the problem and to facilitate access to the accumulated experience, mainly for small and potential producers of ^{99}Mo.

Preparation of this report was accomplished through two consultants meetings held in Vienna in December 1995 and in September 1996. The IAEA would like to express its thanks to all those who took part in the preparation of the report. The IAEA officer responsible for this publication was V. Efremenkov of the Division of Nuclear Fuel Cycle and Waste Technology.

CONTENTS

1. INTRODUCTION

Molybdenum-99 (^{99}Mo) and its daughter product ^{99m}Tc are the most important radionuclides used in nuclear medicine practice. The importance of these radionuclides stems from the fact that ^{99m}Tc is used in about 80% of all diagnostic nuclear medicine procedures. ^{99}Mo production for ^{99m}Tc generators gives rise to a number of waste streams, some of which need special care in handling. Radioactive waste from the production of ^{99}Mo must be managed in such a way that the protection of people and the environment is ensured, both now and in the future. The production and use of ^{99}Mo and ^{99m}Tc have been discussed in detail at various meetings and in scientific journals over the last decade. The IAEA has contributed to this discussion in the form of two technical documents on Fission Molybdenum for Medical Use [1] and Alternative Technologies for ^{99m}Tc Generators [2].

^{99}Mo is produced mainly by nuclear fission of ^{235}U. Processing of irradiated uranium targets for ^{99}Mo production generates considerable amounts of radioactive wastes containing fission products as well as small quantities of transuranium elements. Small amounts of ^{99}Mo are also produced by neutron activation of ^{98}Mo targets. However, this method is not yet fully developed for medium or large scale production. In recent years, conceptual studies on the use of accelerators and aqueous homogeneous nuclear reactors to produce ^{99}Mo have been undertaken as well.

Radiochemical processing of short-cooled irradiated uranium targets offers the opportunity to produce, apart from ^{99}Mo, some other radionuclides for medical use, e.g. ^{133}Xe and ^{131}I. Non-fissioned uranium should also preferably be recovered for re-use. Each of these processes has an influence on waste management practices associated with ^{99}Mo production.

The objective of this technical document is to provide a comprehensive overview of the radioactive waste implications of ^{99}Mo production. This should be of use to the following organizations in different Member States:

- Present large-scale producers of ^{99}Mo having either waste management processes in place or those who are still accumulating waste for future processing, e.g. by supplying information on current waste management practices or possibilities for the optimization of such processes.
- Centralized waste management organizations having to treat ^{99}Mo waste streams.
- Smaller producers of ^{99}Mo, or those who are developing production processes, in order to assist them in evaluating the waste implications of the various routes and methods for ^{99}Mo production.
- Countries contemplating production, even on a very small scale, for their own use to ensure that ^{99}Mo production is not viewed in isolation without giving early recognition to the necessity for considering waste management aspects.

Waste management in the context of this report encompasses all waste-related aspects, for example, handling, treatment, conditioning, storage, transport, and disposal. Relevant information on ^{99}Mo production processes (present, as well as envisaged for the future) is included in order to supply the background against which waste management practices should be considered. At present most ^{99}Mo is produced by means of the fission of ^{235}U, as this is still the only proven process for the production of large quantities of ^{99}Mo with very high specific activity. For this reason most of the available information pertains to waste from fission ^{99}Mo production. This waste is also the most difficult to handle due to the presence of fission products. Although process flowsheets vary considerably, there are certain similarities in the resultant waste, for example, acid dissolution of irradiated targets produces waste containing uranium and most of the fission products, while dissolution in an alkaline medium leaves uranium undissolved and only some fission products in the waste solution. Information on waste from other ^{99}Mo production methods is supplied as far as it is available.

Apart from the difficulties of handling and the wide implications of the waste generated during such a process, as described in this report, a number of other aspects should also be considered. Examples of these are the following:

- the availability of enriched uranium;
- the concomitant nuclear materials proliferation and safeguards concerns;
- access to a high flux reactor;
- the availability of target manufacturing technology;
- the existence of a ^{99}Mo extraction and purification process;
- the existence of shielded hot-cell facilities;
- the existence of trained manpower;
- the availability of shipping containers;
- the existence of licensing, quality assurance and good manufacturing practices, and
- the financial and market implications.

Following a short description of the basic nuclear and physical properties of ^{99}Mo and ^{99m}Tc in Section 2 of this report, an overview of past, present and possible future production methods is supplied in Section 3. In the next chapter the characteristics of the various waste streams produced by the aforementioned processes are given to enable the reader to appreciate the complexities of waste issues related to the various ^{99}Mo production routes. Section 5 provides a description of the necessary waste management practices needed to handle the relevant waste streams in a responsible and internationally-accepted manner. In the last section conclusions and recommendations are given.

2. BASIC PROPERTIES OF MOLYBDENUM-99 AND TECHNETIUM-99m

As stated in the introduction, molybdenum-99 (^{99}Mo) is probably the most important man-made radionuclide because of the wide use of its daughter nuclide, technetium-99m (^{99m}Tc), in nuclear medicine. A brief summary of typical nuclear and physical properties of ^{99}Mo and ^{99m}Tc radionuclides is given in the following sections.

2.1. NUCLEAR PROPERTIES

Molybdenum-99 decays via β^- mode to ^{99m}Tc (87.5%) and ^{99}Tc (12.5%). The half-life of ^{99}Mo is 66.02 h [3, 4]. ^{99m}Tc decays by isomeric transition (IT) according to the decay scheme shown in Fig. 1. The principal gamma-emission associated with the transition to the ground state of ^{99}Tc is γ_2, 0.1404 MeV photons which occurs at a frequency of 0.89 per disintegration. Apart from a very low frequency, direct beta decay to ruthenium-99, ^{99}Ru (Fig. 2) particulate radiation associated with the decay of ^{99m}Tc is limited to internal conversion and auger electrons.

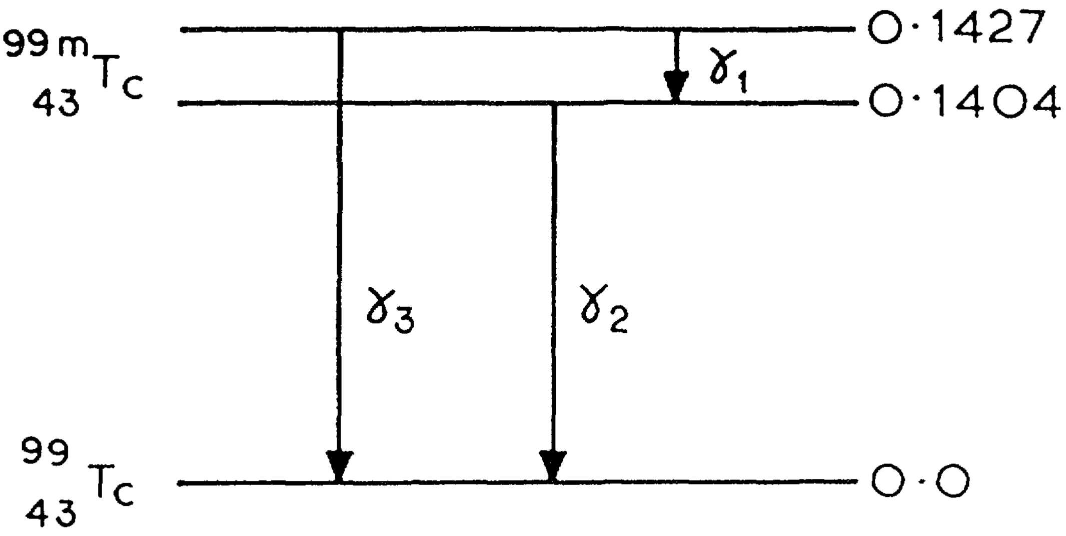


FIG. 1. Isomeric level decay of ^{99m}Tc to ^{99}Ru.

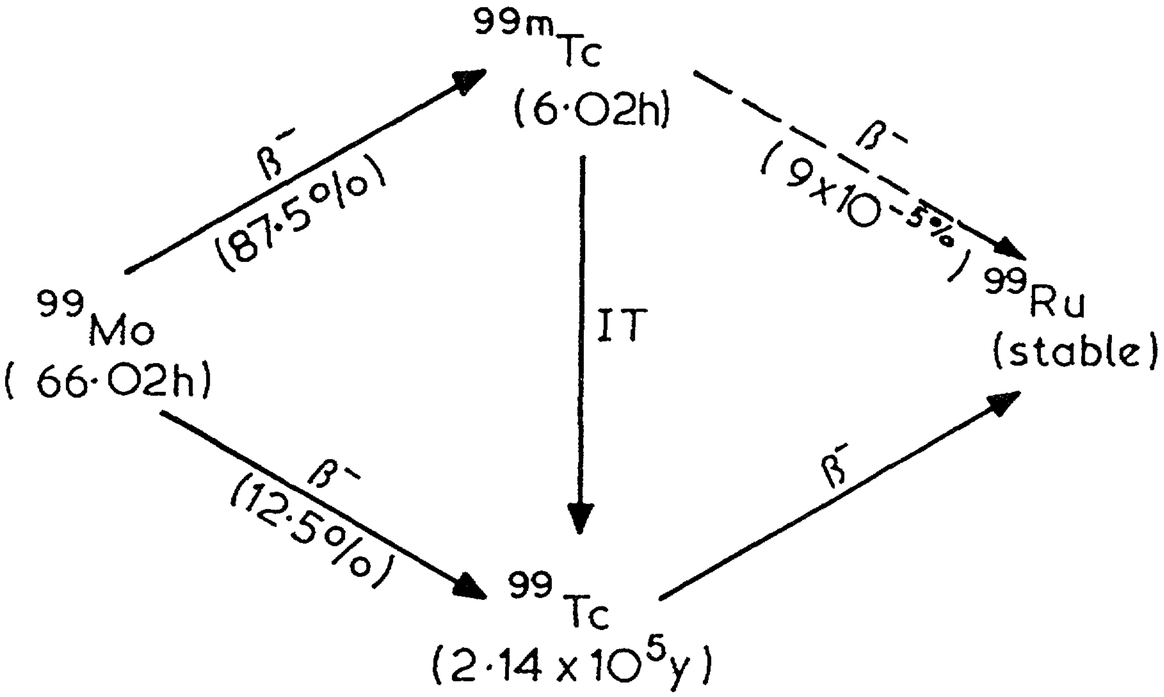


FIG. 2. Decay sequences of ^{99}Mo to ^{99}Ru.

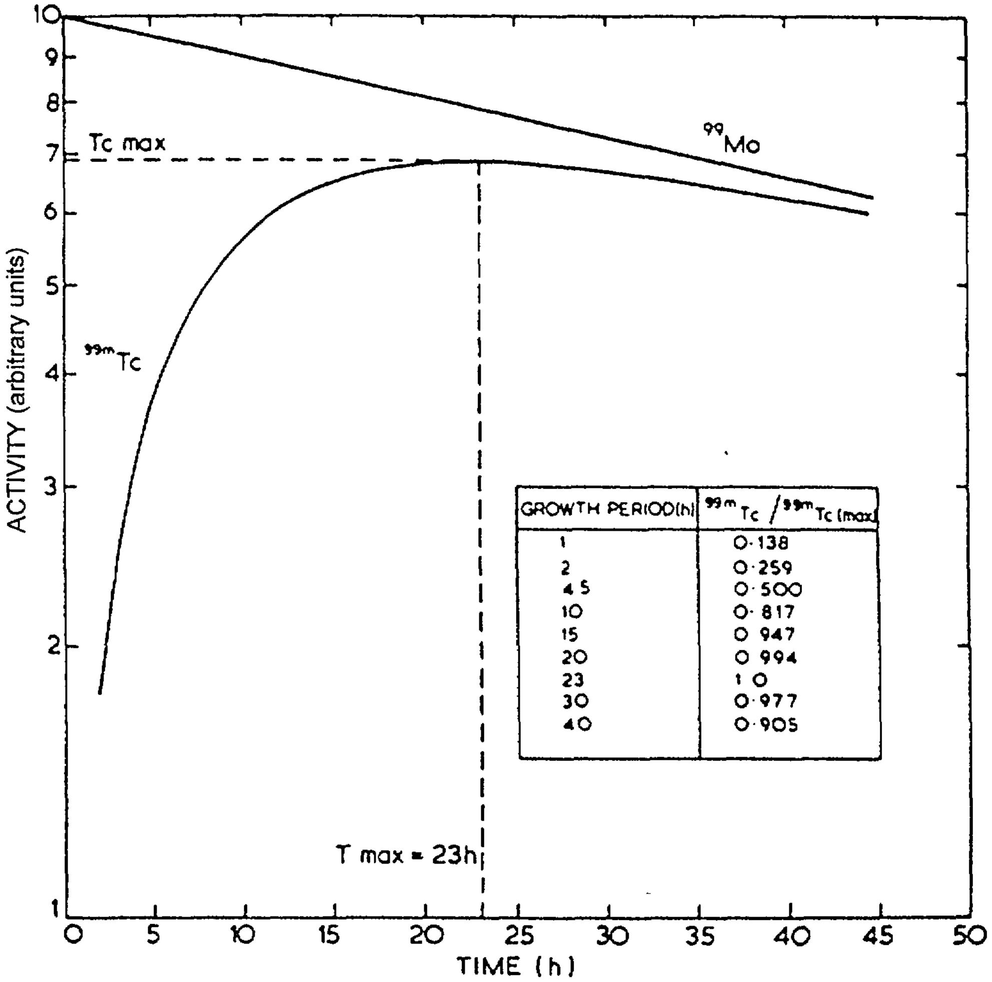
2.2. DECAY SCHEME OF ^{99}Mo

Figure 2 shows the decay scheme for the ^{99}Mo:^{99m}Tc:^{99}Tc:^{99}Ru system with respective half-lives and branching factors. The decay growth curve for the ^{99}Mo:^{99m}Tc system is shown in Fig. 3 [3]. The radioactivity attributable to ^{99}Tc is insignificant because of its very long half-life at $2.14 \cdot 10^5$ years; for example, 1 curie of ^{99}Mo will produce only $4 \cdot 10^{-8}$ curies of ^{99}Tc.

2.3. EXTERNAL IRRADIATION

The maximum range of ^{99}Mo beta particles is 540 mg/cm^2. The gamma exposure rate at a distance of 1 cm from a 37 MBq point source (K factor) is 1.78 μGy/s (or 3.42 μGy/s including ^{99m}Tc in equilibrium). The tenth-thickness in lead for the 0.74 and 0.78 MeV gamma radiation is 3.0 cm; owing to their low percentage (the sum of both is 16.8%), a lead wall 5 cm thick represents a practical shield for the handling of ^{99}Mo activities up to around 37 GBq. However, much thicker shielding is necessary when dissolution of the irradiated targets and separation of ^{99}Mo from fission products are being carried out due to high energy radiation from the fission products [4].

The gamma exposure rate at a distance of 1 cm from a 37 MBq point source (K factor) of ^{99m}Tc is 2.0 μGy/s. The tenth-thickness in lead for 0.14 MeV gamma radiation of ^{99m}Tc is 0.45 cm; 2 cm of lead are sufficient for shielding of activities up to 370 GBq [4].



GROWTH PERIOD(h)	^{99m}Tc / ^{99m}Tc (max)
1	0·138
2	0·259
4 5	0·500
10	0·817
15	0 947
20	0 994
23	1 0
30	0·977
40	0·905

FIG. 3. Decay-growth ^{99}Mo: ^{99m}Tc system.

2.4. INTERNAL CONTAMINATION

Both ^{99}Mo and ^{99}Tc are classified as isotopes with medium radiotoxicity (lower sub-group B), while ^{99m}Tc is classified as an isotope with low radiotoxicity [4].

The effective half-life of ^{99}Mo referred to the total body is 1.8 d, the liver being the critical organ. The effective half-lives of ^{99m}Tc and ^{99}Tc are 0.2 d and 1 d respectively, for the total body [4].

2.5. EXTERNAL CONTAMINATION AND DECONTAMINATION METHOD

Contamination of equipment and containers can occur during purification and handling of ^{99}Mo solution and ^{99m}Tc generators. This type of contamination is easy to decontaminate.

Molybdenum-99 can be satisfactorily removed from surfaces with hot water, in the absence of calcium with dilute alkalies, in the presence of calcium with diluted acids. The presence of ammonium molybdate as the carrier increases the efficiency of decontamination.

^{99}Tc and ^{99m}Tc, which are mainly present as pertechnetate ions (TcO_4^-), are easily decontaminated with any aqueous solution of diluted acids.

2.6. PURITY SPECIFICATION OF ^{99}Mo PRODUCT

The medical end use of ^{99m}Tc requires stringent purity specifications for its parent nuclide ^{99}Mo. Every ^{99}Mo producer has their own purity specifications; however, they are quite similar. A typical purity specification of ^{99}Mo is summarized below [1].

Impurity:

^{131}I/^{99}Mo	$\leq$5E-05
^{103}Ru/^{99}Mo	$\leq$5E-05
^{89}Sr/^{99}Mo	$\leq$6E-07
^{90}Sr/^{99}Mo	$\leq$6E-08
Total $\beta\gamma$/^{99}Mo	$\leq$1E-04
Total α/^{99}Mo	$\leq$1E-09

3. PRODUCTION METHODS OF MOLYBDENUM-99

3.1. INTRODUCTION

The possibility of developing a ^{99m}Tc chromatographic generator based on the decay of ^{99}Mo was recognized and then experimentally proven by Brookhaven National Laboratory (BNL) in the late 1950s [5–7]. It took physicians about five to seven years after this event to appreciate the importance of this scientific success when it became obvious that ^{99m}Tc possessed ideal physical properties for many of the diagnostic procedures being used in nuclear medicine [8–10].

In 1966, Brookhaven National Laboratory withdrew from the production and distribution of ^{99m}Tc generators in favor of commercial distributors. The earliest commercial generators, based on the neutron irradiation of stable ^{98}Mo for the production of ^{99}Mo, were developed and distributed by several companies located in the USA. There was a rapid growth of generator production based on (n, γ) ^{99}Mo through the sixties with the next major change being the shift to fission-product ^{99}Mo generators in the early seventies.

The technical specifications of ^{99m}Tc generators are very closely linked to the quality of ^{99}Mo. The use of fission ^{99}Mo improved the quality of ^{99m}Tc generators considerably over production with (n,γ) ^{99}Mo [11]. However, in comparison with the (n, γ) activation of ^{98}Mo, the production process of fission ^{99}Mo is quite sophisticated and many aspects have to be taken into consideration, for example the targeting, the irradiation conditions, the extremely high exposure rates, off-gas filtration and trapping, product quality, target material recycling, waste management (including treatment, conditioning, storage and final disposal), and safeguard regulations.

The biggest producer of fission ^{99}Mo in the world is Nordion International, Canada. A typical batch of ^{99}Mo produced by Nordion measures between 5000–6000 Ci[1] (at the end of production) and Nordion processes several batches per week [12]. The second largest producer of fission ^{99}Mo is the Institut National des Radioéléments (IRE), Fleurus, Belgium with a capacity for production of ^{99}Mo of roughly 10 000 Ci/week. Both organizations have been commercially marketing ^{99}Mo for a long time. For example, Nordion started with fission ^{99}Mo production in the early 1970s, while the IRE started in 1979 with medium size batches and upgraded production at the beginning of the 1980s [1, 12].

Recently two additional plants were constructed for production of ^{99}Mo on a large scale, namely, the Atomic Energy Corporation of South Africa with a ^{99}Mo production capacity of about 8000 Ci/week (at the end of the production) [13], and Mallinckrodt in the Netherlands with a capacity of about 10 000 Ci/week.

There are also several medium scale producers of fission ^{99}Mo, for example, the Institute of Physics and Power Engineering, Russian Federation, where about 300 Ci/week (at the end of the production) can be generated. Routine production of ^{99}Mo began in Australia in 1967; the current production by the Australian Nuclear Science and Technology Organization (ANSTO) is 300 Ci/week. In Argentina about 100–150 Ci of ^{99}Mo can be produced weekly.

In all the above-mentioned cases, where both highly enriched uranium (HEU) or low enriched uranium (LEU) targets are used, the quality of fission ^{99}Mo is very high and fulfils international requirements.

Many countries have tried to improve (n, γ) activation production methods. Over the last two decades, sublimation, extraction and gel type ^{99m}Tc generators have been developed, for example in Australia, China, India, Hungary, and some other countries [14].

Japan Atomic Research Institute has experience in (n, γ) and (n, f) methods of ^{99}Mo production. China, India and Hungary developed their own capabilities in the production of activation (n, γ) ^{99}Mo for use in several types of ^{99m}Tc generators [2].

---

[1] 1 Ci = 37 GBq.

Several countries have worked at developing ^{99}Mo production technology, but have discontinued the research work for different reasons. This happened in the USA, Japan, the former German Democratic Republic, the former Federal Republic of Germany, and France [11, 16]. The main reasons for this were:

- the increase in the safety risk using old reactors for ^{99}Mo production (for example: General Electric Testing Reactor (USA) in 1977; Amor-I and Amor-II plants (former German Democratic Republic) at the end of the 1980s).
- uneconomic technology, no longer able to be competitive where there was a sudden drop in the price of ^{99}Mo on the world market (Saclay Nuclear Research Center, France, 1989);
- unavailability of a regular supply of enriched uranium and unsatisfactory progress in technology development (Japan Atomic Energy Research Institute, 1978);
- incompatibility of commercial production and scientific research (Nuclear Research Center of Karlsruhe, Federal Republic of Germany, end of the 1980s).

The ^{99}Mo production capacity ranged from 20 Ci/week to 2000 Ci/week (end of production) at these discontinued facilities.

Recently, alternative possibilities for the production of high quality ^{99}Mo have been suggested. These include an aqueous homogeneous reactor as well as the use of cyclotrons and accelerators [17, 18]. However, these new technologies are in the early experimental stages. It will take some time before life-cycle costs can be established to determine how competitive the newer technologies will be. A key factor in the life-cycle costs is the handling, treatment and disposal of associated wastes.

Methods utilized for ^{99}Mo production are summarized in Table I. More details on these methods are discussed in the following sections.

3.2. FISSION OF HIGHLY ENRICHED URANIUM

3.2.1. ^{99}Mo production at Chalk River Laboratories (CRL), Canada

The ^{99}Mo target material consists of 93% ^{235}U enriched uranium metal alloyed with aluminum. The cylindrical target is manufactured with external fins to enhance heat transfer. The targets are assembled into fuel strings and irradiated at a thermal neutron flux of $(2\text{-}3)\cdot 10^{14}$ $n\cdot cm^{-2}\cdot s^{-1}$ for a scheduled period [1]. Targets are currently irradiated in National Research Universal (NRU) reactor. The first of two MAPLE reactors are presently being constructed and when both of them are operational they will become the main supply facilities.

The current CRL ^{99}Mo recovery process is shown in Fig. 4. After a short cooling period, the irradiated target is stripped in a hot cell to remove external cladding, then dissolved in nitric acid. Xenon-133 (^{133}Xe) in the off-gas stream from the dissolver vessel is occasionally trapped (depending on the demand of ^{133}Xe). The ^{133}Xe is shipped to the off-site customers.

The solution containing uranium and fission products is drawn through an alumina (Al_2O_3) column where molybdenum is retained. The column is washed sequentially to remove the other fission products and uranium. The molybdenum is then eluted from the column using ammonium hydroxide.

TABLE I. SUMMARY OF METHODS OF ^{99}Mo PRODUCTION

Method	**Advantages**	**Disadvantages**
1. Nuclear fission of ^{235}U using HEU (≥20% ^{235}U).	1. High specific activity ^{99}Mo; 2. Suitable for the preparation of chromatographic ^{99m}Tc; 3. Well proven process.	1. High investment required; 2. Sophisticated technology for purification of ^{99}Mo required; 3. Formation of relatively high quantity of radioactive waste; 4. Need for the availability of HEU; safeguards.
2. Nuclear fission of ^{235}U using LEU (<20% ^{235}U).	1+2 as above. 3. Proven process in some cases.	1,2+3. As above; 4. No proven target plate technology for uranium silicide.
3. Thermal neutron irradiation of ^{98}Mo. Material for production of ^{99}Mo can be either: • highly enriched stable isotope ^{98}Mo (>95%); or • material with natural content of ^{98}Mo (24.4%).	1. Low quantity of contaminant nuclides; 2. Less sophisticated design of target; 3. Simple technology for purification of ^{99}Mo; 4. Suitable for small-scale production.	1. Low specific activity ^{99}Mo (with carrier); 2. Not ideal for preparation of chromatographic generators.
4. Acceleration of charged particles (protons). Material for production of ^{99}Mo can be either: • highly enriched isotope ^{100}Mo (>98%) or • material with natural content of ^{100}Mo (9.6%).	1. Low quantity of contaminant nuclides; 2. Short period of irradiation; 3. Simple technology for purification of ^{99}Mo.	1. Medium specific activity of ^{99}Mo (with carrier); 2. Necessity of use cyclotrons or accelerators with the energy of bombarding particles >40 MeV; 3. No technology presently available.

The waste solution and the column wash solutions are transferred to temporary storage for later treatment and disposal.

3.2.2. ^{99}Mo production at the Institut National des Radioéléments (IRE), Fleurus, Belgium

The ^{99}Mo target consists of 30 g aluminum and 4.2 g of 89% to 93% enriched uranium. The dimensions of the target are: external diameter, 22 mm; internal diameter, 18.46 mm; wall thickness, 1.27 mm and length, 160 mm [19].

In general, the ^{99}Mo targets are exposed to a flux of $1.5 \cdot 10^{14}$ $n \cdot cm^{-2} \cdot s^{-1}$ for 120 h. Currently, the irradiated targets are supplied from the BR2 (Belgium Research Center at Mol), HFR, SILOE and OSIRIS reactors [20]. The irradiated targets are cooled in the reactor pool for a few hours before being placed in containers and shipped to IRE, Fleurus.

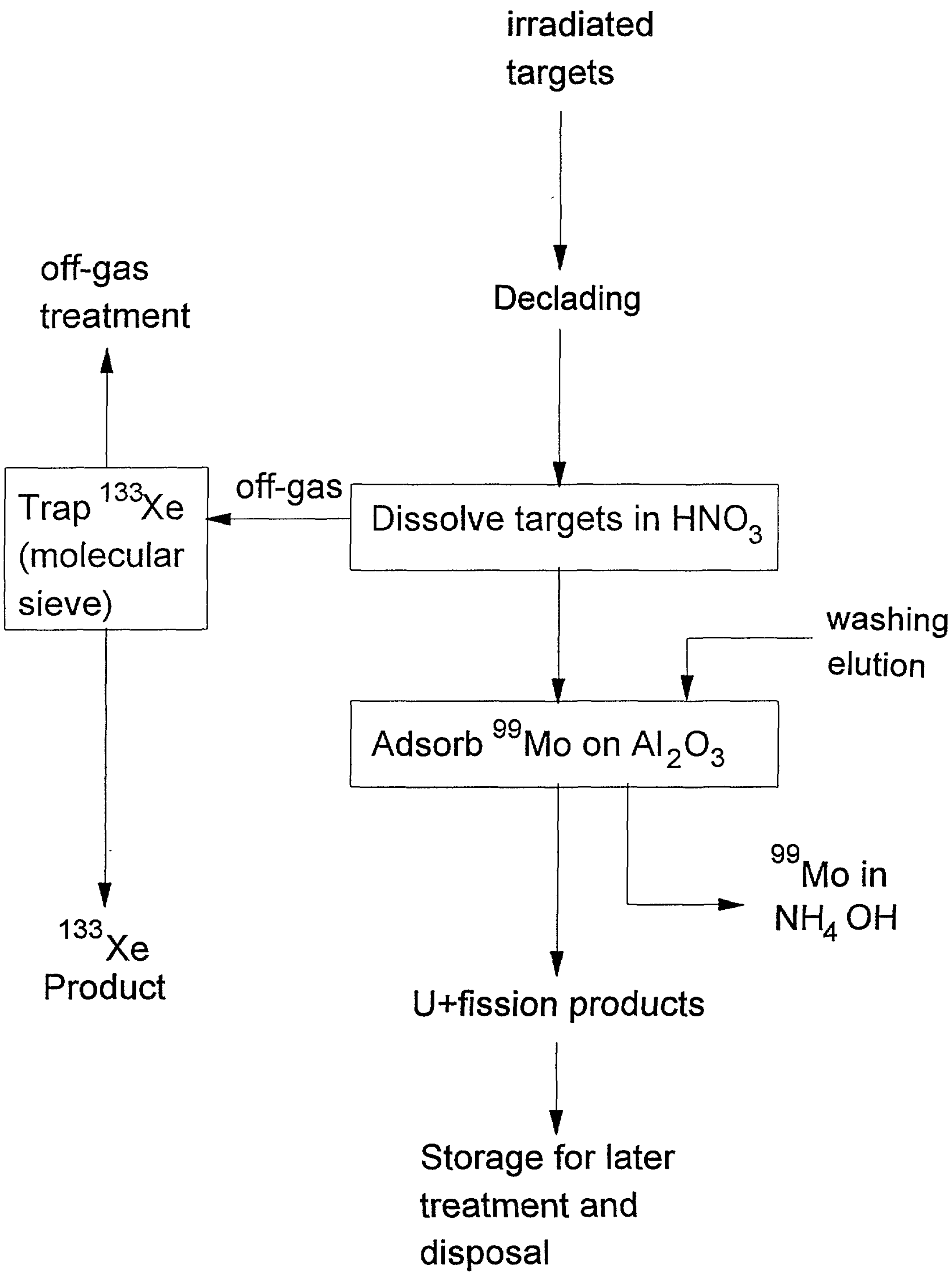


FIG. 4. Flow diagram for ^{99}Mo recovery process at CRL, Canada.

At Fleurus a process is installed for triple production of ^{99}Mo, ^{131}I and ^{133}Xe from the irradiated targets [20]. A simplified flow diagram of the isotope production facility is shown in Fig. 5. The irradiated targets are dissolved in 3M NaOH and 4M $NaNO_3$. Radioxenon is released from the irradiated targets during the dissolution and trapped on copper clippings, then purified prior to deliver to the customers. The uranium and most of the fission products are precipitated as hydroxides in the dissolving solution; however, radioiodine and molybdenum are still in ionic forms as I^{-1} and MoO_4^{-2}, respectively. Iodine and molybdenum are separated from the uranium and the other fission products by filtration. The filtrate is then acidified using concentrated nitric acid. The iodine is released in this step. The released iodine is trapped on platinum asbestos and then purified. The acidified solution is passed through an alumina column where molybdenum is retained and thereafter eluted using NH_4OH. The ^{99}Mo solution is purified using ion-exchange resin and activated carbon. The ^{99}Mo yield of this operation is about 85% to 90%. A flow diagram for ^{99}Mo separation and purification is given in Fig. 6.

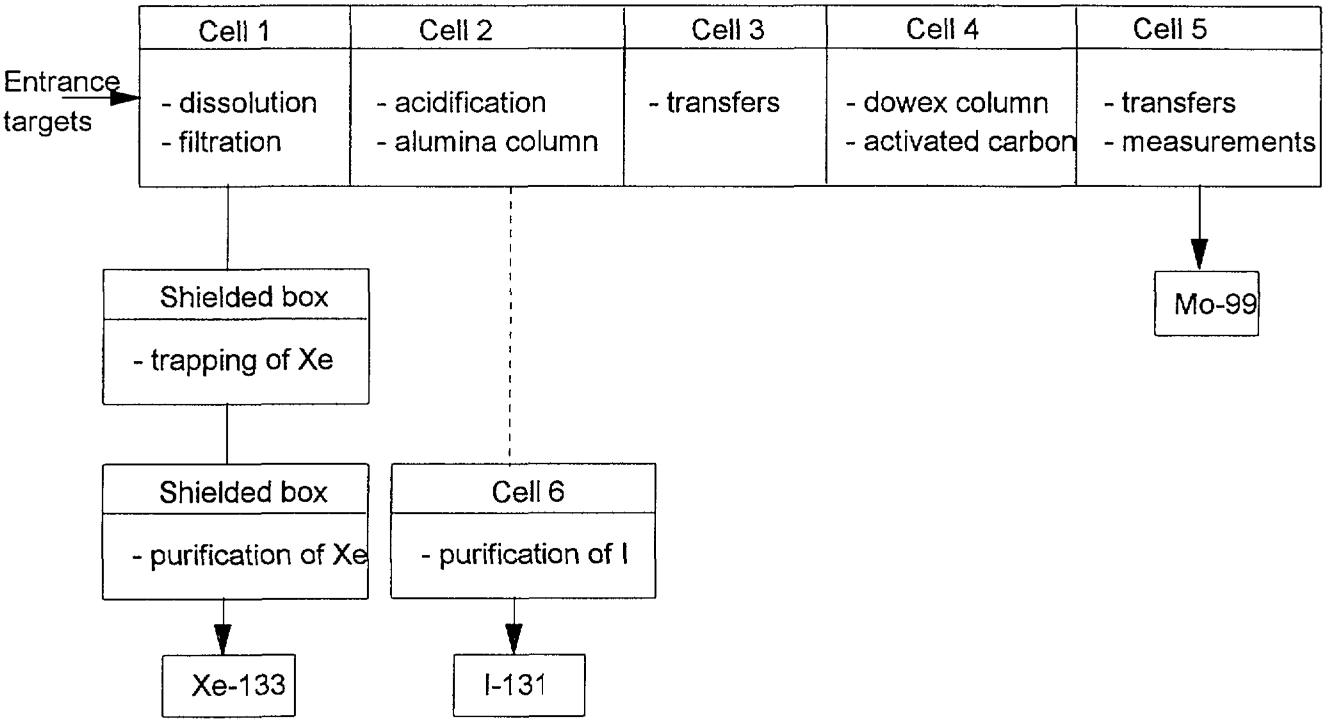


Fig 5. Flow diagram of the IRE isotope production facility, Belgium.

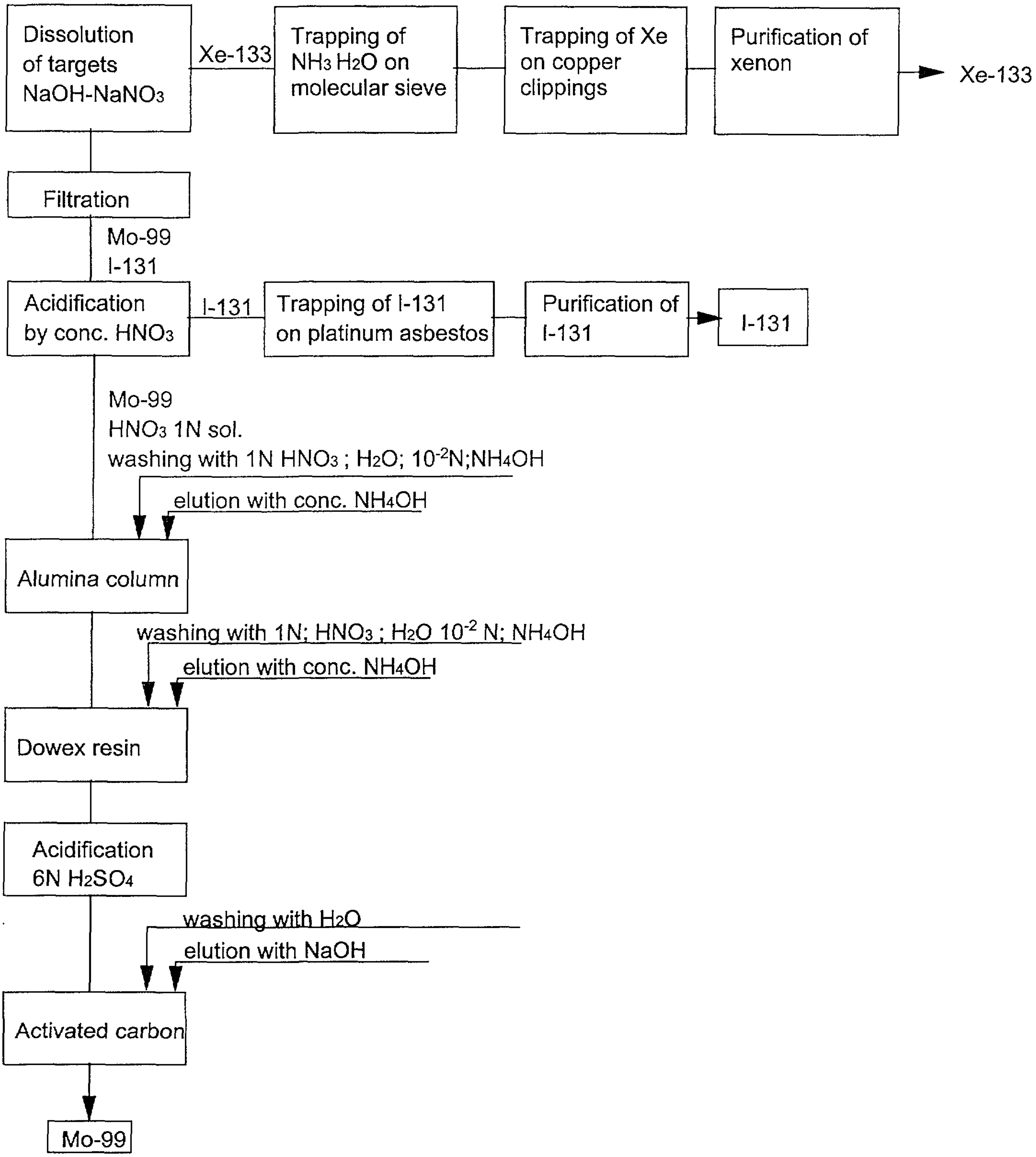


Fig 6. Flow diagram of isotope separation and purification process, Belgium.

3.2.3. ^{99}Mo production at Mallinckrodt/ECN, Petten, Netherlands

A fission ^{99}Mo process cycle which is integrated with the nuclear material recycling and target fabrication was developed and used at KfK, Germany [21]. A new production facility based on this method has been built by Mallinckrodt at the Research Center ECN, Petten, Netherlands, which has started operation recently (Sept. 1996) to produce ^{99}Mo. The target plates, produced in Scotland, consist of an aluminum-cladded uranium/aluminum alloy of the approximate composition UAl_3. To achieve the highest possible fission product yields uranium, enriched to 90–93%, is irradiated with a thermal neutron flux of $1 \cdot 10^{14}\ \mathrm{n \cdot cm^{-2} \cdot s^{-1}}$. Figure 7 shows a simplified scheme of the complete ^{99}Mo production process including the route for recycling of uranium.

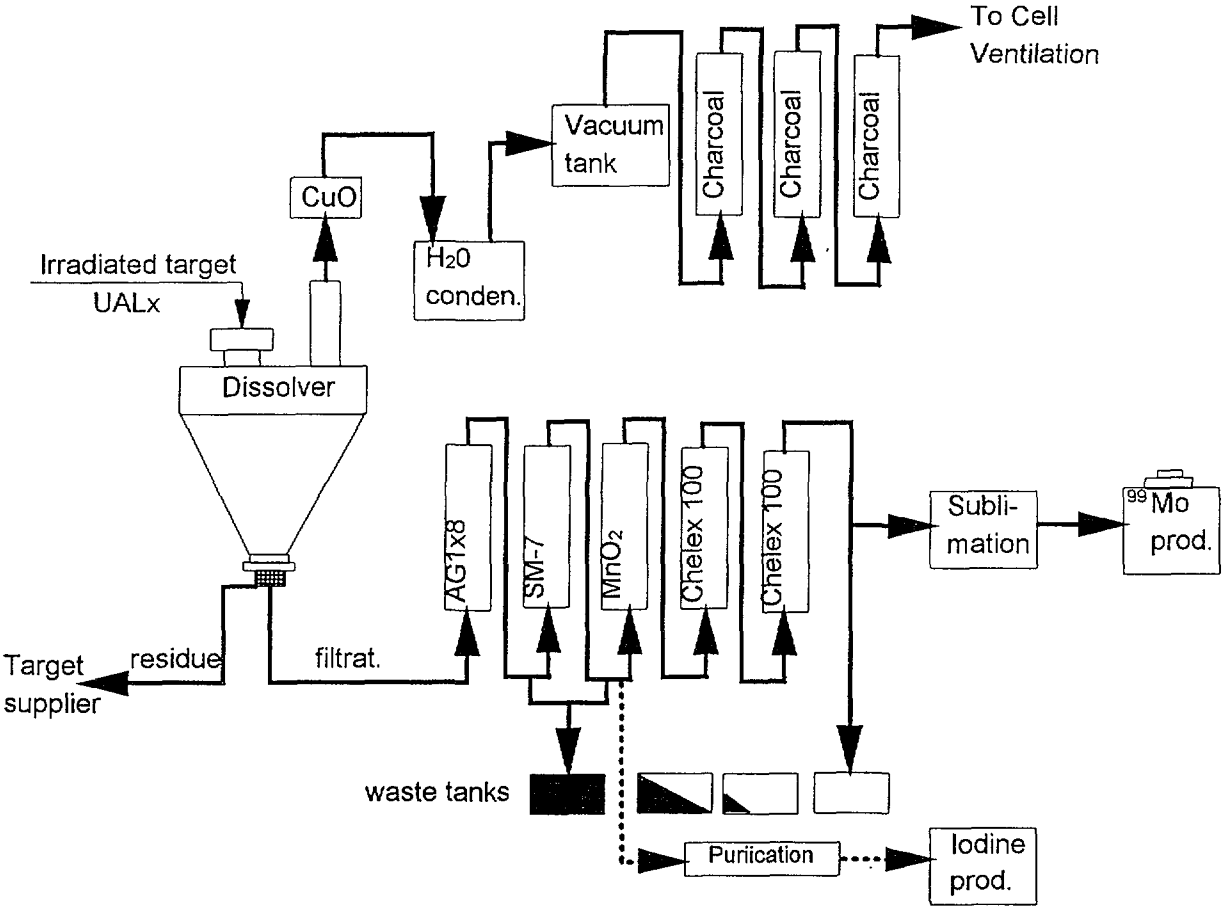


Fig. 7. Flow diagram of ^{99}Mo production facility at Mallinckrodt/ECN in the Netherlands.

The first chemical step consists of the alkaline (NaOH) digestion of the target during which the main constituents of the UAl_3 targets are dissolved, namely, Al and the fission products soluble in the alkaline (such as alkaline and alkaline earth cations as well as antimony, iodine, tellurium, tin, and molybdenum). The alkaline solution containing molybdenum passes through a column filled with the strong-base ion exchanger (AG1x8). Molybdenum is quantitatively adsorbed on the AG1x8 ion exchanger together with a significant proportion of other fission products. Cationic impurities such as cesium, strontium and barium pass through the column together with hydroxyaluminate, telluric and iodine anions and follow the route to waste. Molybdenum and the main part of the accompanying fission products are eluted with sulphuric acid.

The acidified molybdenum solution first enters a column filled with an ion-exchange resin (SM-7) to retain the iodine and then enters the chromatographic column filled with manganese oxide (MnO_2). In this column molybdenum is purified from sodium salts and traces of fission products. The MnO_2 containing ^{99}Mo is then dissolved completely with a mixture containing sulphuric acid.

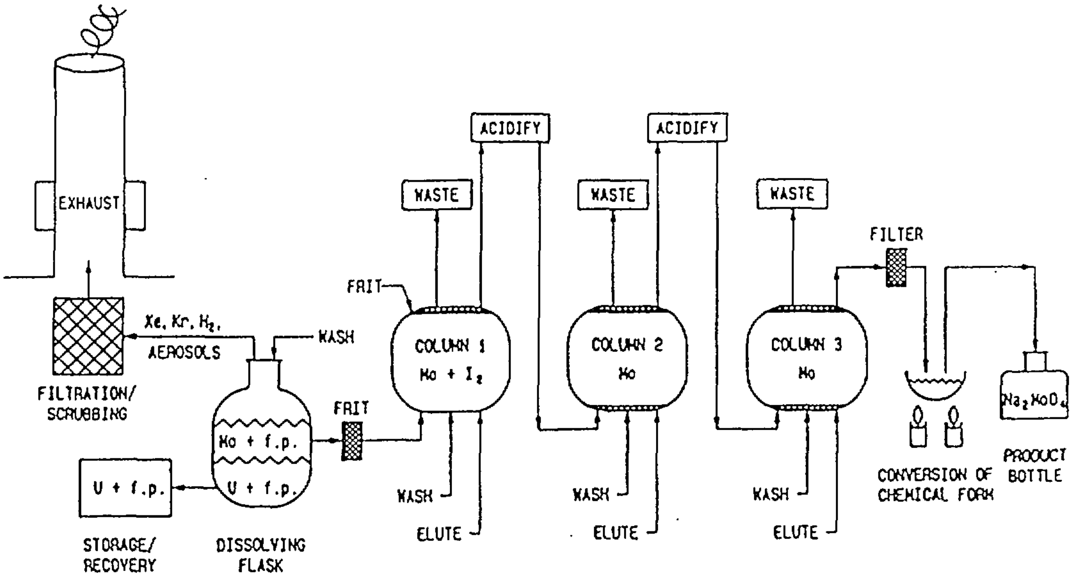
From this solution, the molybdenum is retained quantitatively on two columns containing Chelex-100 resin. The remaining fission products go with the solution to waste. Molybdenum is then eluted with ammonium hydroxide and transferred as ammonium molybdate to the sublimation cell for final purification.

The insoluble residue in the dissolver contains about 99% of the initially irradiated uranium together with the insoluble fission products, including ruthenium, zirconium, niobium, the lanthanides, Am and Pu. This residue is conditioned in the preparation for return to the target producer in Scotland, where it will be further reprocessed.

3.2.4. ^{99}Mo production at the Atomic Energy Corporation (AEC), South Africa

Target plates for the AEC process consist of an uranium–aluminum alloy (containing 46% enriched uranium) cladded with aluminum. These are irradiated in the 20 MW SAFARI-1 reactor at Pelindaba at a neutron flux of $(1–2)\cdot 10^{14}$ $n\cdot cm^{-2}\cdot s^{-1}$ for 50–200 hours, depending on customer demand.

The flowsheet of the process is shown in Fig. 8. The target plates are dissolved in concentrated sodium hydroxide. A suitable oxidizing agent is added to ensure that all molybdenum is dissolved, while at the same time leaving the non-fissioned uranium as a solid residue for recovery at a later stage. Nuclides of only a few elements are dissolved with molybdenum, and three specific purification steps have been designed to ensure proper decontamination of ^{99}Mo with respect to these elements, while at the same time giving good product recovery.



f.p. = fission products

FIG. 8. Process scheme for molybdenum extraction and purification at AEC in South Africa.

Purification of ^{99}Mo is carried out by means of two anion exchange resins and one chelating resin, all commercially available. Each purification step consists of adsorption of Mo, washing to remove any residual source solution, and elution of Mo under conditions suitable for separation from any impurities that might still be present at the particular stage. The ammonium hydroxide eluate of the third column is filtered, evaporated to dryness, and redissolved in 0.2M NaOH to convert the product into sodium molybdate. Sodium hypochlorite is sometimes added at this stage to ensure that Mo is maintained in the molybdate form.

Dissolution of the irradiated target plates, separation and purification of ^{99}Mo, followed by quantification, dispensing and packaging of the final product, take place in five adjoining lead shielded hot cells.

3.2.5. ^{99}Mo production at IPPE, Russian Federation

There are two main ^{99}Mo producers in the Russian Federation: the Federal Research Center – Institute of Physics and Power Engineering (IPPE) and the branch of Moscow Physico-Chemical Research Karpov Institute. The technologies of ^{99}Mo production used by both of the above mentioned institutes are similar. In this section the practice of IPPE is described.

The target material consists of uranium dioxide (UO_2), enriched to 90% ^{235}U, pressed with magnesium oxide (MgO) in the form of hollow cylinder. The target cladding is made from stainless steel. The small gap between the target and cladding is filled with metallic magnesium for better heat transfer. Targets irradiation is currently carried out in research reactors (the thermal neutrons reactor AM-1 with a thermal neutron flux of $3 \cdot 10^{13}$ $n \cdot cm^{-2} \cdot s^{-1}$ and in the fast neutron reactor BR-10 with a fast neutron flux of $1 \cdot 10^{14}$ $n \cdot cm^{-2} \cdot s^{-1}$ into its thermal tube).

After cooling for 10–24 hours the targets are shipped to the isotope production facility. The principal flowsheet of ^{99}Mo recovery process from irradiated uranium targets is shown in Fig. 9. After cutting the targets their content is dissolved in nitric acid. Released ^{133}Xe, the other isotopes of noble gases and isotopes of iodine (for example, ^{131}I, ^{132}I and ^{133}I) are trapped by means of absorption and adsorption techniques and filling of preevacuated tanks. If there are any demands for these isotopes they are additionally purified and delivered to customers. This off-gas is the main radioactive aerosol and gaseous waste stream.

The acidic solution of fission products is transferred into the extractor and the next process steps are carried out. Only ^{99}Mo and a few nuclides are separated into organic phase from most fission products, which are retained in the aqueous phase at the extraction step. After the re-extraction step the main component of aqueous solution is ^{99}Mo. The aqueous phase after the first separation step and the organic phase after reextraction of ^{99}Mo contain nearly all non-fissioned uranium and the rest of the fission products and represent the main radioactive liquid waste streams.

Further purification of crude ^{99}Mo is carried out by means of extraction chromatography. The purification consists of extraction of molybdenum on the surface of special prepared adsorbent, washing the column to remove any residual impurities and then re-extraction of ^{99}Mo. The final aqueous solution is evaporated to dryness and than ^{99}Mo is converted to the needed chemical form by dissolution in acid or alkali. After testing and packaging ^{99}Mo species are ready for delivering to customers.

All technological steps of ^{99}Mo recovery process are carried out under strict control to ensure high and stable quality of the end product. The yield of the ^{99}Mo recovery process is about 90%.

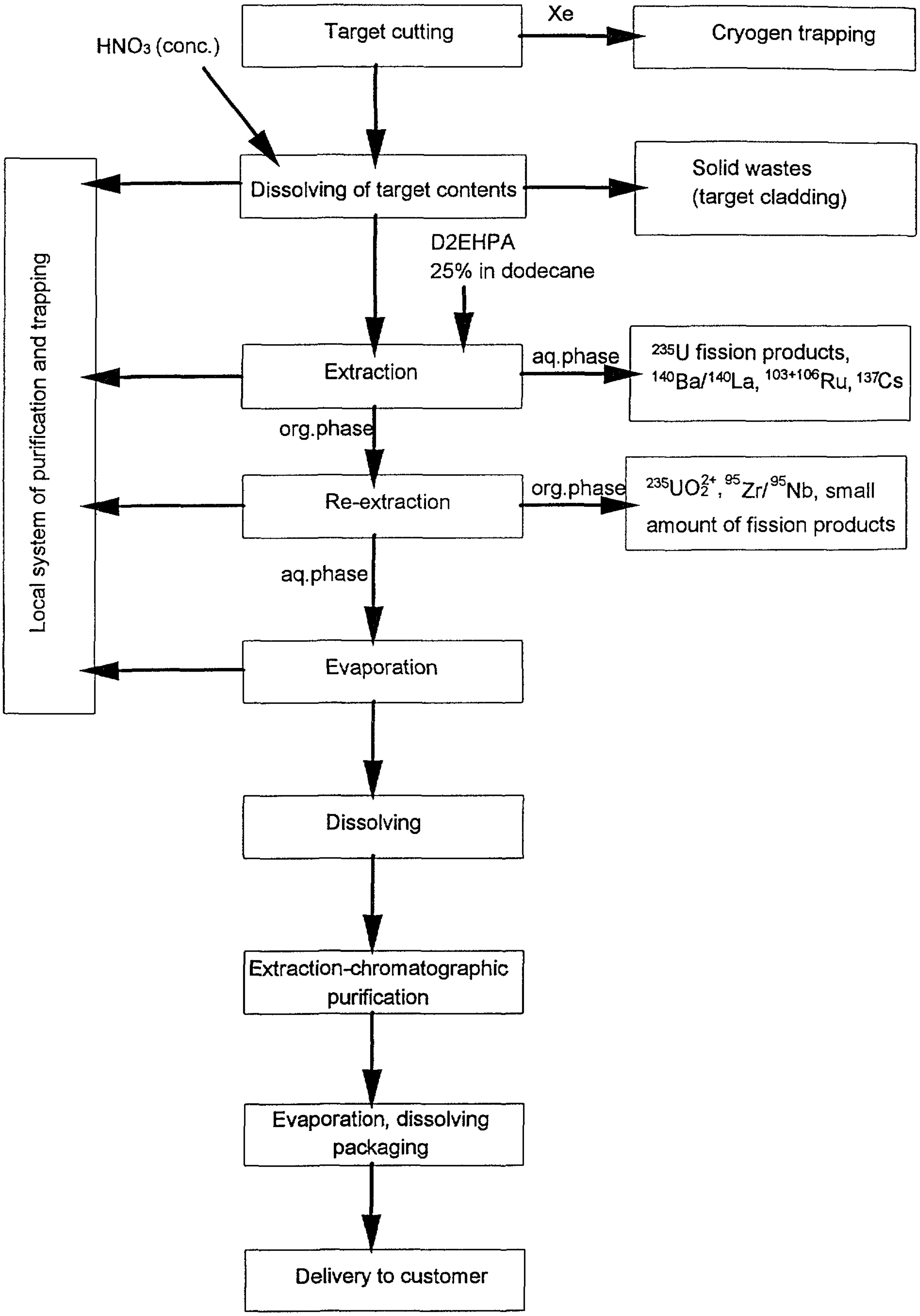


FIG. 9. Principal scheme of Mo-99 production in the Russian Federation.

3.2.6. The aqueous homogeneous nuclear reactor as a prospective ^{99}Mo source

IPPE, in cooperation with several other Russian Institutes, proposed the concept of a specialized aqueous homogeneous nuclear reactor as a prospective convenient ^{99}Mo source. The general idea of this concept is to operate a small aqueous homogeneous reactor equipped with two additional separation and trapping systems. The first system comprises a gaseous recuperation and a trapping unit which is used for safe recombination of formed radiolytical hydrogen and oxygen, and delaying aerosol and volatile nuclides for decaying or further purposes. The second one consists of a specially designed liquid bypass directly connected to the reactor and a removable exchange column for ^{99}Mo selective extracting and retaining while the fuel components of operated reactor are passing through it by means of a special pump. Additional purification of extracted ^{99}Mo will be carried out with existing proven technology. The control rods are inserted at the top of the reactor. The reactor will use internal cooling coils to remove the heat from the reactor solution. The temperature of the fuel solution is about 80°C. The radiation shield of this reactor comprises a graphite and cadmium composition. The fuel can be an aqueous solution of uranyl sulphate, both HEU (90% enrichment) and LEU (21% enrichment). Calculations carried out at IPPE showed that it is possible to produce 500 Ci ^{99}Mo per day (at the date of production) based on one 50 kW homogeneous reactor.

A similar study using a homogeneous reactor for isotope production has been carried out in the USA [17]. The Medical Isotope Production Reactor (MIPR) is an aqueous homogeneous reactor operating at 200 kW, and its configuration is an aqueous solution of uranyl nitrate in water, contained in an aluminum cylinder immersed in a large pool of water which can provide both shielding and a medium for heat transfer. The control rods are inserted at the top through re-entrance thimbles. Provision is made to recombine radiolytic hydrogen and oxygen and to remove other radiolytic gases emitted. Small quantities of the solution can be continuously extracted and replaced after passing through selective ion exchange columns, which are used to extract the desired fission products, e.g. ^{99}Mo. This type of reactor is known for its large negative temperature coefficient, the small amount of fuel required for criticality, and the ease of control. Calculations showed that a 20% enriched ^{235}U system, water reflected, can be critical with 73 liters of solution. The MIPR could produce over 2000 Ci ^{99}Mo per day.

The advantages of the conception of aqueous homogeneous nuclear reactor for production of ^{99}Mo are related to the uniqueness of such type of reactors. The expenditure on constructing and operating a small solution reactor dedicated to ^{99}Mo production should be less than existing or new research reactors. The expense of the management and treatment of the research reactors spent fuel is also higher than for the small homogeneous reactor. At last the whole ^{99}Mo production technology at homogenous reactor is expected to be less complicated in comparison with the existing target technology. All parts of this planned combined production process have been investigated separately. However, this concept has not been tested until now as a process at any facility.

3.2.7. ADONIS: an alternative to nuclear reactors

As an attractive, competitive alternative to nuclear reactors, a subcritical, cyclotron-based spallation neutron source, with neutron multiplication by fission is proposed [23, 24]. The proposed ADONIS system (Acceleration Driven Optimized Nuclear Irradiation System) is designed for the production of fission ^{99}Mo. It is based on the following elements:

1. A proton cyclotron, able to accelerate 2 mA of beam at 150 MeV with low acceleration losses and almost 100% extraction efficiency;
2. A beam transport system, transporting the proton beam without losses to a neutron source;
3. A neutron source including:
   - a primary beam target, where the proton beam strikes a molten Pb–Bi target, producing spallation (mostly evaporation) neutrons;
   - a water moderator surrounding the primary target;
   - a number of secondary targets made of highly enriched ^{235}U.

The neutron multiplication obtained in such a system can be shaped by the amount of secondary targets, but will ultimately remain far from criticality. Besides, due to the inherent limitation of the maximum amount of uranium put into the system, its behavior is non-critical. The concept of sub-criticality indicates the completely different nature of this system compared to reactors. The use of a cyclotron as a driver also allows the quasi-instantaneous shutdown of the system if necessary. The combination of sub-criticality, externally driven neutron source, and the design of the system itself makes ADONIS inherently safe.

3.3. FISSION OF LOW ENRICHED URANIUM

3.3.1. ^{99}Mo production at ANSTO, Lucas Heights, Australia

At ANSTO, ^{99}Mo is produced from 2% enriched uranium which is irradiated as UO_2 pellets in the HIFAR reactor for up to 7 days. The small gap between the fuel pellet and the aluminum irradiation container is filled with MgO to assist in heat dissipation.

The flowsheet of the process of target dissolution and ^{99}Mo separation is shown in Fig. 10.

After irradiation the pellets are separated from the bulk of the MgO powder by sieving, followed by dissolution in concentrated nitric acid. On passing this solution through an alumina column, ^{99}Mo is sorbed and the solution containing uranium and most of the fission products runs through. The column is washed in sequence with nitric acid, water and dilute ammonia solution to remove traces of contaminants. The purified ^{99}Mo is eluted from the column with concentrated ammonia solution, followed by boiling the solution for removal of residual nuclides of iodine and ruthenium [25].

3.3.2. LEU–U_3Si_2 process developed at Karlsruhe, Germany

The Karlsruhe process for the production of ^{99}Mo from UAl_3 target plates has been adapted for the use of U_3Si_2 targets containing LEU [26]. The main differences concern the dissolution of the irradiated target plates. In this regard the normal alkaline dissolution step is followed by treatment of the solid residue with a mixture of HF, H_2O_2 and KIO_3. Afterwards an excess of KOH is added and H_2O_2 is destroyed by boiling. Separation and purification of ^{99}Mo is carried out in a similar way as described in Section 3.2.3. The general flowsheet of the process is presented in Fig. 11.

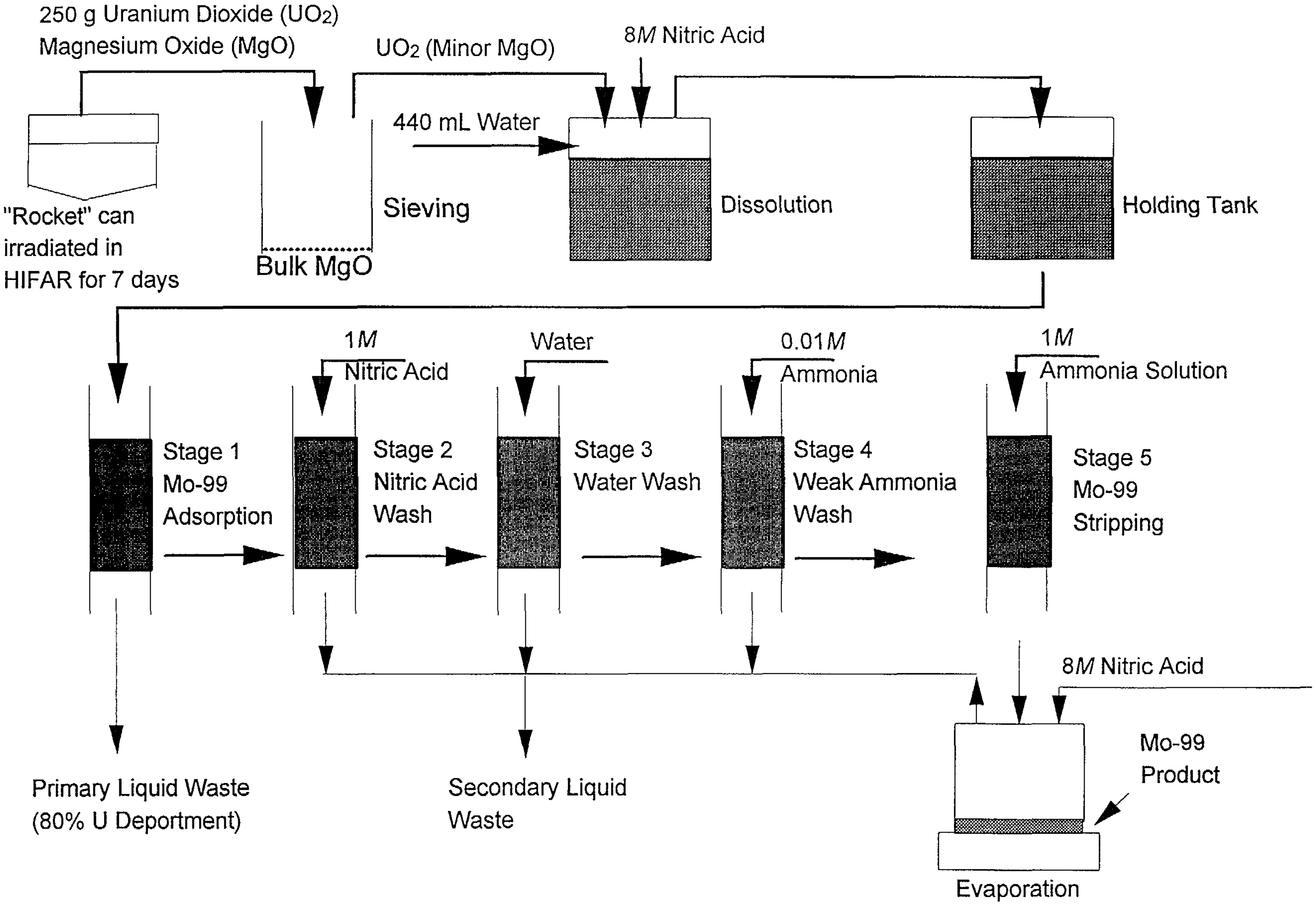


FIG. 10. Schematic process for ^{99}Mo production in Australia.

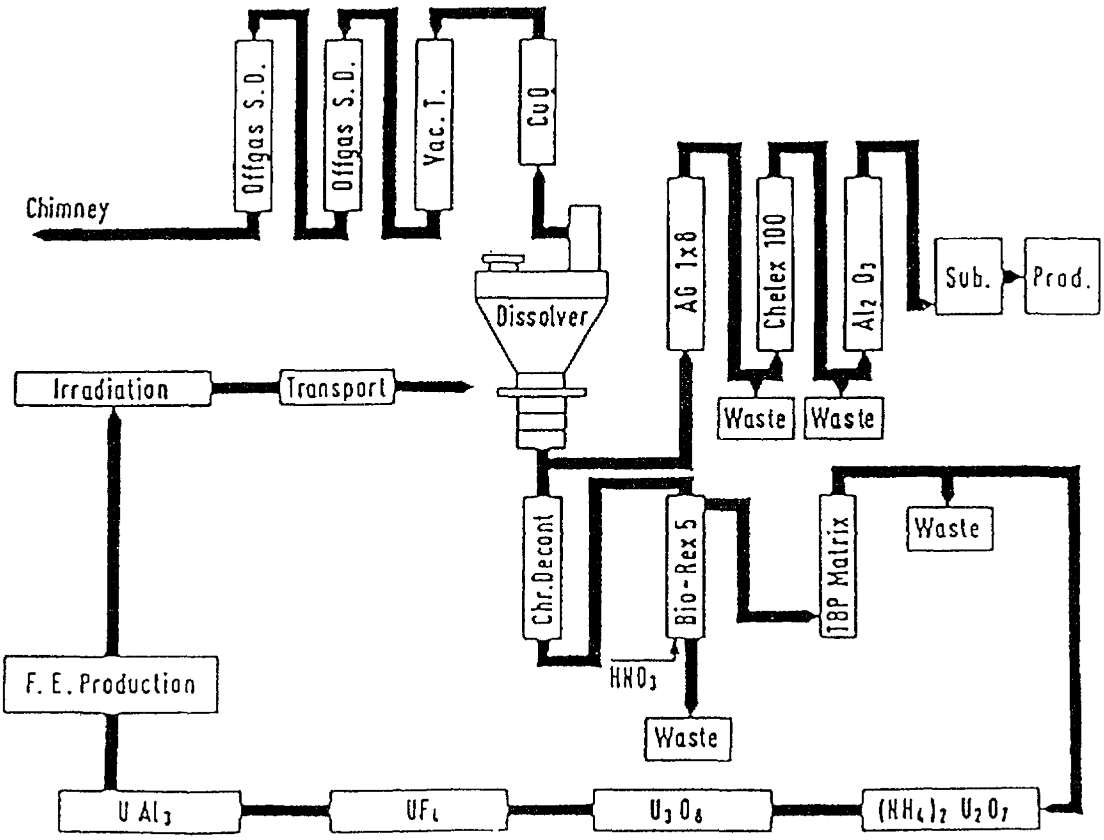


FIG. 11. LEU–U_3Si_2 process developed at Karlsruhe, Germany.

3.3.3. Production of ^{99}Mo using LEU silicide targets developed at Argonne National Laboratory, USA

In October 1992, the US Congress passed an amendment to the Atomic Energy Act of 1954 which prohibits export of HEU for use as fuel or target in research or test reactors unless the following conditions are met:

(1) no alternative low enriched uranium fuel or target can be used;
(2) the US is actively developing an LEU fuel or target for that reactor, and
(3) the proposed recipient of the HEU provides assurances that, whenever possible, an LEU fuel or target will be used in the reactor [27].

Over the last ten years, uranium silicide fuels have been under development as LEU targets for ^{99}Mo production. The use of LEU silicide is aimed at replacing the UAl_x alloy in the HEU. The problem with using uranium silicide targets is that the procedure used to dissolve UAl_x targets is not applicable for the silicide targets, since the latter does not readily dissolve in base. In acid, silica is precipitated in the dissolution process and ^{99}Mo cannot be recovered from the solution [1].

A procedure was proposed to dissolve uranium silicide targets based on the results of research and development work [27–31]. A conceptual flow diagram of the dissolution process is shown in Fig. 12. Initially, the cladding and the aluminum in the irradiated target are dissolved in 3M sodium hydroxide. Sodium nitrate ($NaNO_3$) is added to the system to minimize hydrogen production during the dissolution. A gas sparge is used during the dissolution to remove the gaseous fission products and mix the dissolver contents. Once the cladding is dissolved, the cladding solution is removed from the dissolver leaving the dense uranium silicide behind. A 30% by weight solution of hydrogen peroxide (H_2O_2) is then added with NaOH (5M) to dissolve uranium silicide. The dissolver solution is heated to destroy the H_2O_2 and peroxocomplexes and allow the dissolved uranyl hydroxide to precipitate. This solution is filtered to recover the uranium and remove the base-precipitated fission products. It is then acidified for subsequent recovery of ^{99}Mo using an alumina column.

The problems encountered for the use of LEU silicide targets are:

(1) The uranium silicide target is relatively difficult to dissolve. Although the unirradiated targets can be dissolved by the above procedure, irradiated targets are not readily dissolved. It is suspected that the formation of a $U(AlSi)_3$ surface layer interferes with the dissolution. Further study will be required to resolve this problem [27], and

(2) Loss of ^{99}Mo to the cladding solution is high in this proposed procedure. About 20% of ^{99}Mo is not recovered.

The benefit of using this dissolution procedure is the high degree of separation achieved. As ^{99}Mo is dissolved during the alcalic dissolution, it is purified from U, Np, Pu and many fission products that have insoluble hydroxide salts. A second benefit is that radioxenon can be collected separately from radioiodine.

Substitution of uranium metal for uranium oxide in the target is an alternative for the replacement of HEU by LEU. A detailed discussion of this aspect was given in Ref. [1].

3.4. ACTIVATION OF ^{98}Mo

From the previous two sections it is obvious that the production of ^{99}Mo from the fission of ^{235}U requires very special infrastructure and a strong commitment to large scale production. For smaller countries or organizations, such as the developing Member States, the ^{98}Mo(n, γ) ^{99}Mo route of production offers a viable alternative.

This method consists of the irradiation of targets containing ^{98}Mo (either natural Mo or material enriched in ^{98}Mo), with thermal neutrons in a flux of 10^{13}–10^{14} $n\cdot cm^{-2}\cdot s^{-1}$. Apart from the significantly lower specific activity of ^{99}Mo obtained in this way (about 3 to 4 orders of magnitude lower than fission ^{99}Mo), the production of considerable quantities of ^{99}Mo requires large targets, e.g. 100 g of natural Mo irradiated in a neutron flux of $>10^{14}$ $n\cdot cm^{-2}\cdot s^{-1}$ for 4 to 7 days to produce 100 Ci of ^{99}Mo [1]. Another limitation of this method is that neutron irradiated MoO_3 targets do not produce ^{99}Mo specific activities sufficiently high for the preparation of chromatographic generators with acceptable ^{99m}Tc elution profiles due to the limited adsorption capacity of the alumina on which molybdate anions are adsorbed. This limitation is, however, being overcome by the use of metallic molybdate gels of titanium and zirconium. It is interesting to mention that this type of generator was very much in used, in

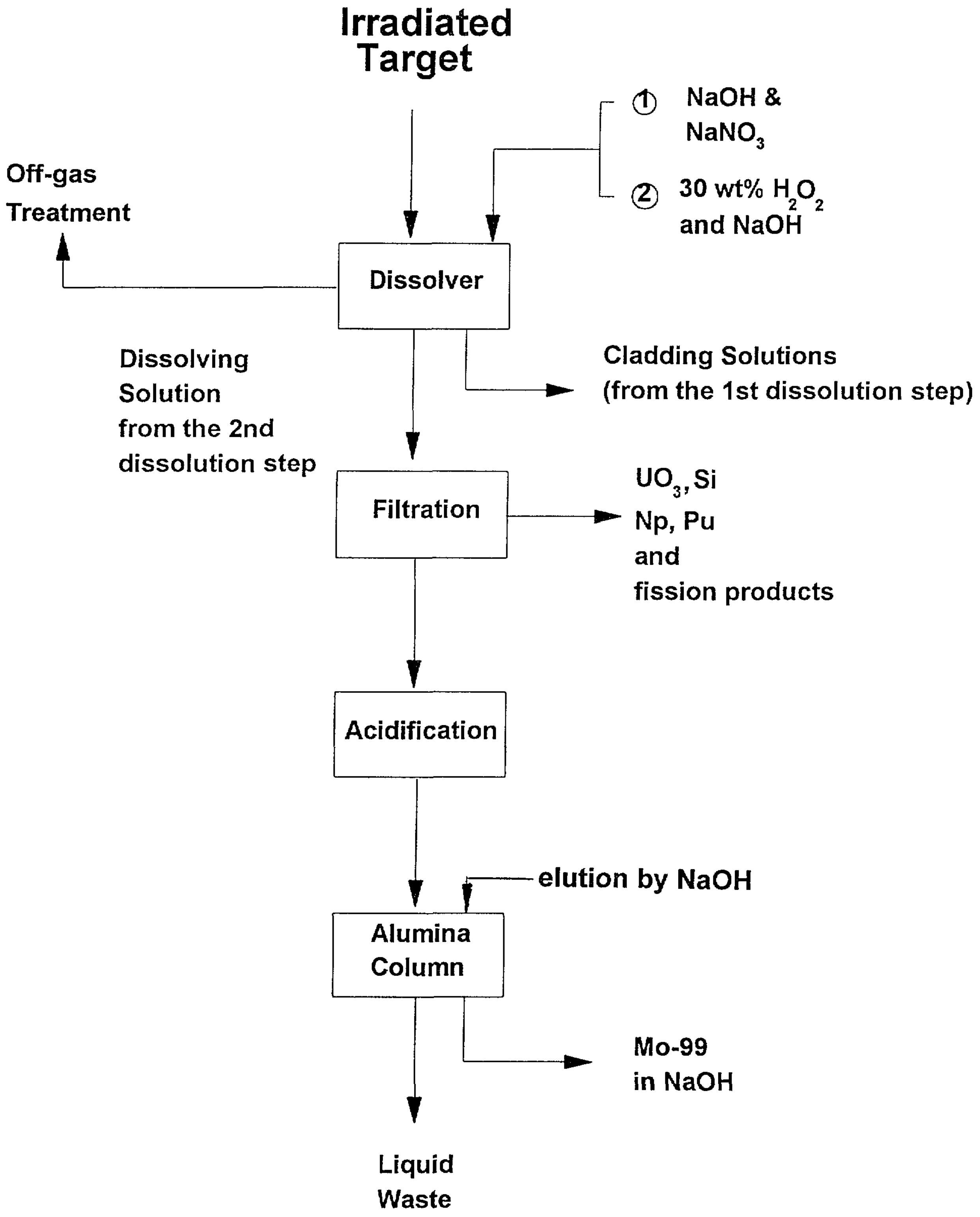


FIG. 12. Conceptual flow diagram of LEU silicide dissolution process, USA.

addition to the solvent extraction system, before the introduction of fission based ^{99}Mo generators at the beginning of the 1970s. The acceptance of the fission chromatographic generators by the nuclear medicine community was almost immediate replacing the neutron irradiated version in technologically advanced countries. Today the fission based generator is considered the gold standard of the technology.

Nevertheless, countries operating nuclear research reactors but not able or not willing to commit the required technological and economical resources to set up the required radiochemical facilities for the production of fission ^{99}Mo, have developed and further refined the technology for the separation of ^{99m}Tc from ^{99}Mo using the solvent extraction and sublimation alternative methods. Based on these two methods, several developing countries today supply reliably (although in limited amounts and sometimes as a supplement to imported fission-based ^{99}Mo generators) the so-called instant ^{99m}Tc to hospitals on a daily basis.

The irradiation of high purity MoO_3 in nuclear reactors does not produce appreciable amounts of other radionuclides that contribute to the radioactive waste inventory in any significant fashion.

Solvent extraction and sublimation separation systems render themselves more suitable for centralized production-distribution facilities as practice in such countries like India, Peru, Thailand and many others. As a matter of fact, in India, small solvent extraction units have been installed in hospitals across the country which are weekly supplied with molybdate solutions containing neutron activated ^{99}Mo. The preparation of solvent extraction and sublimation generators is described in detail in the IAEA-TECDOC-515 [1].

^{99}Mo/^{99m}Tc separation processes in solvent extraction systems are carried out in well shielded hot cells usually installed within the premises of a nuclear research establishment under the supervision of well trained and qualified staff. It essentially consists of a glass-quartz, sometimes stainless steel, apparatus where activated $^{99}MoO_3$ is dissolved in an acid or base (HCl, NaOH) and mixed with an organic solvent, usually methyl-ethyl-ketone (MEK), which has the property to preferentially concentrate ^{99m}Tc over ^{99}Mo. Once equilibrium is reached, the organic solvent containing ^{99m}Tc as a decay product of ^{99}Mo is separated from the aqueous layer into a different vessel and evaporated out leaving behind a white powder of a technetium salt. This water soluble salt is then dissolved in a sterilized physiological saline solution (0.9 % NaCl) which is then ready for distribution to the users in hospitals after proper quality control procedures have been satisfactorily performed. The parent ^{99}Mo contained in the aqueous layer continues to produce further ^{99m}Tc which, after a proper equilibration period of 1–2 days, is ready for subsequent extractions with an organic solvent.

In the sublimation generator system the separation of ^{99m}Tc from ^{99}Mo is achieved by subjecting the activated MoO_3 to high temperatures of over 900°C, a temperature at which an oxide of ^{99m}Tc (Tc_2O_7) is sublimed and collected in a separated vessel. The oxide of technetium is then dissolved in a physiological saline solution which is ready further use. Sublimation systems produce ^{99m}Tc of the highest purity and high radioactivity concentration. As in the case of solvent extraction systems, sublimation apparatuses have to be installed in a shielded hot cell. This system has been used extensively for quite some time in Australia and Hungary. Several versions of sublimation systems have been developed in the recent past using different substrates, such as $SiC\text{-}MoO_3\text{-}V_2O_5$ containing activated ^{99}Mo from which ^{99m}Tc can be separated out at a lower temperature , i.e. 640°C.

Solvent extraction and sublimation systems have proved practical and economical, and capable of providing ^{99m}Tc solutions of high activity and quality for the labelling of radiopharmaceuticals kits.

Medium to high energy nuclear reactions such as ^{100}Mo(p,2n)^{99}Mo and ^{100}Mo(p,pn)^{99}Mo are currently being investigated to assess the viability of using accelerators to produce ^{99}Mo economically and in significant amounts. The above reactions would required a source of enriched ^{100}Mo and accelerators able to deliver proton beams of about 70 MeV at high currents [18]. The issues that need to be addressed in the context of ^{99}Mo production for the preparation of ^{99m}Tc generators for medical use include radionuclidic impurities, specific activities, targetry, distribution logistics, licensing process and economics.

Although the capital layout and operating costs of accelerators will be much lower than for reactors, such a ^{99}Mo production route will still require a substantial investment.

Another interesting way of ^{99}Mo production which is being considered, is the use of a cyclotron to bombard a target of molten lead and bismuth with 150 MeV H^+ ions, thus creating neutrons by spallation. This alternative is discussed in more detail in Section 3.2.7. Although similar waste to other fission ^{99}Mo processes will be formed, this method of ^{99}Mo production should be safer and cheaper to operate than a reactor [32].

Although the use of accelerators offer interesting possibilities for the future, the technology has not reached the stage where it can be considered for ^{99}Mo production within the next few years.

4. WASTE CHARACTERISTICS FROM ^{99}Mo PRODUCTION

4.1. INTRODUCTION

From Section 3 it is clear that the production of ^{99}Mo by means of neutron induced fission of ^{235}U (as HEU or LEU) is operated in some countries on an industrial scale. Although this production method supplies nearly the whole worldwide demand for ^{99}Mo, it also generates the most radioactive waste of all production methods. This arises from the fact that a wide range of nuclides are formed during ^{235}U fission.

The complexity of the radionuclidic source term to be handled during fission ^{99}Mo recovery and purification can be illustrated by calculated data for the nuclides present in ^{235}U target, after irradiation by a neutron flux in reactor. The implication of this irradiation is that radionuclides with widely differing chemical and physical properties must be handled in a safe way. As must also be appreciated, this combination of nuclides creates very large exposure rates, with the risk of radiation doses to personnel.

Apart from the radiation dose, the processing of irradiated uranium targets presents two particular problems from the perspective of waste management. First, xenon and krypton, as well as iodine (depending on the process conditions), will be liberated in gaseous form during target dissolution. These gases must be removed efficiently from hot cell off-gas streams to prevent their release into the environment. In all countries strict regulations are in force regarding the release of such nuclides, especially for ^{131}I and ^{133}I (mostly <1 Ci per year per installation).

Second, the presence of long lived nuclides must be taken into consideration when the recovery and purification of unfissioned uranium (a valuable resource) are carried out or are under consideration. Careful planning and process development is necessary to protect personnel against high radiation doses during handling, treatment, storage, and disposal of waste or to prevent the generation of large waste volumes (which must be subsequently disposed of).

4.2. THEORETICAL ASSESSMENT OF THE FISSION

A simplified theoretical calculation code giving the instantaneous activity of a considered fission product as a function of mass of ^{235}U, its cross-section, the average neutron flux and the irradiation time, only provides an approach for the estimate of the radionuclide spectrum upon leaving the reactor. It does not take into account the radionuclides which are formed by neutron activation of the non fissile elements in the target during the irradiation period, nor the radioactive decay products of radionuclides formed by fission or by activation in the same period. The calculation model drawn up for the ORIGEN programme presents a more complete solution, which takes into account these elements and gives a complete radionuclide spectrum of a target upon leaving the reactor and/or given cooling period.

The study of the evolution in time of the isotopic composition of a given mixture, whether or not under neutron flux, requires the solution of a system of first order differential equations (the Bareman equations). In the case of the waste produced by ^{235}U fission, this mixture contains several hundreds of isotopes. This problem could be solved using a computer code ORIGEN 2.1 for PC [33]. This programme allows one to solve the above mentioned equations and to produce the isotopic composition of the mixture at any time of irradiation of the targets and of decay of radioisotopes produced. The calculation was made with a library of cross-sections calculated for thermal neutrons.

A calculation was done on the basis of a real process involving 9 targets totaling 35.4 g of ^{235}U, irradiated at $1.54 \cdot 10^{14}$ n/cm^2·s for 103.5 h. The initial composition of the target is presented in Table II.

The obtained activities have been converted into MBq upon leaving the reactor, at the date of calibration (+7 days) and after one and two years of cooling. The results of calculation are discussed below separately for activation products, actinides and fission products.

4.2.1. Calculation of the activation products

The influence of the elements other than the uranium isotopes, which are found in the composition of the targets, was determined by a "blank" calculation simulating an irradiation without uranium. Only the aluminum (UAlx + Al cladding) and the impurities of uranium and aluminum was taken into account. The same irradiation conditions were applied.

Upon leaving the reactor, a list is obtained of 688 activation products formed including their decay products. 157 of these are radioactive, generally very weakly. The total activity is of the order of $2 \cdot 10^8$ MBq upon leaving the reactor. The activity decreased to $7 \cdot 10^3$ MBq after seven days and to $1.9 \cdot 10^3$ MBq after one year. Lists of radionuclides in Tables III and IV are drawn in the order of the decreasing activities. If the radionuclides with an original activity of less than 1 MBq are disregarded, 50 remain, but the activity is mainly concentrated on the ^{28}Al with a very short half-life. The main activation products are given in Table III.

TABLE II. INITIAL COMPOSITION OF ^{235}U TARGET BEFORE IRRADIATION

Composition:	g	%(U)
^{234}U	4.37E–01	1.15
^{235}U	3.54E+01	93.20
^{236}U	7.60E–03	0.02
^{238}U	2.15E+00	5.65
U total	3.80E+01	
	g	**% total**
Ag	7.60E–05	1.92E–05
Al	3.19E+02	8.05E+01
B	3.23E–03	8.15E–04
C	5.17E–03	1.30E–03
Ca	3.23E–04	8.14E–05
Cd	2.17E–03	5.47E–04
Co	7.60E–05	1.92E–05
Cr	6.13E–03	1.55E–03
Cu	8.36E–03	2.11E–03
Fe	7.52E–01	1.90E–01
Li	3.23E–03	8.15E–04
Mn	1.44E–02	3.63E–03
Mg	7.77E–03	1.96E–03
Mo	3.45E–03	8.71E–04
Ni	1.33E–03	3.35E–04
P	4.94E–04	1.25E–04
Pb	3.61E–04	9.10E–05
Si	2.10E–01	5.30E–02
Sn	3.80E–04	9.58E–05
Ti	4.79E–02	1.21E–02
W	2.28E–04	5.75E–05
Zr	5.13E–04	1.29E–04
Zn	3.51E–02	8.86E–03
Total	3.96E+02	

TABLE III. MAIN INITIAL ACTIVATION PRODUCTS IN THE IRRADIATED TARGET

Radionuclides	**$T_{1/2}$ (h)**	**Activity (MBq)**	%
	Total:	2.03E+08	100.000
^{28}Al	3.73E–02	2.03E+08	99.842
^{56}Mn	2.58E+00	2.60E+05	0.128
^{64}Cu	1.27E+01	3.05E+04	0.015
^{69}Zn	9.50E–01	8.06E+03	0.004
^{66}Cu	8.50E–02	6.56E+03	0.003
^{110}Ag	6.83E–03	2.36E+03	0.001
^{60m}Co	1.75E–01	2.02E+03	0.001
^{131}S	2.62E+00	1.85E+03	0.001
^{3}H	1.08E±5	1.60E+03	0.001
^{8}Li	2.34E–04	1.22E+03	0.001
^{8}Be	5.56E–10	1.22E+03	0.001
^{108}Ag	3.95E–02	9.80E+02	0.001

After one year, only 4 radionuclides with an activity of more than 1 MBq remain (Table IV).

TABLE IV. MAIN ACTIVATION PRODUCTS IN THE TARGET ONE YEAR AFTER IRRADIATION

Radionuclides	**$T_{1/2}$(h)**	**Activity (MBq)**	%
	Total	1.90E+03	100.00
^{3}H	1.08E+05	1.51E+03	79.46
^{55}Fe	2.28E+04	3.19E+02	16.73
^{65}Zn	5.85E+03	6.66E+01	3.50
^{60}Co	4.62E+04	5.12E+00	0.27
^{59}Fe	1.08E+03	8.34E–01	0.04

From the above data it could be concluded that the weak activities resulting from the activation of the aluminum and of the impurities of the uranium constituting the targets are fully negligible.

4.2.2. Calculation of actinides

On the basis of the mentioned uranium composition, a list is obtained of 129 nuclides representing the actinides and their decay products. The activities below $1 \cdot 10^{-3}$ MBq are disregarded. The remaining list of 17 radionuclides is given in decreasing order of activity (Table V).

TABLE V. INITIAL COMPOSITION OF ACTINIDES IN IRRADIATED TARGET

Actinides	**$T_{1/2}$ (h)**	**Activity (MBq)**	**%**
	Total:	3.97E+06	100.000
^{239}U	3.92E–01	2.26E+06	57.062
^{239}Np	5.65E+01	1.62E+06	40.760
^{237}U	1.62E+02	8.28E+04	2.089
^{240m}Np	1.23E–01	2.28E+03	0.058
^{240}Np	1.08E–00	1.02E+03	0.026
^{234}U	2.14E+09	1.00E+02	0.003
^{238}Np	5.08E+01	8.07E+01	0.002
^{240}U	1.41E+01	1.54E+01	0.000
^{235}U	6.17E+12	2.72E+00	0.000
^{231}Th	2.55E+01	2.68E+00	0.000
^{236}U	2.05E+11	4.91E–1	0.000
^{239}Pu	2.11E+08	3.25E–01	0.000
^{238}U	3.92E+13	2.68E–02	0.000
^{241}Pu	1.26E+05	2.00E–02	0.000
^{232}Pa	3.14E+01	1.18E–02	0.000
^{240}Pu	5.73E+07	9.22E–03	0.000
234Th	5.78E+02	3.12E–03	0.000
^{234m}Pa	1.95E+02	3.12E–03	0.000
^{238}Pu	7.69E+05	1.65E–03	0.000

After one year, the total activity has decreased from $4 \cdot 10^{6}$ MBq to $1.1 \cdot 10^{2}$ MBq. If activities of less than $1 \cdot 10^{-3}$ MBq are disregarded, the reduced list would be as follows (Table VI).

TABLE VI. MAIN ACTINIDES REMAIN IN THE TARGET ONE YEAR AFTER IRRADIATION

Actinides	$T_{1/2}$ **(h)**	**Activity (MBq)**	**%**
^{234}U	2.14E+09	1.00E+02	93.648
^{231}Th	2.55E+01	2.72E+00	2.538
^{235}U	5.17E+12	2.72E+00	2.538
^{239}Pu	2.11E+08	7.62E–01	0.710
^{236}U	2.05E+11	4.91E–01	0.457
^{234}Th	5.78E+02	2.68E–02	0.025
^{234m}Pa	1.95E+02	2.68E–02	0.025
^{238}U	3.92E+13	2.68E–02	0.025
^{241}Pu	1.26E+05	1.91E–02	0.018
^{240}Pu	5.73E+07	9.25E–03	0.009
^{238}Pu	7.69E+05	6.93E–03	0.006

After two years this spectrum is practically the same.

4.2.3. Calculation of fission products

Upon leaving the reactor, a list of the resulting fission products is obtained on 879 nuclides totaling almost $4 \cdot 10^{10}$ MBq. Some 640 radionuclides show an activity between 1 MBq and $6 \cdot 10^{8}$ MBq.

After one year of decay, the total activity decreases to $7.1 \cdot 10^{6}$ MBq and after two years to $2.6 \cdot 10^{6}$ MBq. If the radionuclides with an activity after one year of less than 1 MBq are disregarded (this corresponds to a sum of 0.01% of the total activity), this list is reduced to 33 radionuclides, the activity of which is comprised between 1 MBq and $1.9 \cdot 10^{6}$ MBq (Table VII).

4.3. ^{99}Mo PRODUCTION FROM HIGHLY ENRICHED URANIUM

4.3.1. Characterization of waste generated by ^{99}Mo production at CRL, Canada

A variety of waste streams are generated by the ^{99}Mo production process at CRL. The wastes are processed or temporarily stored according to established procedures. The compositions and main radionuclides in the waste streams for the liquid and solid wastes are summarized in Tables VIII and IX, respectively.

The fissile liquid waste containing uranium and fission products generated by the ^{99}Mo production process is in the solution from the dissolver and the first wash solution from the alumina column. This liquid waste is classified as high level waste (HLW) based upon its activity and radiation.

TABLE VII. COMPOSITION OF FISSION PRODUCTS IN THE TARGET ONE OR TWO YEARS AFTER IRRADIATION

Fission products	$T_{1/2}$ (h)	**Activity after 1 year (MBq)**	%	**Activity after 2 years (MBq)**	%
Total		7.11E+06	100.00	2.58E+06	100.00
^{144}Ce	6.82E+03	1.89E+06	26.53	7.74E+05	30.00
^{144}Pr	2.88E–01	1.89E+06	26.53	7.74E+05	30.00
^{147}Pm	2.30E+04	4.40E+05	5.18	3.37E+05	13.07
^{137}Cs	2.63E+05	1.30E+05	1.83	1.28E+05	4.94
^{90}Y	6.40E+01	1.27E+05	1.78	1.24E+05	4.79
^{90}Sr	2.55E+05	1.27E+05	1.78	1.24E+05	4.79
^{137m}Ba	4.25E–02	1.23E+05	1.73	1.21E+05	4.67
^{106}Ru	8.84E+03	1.28E+05	1.80	6.43E+04	2.49
^{106}Rh	8.31E–03	1.28E+05	1.80	6.43E+04	2.49
^{95}Nb	8.44E+02	1.04E+05	14.63	2.06E+04	0.80
^{85}Kr	9.40E+04	1.55E+04	0.22	1.45E+04	0.56
^{144m}Pr	1.20E–01	2.26E+04	0.32	9.29E+03	0.36
^{95}Zr	1.54E+03	4.69E+05	6.59	8.97E+03	0.35
^{91}Y	1.40E+03	3.27E+05	4.60	4.32E+03	0.17
^{125}Sb	2.43E+04	5.49E+03	0.08	4.27E+03	0.17
^{155}Eu	4.35E+04	3.38E+03	0.05	2.94E+03	0.11
^{151}Sm	7.89E+05	2.48E+03	0.03	2.46E+03	0.10
^{125m}Te	1.39E+03	1.32E+03	0.02	1.04E+03	0.04
^{89}Sr	1.21E+03	1.56E+05	2.20	1.04E+03	0.04
^{3}H	1.08E+05	5.50E+02	0.01	5.20E+02	0.02
^{127m}Te	2.62E+03	4.11E+03	0.06	4.03E+02	0.02
^{127}Te	9.35E+00	4.03E+03	0.06	3.95E+02	0.02
^{123}Sn	3.10E–03	1.16E+03	0.02	1.63E+02	0.01
^{134}Cs	1.81E+04	1.13E+02	0.00	8.07E+01	0.00
^{95m}Nb	8.66E+01	3.48E+03	0.05	6.65E+01	0.00
^{103}Ru	9.43E+02	3.15E+04	0.44	5.00E+01	0.00
^{103m}Rh	9.35E–01	2.84E+04	0.40	4.51E+01	0.00
^{154}Eu	7.54E+04	4.58E+01	0.00	4.22E+01	0.00
^{119m}Sn	5.88E+03	5.63E+01	0.00	2.00E+01	0.00
^{99}Tc	1.87E+09	1.91E+01	0.00	1.91E+01	0.00
^{113m}Cd	1.28E+05	1.17E+01	0.00	1.12E+01	0.00
^{141}Ce	7.80E+02	1.88E+04	0.26	7.81E+00	0.00
^{93}Zr	1.34E+10	2.77E+00	0.00	2.77E+00	0.00

TABLE VIII. CHARACTERISTICS OF LIQUID WASTE GENERATED BY THE ^{99}Mo PRODUCTION PROCESS AT CRL

Waste stream	**Composition**	**Main radionuclides**
Fissile liquid waste	NO_3^{-1}. Al^{+3}, Hg^{+2} (low pH)	U, Pu, Ce, ^{3}H, Se, Zr, Mo, Tc, Ru, Rh, Sn, Sb, Cs, Zn
Non–fissile liquid waste	NH_4^{+1}, Na^{+1}	fission products
Decontamination waste	H_2O	low-levels of fission products

TABLE IX. CHARACTERISTICS OF SOLID WASTE GENERATED BY THE ^{99}Mo PRODUCTION PROCESS AT CRL

Waste stream	**Composition**	**Main radionuclides**
Spent alumina column	Al_2O_3	^{3}H, Zr, Mo, Tc, Ru, Rh, Ce, Cs, Co
In-cell waste	aluminum sheath, defective equipment, molecular sieve, charcoal and filters.	Small amounts of U, Pu & fission products
Air filters	mainly HEPA filters	^{3}H, Xe, I, Kr, Cs
Charcoal	carbon	^{3}H, I, Xe, Kr
Molecular sieve	molecular sieve	^{3}H, Kr, Xe, Cs

Non-fissile liquid wastes generated by the ^{99}Mo-production process, such as the second and subsequent alumina column washes and scrubber solutions are designated as intermediate level waste (ILW) based upon its activity and radiation.

Liquid waste is also generated during the decontamination of in-cell surfaces and by steam cleaning. This waste is classified as low level waste (LLW) which is combined with other liquid wastes generated at CRL.

Solid wastes generated from the process include alumina columns, in-cell waste, air filters, charcoal and molecular sieves. The alumina columns and in-cell waste are designated as ILW. Process off-gas filters and molecular sieves are designated LLW.

Process off-gas, mainly oxides of nitrogen and volatile fission products such as Xe, I, Kr and ^{3}H are generated during dissolution of the irradiated targets and is treated to reduce emission below permissible limits then discharged.

4.3.2. Waste arising from ^{99}Mo production at the IRE, Belgium

The distribution of U and fission products during processing of irradiated uranium targets and formation of different waste streams is presented in Fig. 13. The dissolution of the targets liberates xenon, which is used for commercial applications (see Section 3.2.2). During the acidification iodine is released and also used for commercial applications.

From the alumina column (Woelm A, Super 1) the fission products are eluted and fixed in the effluents (ILLW) and in the subsequent washes of 1N HNO_3, H_2O and 0,01N NH_4OH. The main characteristics of the liquid waste from ^{99}Mo production received by Belgoprocess from IRE during several years for processing are summarized in Tables X and XI.

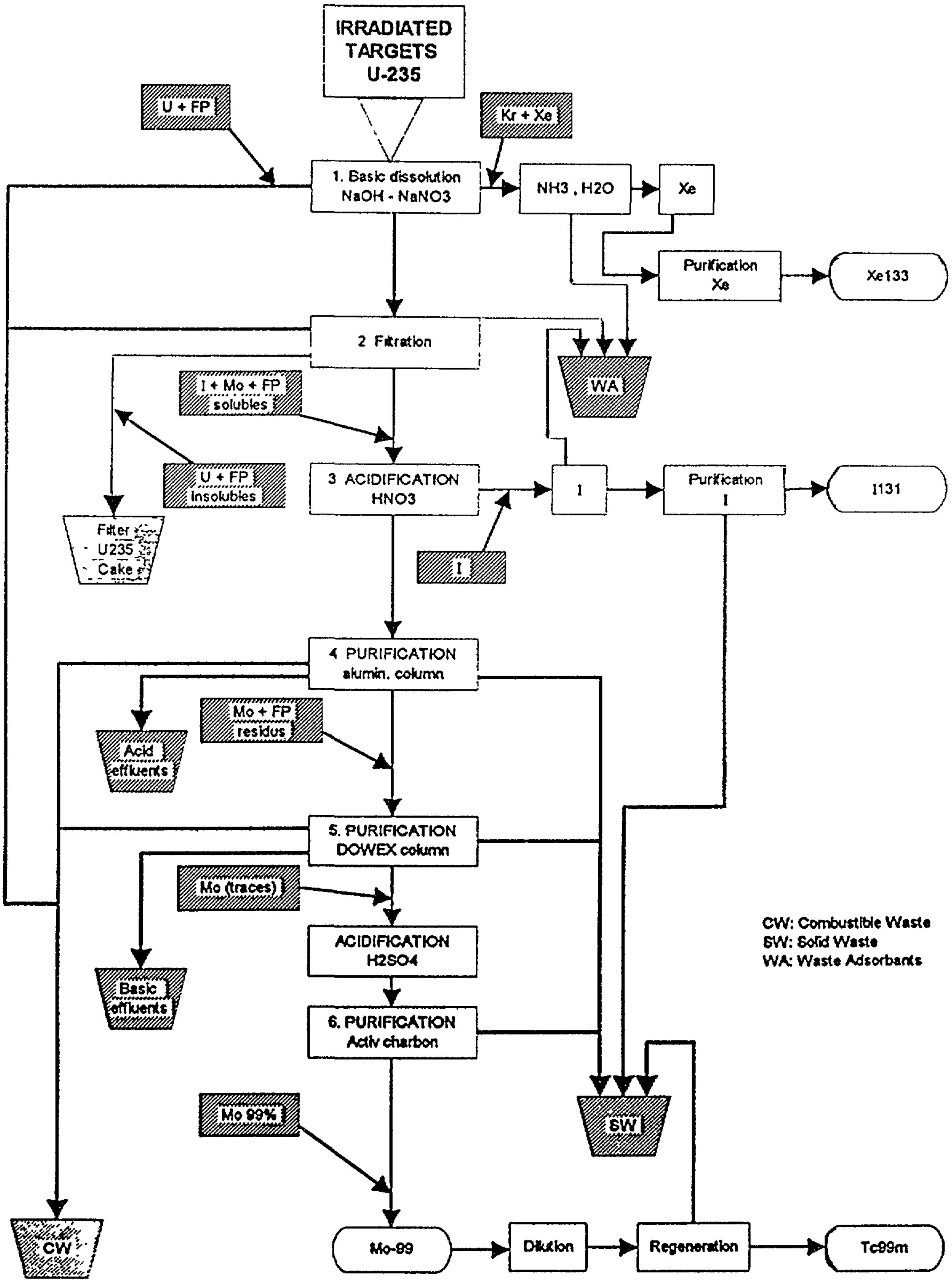


FIG. 13. Distribution of uranium and fission products in the different waste streams of the I.R.E. process, Belgium.

TABLE X. AVERAGE CHEMICAL COMPOSITION (THE MOST IMPORTANT ELEMENTS) OF THE LOW- AND INTERMEDIATE LEVEL WASTE RECEIVED BY BELGOPROCESS FROM IRE FLEURUS FOR PROCESSING

Batch from	Volume In liter	OH^- N	H^+ free N	Al^{3+} M	NH_4 M	Tot. salts g/l	NO_3^- g/l	NO_2^- ppm	Cl^- mg/l	F^- M	Total salts In kg	BWP drums
1985	2181		0,74	0,44	0,03	390	280	<20	25	5,00E-0	851	10,6
1986	2300	0,16		0,15	0,31	71			140		163	2,0
1987	2500		0,63			366	228	5			915	11,4
1987	700		0,15			24	14	<0,5			17	0,2
1988	2600		0,59			397	238	5	100	1,00E-0	1032	12,9
1989	5638					110	74	50	130	1,00E-0	620	7,8
1990	2400					340	126	<5	5,5	2,00E-0	816	10,2
1991	3470					242					840	10,5
1992	3500					213					746	9,3
1993	3487					356					1241	15,5
1995	3012					232	160	<1	24	1,00E-0	699	8,7
1996	4857					336	219	<100	660	0,05	1632	20,4
Average	36645					261	169				9571	

TABLE XI. AVERAGE RADIOLOGICAL COMPOSITION (THE MOST IMPORTANT ISOTOPES) OF LOW- AND INTERMEDIATE LEVEL WASTE RECEIVED BY BELGOPROCESS FROM IRE FLEURUS FOR PROCESSING

Batch from	Volume in liter	Alpha tot. in Bq/dm3	Beta tot. in Bq/dm3	Cs 137 in Bq/dm3	Ce/Pr 144 in Bq/dm3	Ru/Rh 106 in Bq/dm3	Zr 95 in Bq/dm3	Nb 95 in Bq/dm3	Ru 103 in Bq/dm3	Sb125 in Bq/dm3	Sr/Y 90 in Bq/dm3	Pu tot. in Bq/dm3
1985	2181	5,92E+05	9,89E+09	4,08E+09	3,23E+09	4,85E+08	2,37E+08	1,26E+08	1,15E+08			
1986	2300	7,40E+03	6,88E+07	2,00E+07		8,10E+06			2,40E+05	3,00E+07		2,20E+02
1987	2500	1,33E+05	8,28E+09									
1987	700	2,70E+03	1,00E+08									
1988	2600	3,90E+07	7,40E+09	4,60E+09	1,60E+09	5,20E+08	6,00E+07	1,10E+08			5,40E+08	
1989	5638	2,50E+05	2,10E+09	1,19E+09	4,30E+08	1,30E+08	6,00E+07	6,00E+07	1,60E+08		1,30E+08	
1990	2400	7,60E+04	6,60E+09	4,80E+09	1,84E+09	2,20E+07	9,40E+07	9,40E+07				
1991	3470	7,30E+07	1,11E+10									
1992	3500	2,35E+06	1,20E+10									
1993	3487	2,22E+05	9,32E+09									
1995	3012	1,30E+05	5,80E+09	3,00E+09	1,80E+09	7,20E+08						
1996	4857	3,16E+04	3,36E+09	2,19E+09	1,79E+08	3,03E+08	6,27E+06	1,03E+09		3,09E+07	1,76E+08	
Average		6,91E+06	4,77E+09	2,81E+09	1,07E+09	2,46E+08	7,17E+07	3,47E+08				

Batch from	Alpha tot. in Bq	Beta tot. in Bq	Cs 137 tot.in Bq	Ce/Pr 144 tot.in Bq	Rh 106 tot.in Bq	Zr 95 tot.in Bq	Nb 95 tot.in Bq	Ru 103 tot.in Bq	Sb125 tot.in Bq	Sr/Y 90 tot.in Bq	Pu tot. tot.in Bq
1985	1,29E+09	2,11E+13	8,90E+12	7,04E+12	1,06E+12	5,16E+11	2,74E+11	2,50E+11			
1986	1,70E+07	1,58E+11	4,60E+10		1,86E+10			5,52E+08	6,90E+07		5,06E+05
1987	3,33E+08	2,07E+13									
1987	1,89E+06	7,00E+10									
1988	1,01E+11	1,92E+13	1,20E+13	4,16E+12	1,35E+12	1,56E+11	2,86E+11			1,40E+12	
1989	1,41E+09	1,18E+13	6,71E+12	2,42E+12	7,33E+11	3,38E+11	3,38E+11	9,02E+11		7,33E+11	
1990	1,82E+08	1,58E+13	1,15E+13	4,42E+12	5,28E+10	2,26E+11	2,26E+11				
1991	2,53E+11	3,85E+13									
1992	8,23E+09	4,20E+13									
1993	7,74E+08	3,25E+13									
1995	3,92E+08	1,75E+13	9,04E+12	5,42E+12	2,17E+12						
1996	1,53E+08	1,63E+13	1,06E+13	8,69E+11	1,47E+12	3,05E+10				8,55E+11	

Solid waste is generated mainly from spent ion exchange resins and the alumina columns. Another type of solid waste is filter material from off-gas cleaning of the process, such as activated charcoal filters or absolute filters. A relatively small amount of solid waste arises from the replacement of some parts of the ^{99}Mo production plant like valves, pumps, tubes, etc. The remaining U and fission products retained in the dissolver are transferred to a storage tank. The slurry contains the highest activity such as lanthanides, ruthenium, zirconium, etc. In Belgium, this slurry is stored in a vessel awaiting further treatment.

4.3.3. Waste generation from fission of HEU at Mallinckrodt/ECN, Netherlands

The volatile fission products with ^{133}Xe as the main are flushed by a stream of nitrogen. They leave the dissolver at its upper end and pass through a reflux condenser together with the hydrogen generated during the digestion process.

The separation process of the HEU target leads to an alkaline waste stream arising from passing through the AG-1, SM-7, and MnO_2 columns and an acidic waste stream arising from passing through chelex-100 exchangers (see Fig. 6). The alkaline waste solution contains nearly the total amount of aluminum from the target and an essential amount of the fission products. The characteristics of the main liquid-streams are compiled in Table XII. The acidic waste stream consists of sulphuric acid containing thiocyanate, sulphite and the remaining fission products.

TABLE XII. ESTIMATED CHARACTERISTICS OF LIQUID WASTE FROM ^{99}Mo PRODUCTION AT MALLINCKRODT/ECN PER HOT CELL AFTER PRODUCTION OF 3000 Ci ^{99}Mo

Liquid	**Volume (L)**	**Content (g/L)**		**Activity MBq/L after 0.5 year storage on site**	
Cell 1, ILW	13.3	NaOH	244	Sr-89	7000
		Aluminum	20	Sr-90	1500
		Uranium	0.050	Cs-137	7000
Cell 2, ILW	8.7	$NaNO_3$	102	Ru-103	740
		NaOH	29.6	Ru-106	370
				Sb-125	37
Cell 3 LLW	7.6	Na_2SO_4	115	Ru-103	70
		NaI	0.06	Ru-106	40
		Na_2SO_3	121	Sb-125	4
		NaOH	16	Sn-126	1
Cell 4 LLW	9.0	NaOH	21.2	Mo-99	10
		Na_2SO_4	116.4	Tc-99	10
		NaSCN	4.0	Ru, Sb, Sn	20
		Na_2SO_3	1.3		
		KI	0.06		
Cell 5 LLW	3.0	NH_4NO_3	50.4	Ru-103	$4 . 10^{-3}$

The ^{99}Mo purification process generates only small volumes of liquid wastes with relatively low activity level and consist mainly of aqueous solutions or solutions of low concentration of acidic or alkaline media.

Solid waste is generated mainly from spent ion exchange resins and absorber columns. Other types of solid waste are filter material from off-gas cleaning, such as activated charcoal filters or absolute filters from the hot cell. Small amounts of solid waste arise from the replacement of parts of the whole ^{99}Mo production plant like valves, pumps, tubes etc. The characteristics of the main solid wastes are compiled in Table XIII.

TABLE XIII. ESTIMATED CHARACTERISTICS OF SOLID WASTE FROM ^{99}Mo PRODUCTION AT MALLINCKRODT/ECN PER HOT CELL AFTER PRODUCTION OF 3000 Ci ^{99}Mo

Solid waste from	**Components**	**Activity (Ci) some days after production**	
Cell 1, ILW	filters sinter metals tissues pipes/tubes	Estimation 1% of the total start activity	
		U-234	2.5E-5
		U-235	7.9E-7
		U-236	1.0E-7
		Pu-239	3.0E-7
		Sr-89	3.9E0
		Y-90	1.35E2
		Y-91	4.13E0
		Zr-95	4.04E0
		Nb-95	2.7E-1
		Ru-103	3.11E0
		Ru-106	4.44E–2
		Ce-141	7.95E0
		Ce-144	8.48E–1
		Sm-151	4.65E–4
Cell 2, ILW	AG-1, SM-7 and empty MnO_2 columns, PVC-tubes, stainless steel components, tissues	% activity in cell	
		I-131	5%
		Ru-103	90%
		Ru-106	5%
Cell 3, ILW	Chelex-columns, polyethylene bottles	% activity in cell	
		Ru-106	10%
		Ru-103	90%
Cell 4, LLW	tissues	Mo-99 tracers	
Cell 5, LLW	tissues	Mo-99 tracers	
Replacement components when necessary, ILW/LLW	dissolvers drop collector in cell storage tanks	various activity	

The remaining U and fission products are retained in the dissolver by Bio-Rex-5 containing the highest activity such as lanthanides, ruthenium, zirconium, etc. In the Netherlands the material of the spent target is sent back to the target producer for reprocessing. There will be just some traces of U and α-emitters in the liquid waste.

4.3.4. Waste from ^{99}Mo production at the AEC, South Africa

The following waste streams result from the dissolution of target plates and ^{99}Mo purification: gaseous, alkaline liquid containing inter alia cesium, acidic liquid containing short-lived nuclides, and short-lived solid waste. During future recovery and purification of unfissioned uranium, intermediate level liquid and low active solid waste will be produced. Due to the short irradiation period of target plates, no highly active waste and very low quantities of transuranium elements are formed.

Gaseous waste consists mainly of various fission gases, predominantly xenon nuclides and small quantities of krypton. Small amounts of gaseous iodine, mainly ^{131}I and ^{133}I, may also be liberated during plate dissolution or other stages of the process.

The only relatively long lived nuclides in significant quantities in the highly alkaline liquid waste from target plates dissolution are ^{90}Sr, ^{134}Cs and ^{137}Cs (Table XIV). Small quantities of other short lived nuclides are also present. Approximately 1 m^3 of alkaline waste is produced per year.

TABLE XIV. TYPICAL COMPOSITION OF AEC LIQUID WASTE

Chemical composition		**Radionuclide content after 6 months**	
Components	**Concentration**	**Nuclide**	**Activity, (MBq/L)**
Alkaline:		^{137}Cs	10 000–15 000
Al	40–60 g/L	^{95}Zr	2000–4000
NaOH	160–200 g/L	^{95}Nb	200–400
		^{103}Ru	150–300
Acidic:			
Li_2SO_4	50–75 g/L	^{95}Zr	50–150
HNO_3	0.5–0.7 M	^{95}Nb	5–15
H_2SO_4	0.03–0.04 M	^{103}Ru	4–8

Slightly acidic liquid waste is generated during ^{99}Mo purification. This is contaminated only with short lived nuclides such as ^{95}Zr, ^{95}Nb and ^{103}Ru. Less than 1 m^3 of this waste is produced per year.

At present the solid uranium residue which is retained during the alkaline dissolution process, is accumulated in stainless steel containers within hot cells. This residue contains the majority of the fission products formed during target plate irradiation, e.g. nuclides of Sr, Y, Zr, Nb, Ru, Sn, Sb, Te, the lanthanides, and small quantities of Pu. Consumable materials in the hot cell processes (approximately 1 m^3 per year) are contaminated with low levels of short -lived isotopes only.

4.3.5. Waste generation by ^{99}Mo production at IPPE, Russian Federation

A general overview of amount and chemical nature of waste generated during ^{99}Mo production at IPPE is presented in Table XV.

TABLE XV. WASTE FORMED UNDER IPPE TECHNOLOGY OF FISSION ^{99}Mo PRODUCTION DURING ONE YEAR (QUANTITY OF ^{99}Mo PRODUCED DURING ONE YEAR IS ABOUT 18–20 kCi AT THE DAY OF PROCESSING)

Waste type	**Main radioactive components of wastes**	**Quantity of waste**
Gaseous radioactive waste	^{131}I, ^{132}I, ^{133}I, ^{135}I, ^{133}Xe, ^{135}Xe	500 L
Aqueous radioactive waste	^{141}Ce, ^{143}Ce, ^{144}Ce/^{144}Pr, ^{137}Cs, ^{132}Te/^{132}I, ^{140}Ba/^{140}La, ^{131}I, ^{132}I, ^{133}I, ^{135}I, ^{103}Ru/^{106}Rh, ^{89}Sr, ^{90}Sr, ^{91}Sr, ^{90}Y, ^{91m}Y, ^{91}Y	100 L
Organic radioactive waste	UO_2^{2+}, ^{131}I, ^{132}I, ^{133}I, ^{135}I, ^{97}Zr/^{97}Nb, ^{95}Zr/^{95}Nb	40 L
Solid radioactive waste	Targets cladding, chromatographic columns	10 kg

Radioactive noble gases such as krypton and xenon isotopes (particularly ^{133}Xe) are completely released at the stages of cutting and dissolving of targets (see Fig. 8). Iodine isotopes (such as ^{131}I, ^{132}I, ^{133}I and others) are liberated through nearly the whole processing cycle up to the stage of evaporation to dryness. Iodine isotopes are also being grown from the decay of tellurium isotopes during storage of liquid wastes. Small amounts of aerosols are released when the ^{99}Mo recovery process is carried out.

All liquid wastes formed during the ^{99}Mo extraction process can be divided into two different parts — an aqueous waste and an organic waste. The general characterization of aqueous and organic wastes are shown in Table XVI.

The aqueous acid solution contains nearly the total amount of magnesium from the dissolved targets (about 40–50 g/L). The most part of the activity separation step is determined by relatively short lived radionuclides such as ^{143}Ce, ^{132}Te/^{132}I and others. After keeping for several months the total activity of waste is decreased significantly.

The organic solution contains uranium at a concentration of 30–40 g/L as its main valuable component.

The solid wastes consist of direct contaminated technologically removable parts of installations, e.g. the chromatographic columns, and as cumulative contaminated components of trapping and cleaning system of the radioisotope production facility, e.g. various filters. The inventory of main solid wastes of ^{99}Mo recovery process is given in Table XVII.

TABLE XVI. CHARACTERISTIC OF LIQUID WASTE FROM ^{99}Mo PRODUCTION AT IPPE

Type of waste	**According to which unit the waste volume is estimated**	**Waste volume**	**Waste type according to activity level**
Aqueous waste			
1. Aqueous phase from extraction	1 target	350 mL	HLW
2. Waste solution from organic phase rinsing	1 processing	3 × 200 mL	HLW
3. Condensate from evaporation of ^{99}Mo solution	1 processing	300 mL	ILW
4. Aqueous solution from extraction chromatography	1 processing	150 mL	ILW
5. Waste solution from column rinsing	1 processing	3 × 90 mL	ILW
6. Solutions from decontamination of the production equipment before starting the new batch processing	1 processing	2000 mL	ILW
Organic wastes			
Organic phase from extraction and re-extraction of ^{99}Mo	1 target	120 mL	HLW
Waste solution from rinsing of re-extraction phase with organic solution	1 processing	2 × 50 mL	ILW

4.4. ^{99}Mo PRODUCTION FROM LOW ENRICHED URANIUM

Use of LEU rather than HEU for ^{99}Mo production has several effects on radioactive waste generation:

(1) Increased amounts of irradiated uranium by-product that must be solidified, stored, and transported.
(2) Increased radioactive waste generation (spent chemicals and equipment, wipes, etc.) due to the processing of more uranium for the same ^{99}Mo yield.
(3) Increased amounts of Pu and other transuranic elements, which increase the cost and difficulty of disposal [27].

By comparing the data for the production of ^{99}Mo using HEU (93% enriched) with that using LEU (~20% enriched), almost 6 times the amount of uranium must be dissolved to produce the same amount of ^{99}Mo for the case of using LEU. The amount of dissolving solution used also increases by a factor of 3 to 6. As a result, larger process equipment is required.

TABLE. XVII. CHARACTERISTICS OF SOLID WASTES FROM ^{99}Mo PRODUCTION AT IPPE

Type of wastes	**According to which unit the waste volume is estimated**	**Waste amount**	**Waste type according to the activity level**
1. Extraction chromatography column	1 processing	1 piece (100 cm^3)	ILW
2. Stainless steel parts of targets	1 target	20–30 g (100 cm^3)	ILW
3. Filters from gaseous cleaning system of hot cells			
3.1. Ultrafine fibre glass	Change 3–4 times per year	10 kg (0.3 m^3)	ILW
3.2. Activated charcoal modified with Ag-salts	Change every 2 years	120 kg (4m^3)	ILW
3.3. Aerosol filters	Change 1 time per year	100 kg (8.5 m^3)	LLW
4. Activated charcoal filters from local gas cleaning system	Change every 10 processing runs	0,05 kg (0.15 m^3)	ILW

About 26 times more ^{239}Pu is produced in LEU targets than in HEU targets with comparable ^{99}Mo yield. As a result, the waste becomes more difficult for storage/disposal and disposal costs will increase significantly.

4.4.1. Waste from the Australian process

Three liquid waste streams are generated. The primary waste is an uranium-rich solution containing most of the fission products, the secondary waste is a combination of wash solutions, while the third waste stream is small in volume and contains the ammonia-rich condensate from evaporation of ^{99}Mo product (see Fig. 9). Until recently, the second and third waste streams were combined, but this practice has been stopped to reduce the concentration of ammonium nitrate in the secondary waste which complicates waste evaporation and solidification.

Annually, ANSTO generates about 100 L of primary and 250 L of secondary waste.

The major chemical constituents of the liquid waste are nitric acid, uranium, magnesium iron and ammonium ions. The radionuclide inventory is dominated by ^{137}Cs and ^{144}Ce, with significant quantities of ^{106}Ru, ^{155}Eu, ^{95}Zr, ^{95}Nb and ^{91}Y, as well as traces of ^{103}Ru, ^{141}Ce, ^{134}Cs and ^{60}Co (Table XVIII).

TABLE XVIII. TYPICAL COMPOSITION OF ANSTO INTERMEDIATE LEVEL LIQUID WASTE

Chemical composition		Radionuclide content	
Component	**Concentration**	**Nuclide**	**Activity (MBq/L)**
U	20–200 g/L	Ce-144	500–10000
Mg	<1 g/L	Ce-141	<10
Fe	<1 g/L	Cs-137	800–8000
NH_4^+	100–700 g/L	Cs-134	<10
HNO_3	0.6–0.8 M	Ru-106/Rh-106	50–1000
		Ru-103	<15
		Zr-95	10–200
		Nb-95	20–300
		Y-91	50–200
		Eu-155	10–100
		Co-60	<2

4.4.2. Waste from the Karlsruhe silicide process

Liquid waste from this process is similar in composition to that described in Section 3.2.3. for the HEU process presently used in the Netherlands (Mallinckrodt). However, the silicide process produces greater volumes of waste containing more acids and also oxidizing agents, and it also contains more plutonium due to the use of LEU. No specific details are available, as this process has only been carried out on pilot scale.

Gaseous and solid waste is similar to above-mentioned processes using HEU.

4.4.3. Waste generated from ^{99}Mo production using LEU silicide target developed at Argonne National Laboratory, USA

The production process using LEU silicide target is still under development, thus no operational information on waste generation is available. The potential waste streams and their compositions from the process have been estimated based on the proposed dissolution process discussed in Section 3.3.3 and are summarized in Table XIX.

4.5. PRODUCTION BY NEUTRON ACTIVATION

^{99}Mo produced by means of the (n, γ) route contains far less impurities than fission ^{99}Mo. Product purity depends primarily on the chemical purity of the raw material used in target preparation. Significant quantities of ^{186}Re, ^{188}Re, ^{188}W, ^{122}Sb, ^{124}Sn, ^{134}Cs, ^{60}Co, ^{65}Zn, ^{95}Zr, ^{95}Nb, ^{110m}Ag and ^{192}Ir have been detected in (n, γ) produced ^{99}Mo [2]. Further purification of this ^{99}Mo will create low or intermediate level liquid and solid waste containing these nuclides with half-lives up to a few years. To reduce waste volumes arising from this route of ^{99}Mo production, the use of high purity starting material is of the utmost importance. If this is achieved, the spent irradiation containers will constitute the only significant waste.

4.6. ACCELERATOR PRODUCTION

In principle this route will create only low-level radioactive waste. However, this remains to be shown experimentally.

TABLE XIX. POTENTIAL WASTE STREAMS AND THEIR COMPOSITION GENERATED BY THE PROPOSED PROCESS USING LEU SILICIDE TARGETS

Waste stream	**Composition**	**Main radionuclides**
Cladding solution (liquid)	NaOH, Al, Cr, Cu, Mg, Mn, Zn, Si	small amounts of fission products
Dissolving solution (liquid)	SO_4^{2-} (assuming H_2SO_4 is used for acidification)	fission products, Pu, Np
Slurry from the filter (solid)	OH precipitates in oxide and hydroxide forms	$Na_2U_2O_7$, Si, Np, Pu, and various fission products.
Alumina column (solid)	Al_2O_3	Mo, Tc
Cell waste (solid)	in-cell equipment	U, Pu, Np & fission products
Solid waste from off-gas treatment (solid)	e.g. charcoal, molecular sieve, air filter	Xe, Kr, ^{3}H, I

1. The use of accelerators as the source of neutrons would generate the same amount of waste as (n, γ) methods.
2. The use of accelerators for direct production of ^{99m}Tc or ^{99}Mo would result in moderating long lived radioactive impurities such as ^{90}Nb, ^{95m}Nb, ^{86}Zr, ^{87}Zr, ^{89}Zr, ^{87}Y.

4.7. WASTE RELATED TO ^{99}Mo PRODUCTION

4.7.1. ^{99m}Tc generator related waste

During the production of the ^{99m}Tc generators a small amount of liquid containing some ^{99}Tc is produced, which has to be treated as low level liquid waste. At the facilities where ^{99m}Tc generators are used, some liquid and solid wastes contaminated with ^{99}Tc are also produced.

Spent ^{99m}Tc generators are usually sent back to the producer where the generators are dismantled. The device with the decayed ^{99}Mo-source is removed and because of the presence of ^{99}Tc it is usually put into a drum and classified as low level solid waste. This drum is to be treated and conditioned as long lived LLW [34].

4.7.2. Waste from decommissioning of facilities for ^{99}Mo production

In several countries there is already some experience in dismantling and decommissioning of ^{99}Mo production facilities. It is always necessary to carefully plan the strategy of decommissioning. The following steps have to be executed:

1. Emptying the cells of all kinds of movable process components.
2. Inventory of the contamination level of each cell.
3. Determination of the dismantling strategy, i.e. hands-on or remote. This can be done by doing a cost evaluation of the two different methods by taking into consideration the actual available modern techniques.

4. Execution of the work. The following techniques can be used in order to minimize the waste production or to lower the contamination level:
   - rinsing with acids or other aggressive liquids to dissolve of the remaining crude in the vessels, pipes, evaporators and dissolver. It is important to know that there is an economic optimum from which the gain in decontamination factor (D.F.) is too low in comparison with the cost of treatment for the volume of liquid waste that is produced.
   - cutting or sawing the content of the cells in pieces and loading into drums
   - scaling or shaving the cell walls, collecting the dust into drums.

It is important that before starting the dismantling activities:

- all procedures are licensed by the responsible authority.
- all the produced waste can be stored or treated in licensed facilities.
- all materials and equipment should be controlled and monitored before leaving the facility.
- after complete decontamination there is a procedure to exempt from regulatory control.
- special care should be taken in respect to safeguards problems. All procedures and measurements during decommissioning must be documented and records must be kept for the period prescribed by the competent authority.

5. MANAGEMENT OF RADIOACTIVE WASTE FROM ^{99}Mo PRODUCTION

5.1. INTRODUCTION

In the planning, design, construction and operational phase of a ^{99}Mo production facility the waste management concept and implementation plays an important role. Within the licensing procedure the strategy for waste management, including the route for the spent targets and long lived fission products has to be described and approved by the responsible authorities. Other important issues such as criticality of U and decay heat generated by the radionuclides should be taken into consideration.

Waste will be generated as solids, liquids, and/or gases, and will include material in the low, intermediate and even highly radioactive categories. Initial treatment of waste streams is usually required at the production site, prior to short or long term storage. The treatment required is dictated by both the form of the waste and its activity level. This technology is established, and generally available in countries with an existing nuclear industry. In some instances storage facilities may need to be constructed. Final disposal of waste should also be considered right from the beginning of process development.

5.2. ON-SITE PROCESSING OF RADIOACTIVE WASTE FROM ^{99}Mo PRODUCTION

On-site treatment refers to waste treatment carried out in the production facilities while off-site treatment is performed outside the facility (at the same site or elsewhere).

5.2.1. On-site treatment of ^{99}Mo waste at CRL, Canada

The only on-site treatment of ^{99}Mo waste (i.e. in the ^{99}Mo production facility) at CRL is in-cell off-gas treatment. The process off-gas from the dissolver is scrubbed with nitric acid and distilled water. The gas is then collected into a decay system where noble gases are trapped for a period of time prior to release to the cell ventilation system. Radioactive krypton and xenon are trapped in a series of decay columns to allow Kr and Xe to decay.

The cell ventilation system which consists of a series of HEPA and charcoal filters, is designed to retain particulate material and isotopes of iodine, and thus minimize the release of radioactivity to the environment.

Two radiation monitors for noble gases are installed in the exhaust system after the filters. The combined effluent air is also monitored for ^{131}I and β particulate. The average levels of noble gases, ^{131}I and β particulate as ^{137}Cs released from the production facility are 8.0 TBq, 25 MBq and 0.5 MBq per week and they are much lower than the allowable weekly derived release limits of 1600 TBq, 210 GBq and 120 000 MBq, respectively [35].

5.2.2. On-site waste treatment at the IRE, Belgium

Gaseous waste

The released xenon is carried by helium towards the condensation trap, where it is condensed on copper clippings cooled down to –196°C by means of liquid nitrogen. Further treatment of the gases after this first trap takes place. The gas is then routed to an active carbon trap which is also cooled, preventing discharges of xenon into the ventilation system. The trapped xenon is subsequently transferred in a container and shipped to a purification plant. This last step consists of chromatographic separation from the carbon column. It aims at eliminating CO_2, O_2, N_2O, etc. and concentrating of xenon. The released iodine is trapped on platinum asbestos and is processed in order to recover ^{131}I.

Liquid waste

For decay of ^{131}I and other short lived fission products, the alkaline solution is stored in stainless steel tanks for about 1 year. To avoid Al-precipitation the alkaline solution is adjusted with 6M NaOH and limited to 40g/L Al. The acidic waste stream consists of 2M sulphuric acid and the remaining fission products. The transport is made using TNB-165 transport container of Transnubel.

Solid waste

The waste is collected in the hot cells, segregated and partially loaded in 15 L cans. The compactable waste is precompacted in the cell and loaded in cans or racks. Depending on the dose rate (up to 1 Sv) the boxes are stored for a decay period of several months or years. They are stored till their dose rate becomes lower than 2 mSv/h or between 2 mSv/h and 200 mSv/h. This is due to the NIRAS/ONDRAF specifications for treatment and conditioning of low level and medium level solid waste in the installations of Belgoprocess in Dessel.

- Waste with a dose rate between 2 mSv/h and 200 mSv/h is loaded in 200 L drums (for supercompaction) or in 1 m^3 containers (for incineration) for transport to the treatment installations at Belgoprocess.
- The waste with a dose rate >200 mSv/h and a higher decay period are still in storage and waiting for a technical solution.

5.2.3. On-site treatment of waste from fission ^{99}Mo production at Mallinckrodt/ECN, Netherlands

Gaseous waste

The hydrogen is oxidized to H_2O via CuO at 350–400°C and the resulting water is condensed together with ^{3}H. Xenon is collected together with nitrogen in the pre-evacuated stainless steel tank. The gas is then sent to the xenon delay system which consists of carbon filters. To guarantee a safe, leakage-free process all equipment used in this extremely hot part of the process is made of stainless steel.

Liquid waste

To avoid Al-precipitation the alkaline solution is adjusted with 6M NaOH and limited to 40g/L Al. For decay of ^{131}I and other short lived fission products the alkaline solution is stored in stainless steel tanks for about 6 months. The acidic waste stream consists of 2M sulphuric acid and the remaining fission products.

After this storage period (6 months) the liquids are pumped into specially designed liquid holders of 44 L, which are filled until the activity reaches the A-2 transport requirements [36]. The liquid holders are placed in a shielded type A transport container for liquids for shipment to the Central Organization for Radioactive Waste (COVRA) in the Netherlands.

The other low level liquid waste streams are stored in buffer tanks for a period of several months. Thereafter the liquid is pumped in standardized 30 liters liquid waste drums and transported unshielded to COVRA.

Solid waste

The waste is collected within the hot cells and loaded into small waste boxes and racks. Spent columns are stacked for a decay-period of several months within the cell. Other waste is put in so called Syntacs-containers with a diameter of 210 mm and a length of 650 mm. For replacement of bigger components a Syntacsbus-container of 295/690 is available. These containers with waste are packed in shielded type A transport containers for direct shipment to COVRA.

The material of the spent target is sent back to the producer of the targets. It is possible that in the future waste from the reprocessing will be sent back to the user.

5.2.4. On-site processing of waste from fission ^{99}Mo production in South Africa

Gaseous waste

Dissolution of irradiated target plates and purification of ^{99}Mo are carried out in lead-shielded alpha-tight hot cells. The xenon which is released during plate dissolution is adsorbed onto standard coconut shell-based activated carbon in stainless steel columns. These columns are then closed off for a period long enough to allow decay of all Xe isotopes. Nuclides of krypton are not retained completely. Small amounts of gaseous iodine are removed from the hot cell off-gas by using suitably impregnated activated carbon. Complete back-up capacity for all filtration systems is available.

Hydrogen which is evolved during target plate dissolution is either oxidized by hot copper oxide, or the dissolution rate is controlled in such a way that the hydrogen concentration in the ventilation system is within safety limits. Monitoring systems for noble gases, iodine, α and β/γ-emitters as well as hydrogen are distributed throughout the ventilation system for process control measures as well as the quantification of releases to the environment.

Liquid waste

Alkaline liquid waste is accumulated in shielded stainless steel tanks and then immobilized in a cement-vermiculite mixture by means of a vacuum intrusion technique. This entails the evacuation of a drum filled with the cement mixture and then using the vacuum to suck the solution from the tank into the drum. Before cementation, the drum is placed into a high density concrete container which provides the necessary radiation shielding during handling and transport to the Vaalputs waste repository for low and medium active waste for shallow land burial.

Slightly acidic liquid waste solutions are accumulated and left to decay to levels where they can be transferred to the Waste Management Department (by underground pipeline). There the solutions are further treated and conditioned, sometimes together with low-active waste from other operations.

Future processing of the uranium-containing residues to recover and purify uranium for re-use can be done with an alkaline process using ion exchangers and/or an acidic purification route similar to the Purex process. Alkaline liquids or neutralized acids are immobilized in the same way as the alkaline waste mentioned above. Alternatively, it can be immobilized in a mixture of Portland cement and blast furnace slag in stainless steel containers within a hot cell.

Solid waste

Consumable materials used in the hot cell processes are contaminated with low levels of short lived isotopes only. These materials are left to decay in hot cells or elsewhere in the facility to radiation levels where it can be transferred to the Waste Management Department for further processing. Plant materials such as ventilation filters are handled in a similar way.

If contamination of solid waste by Cs nuclides is suspected, this waste is segregated and handled separately due to the need for more shielding. Partitioning of other waste according to contamination levels or radionuclide content is practiced wherever possible.

5.2.5. On-site treatment of waste from fission ^{99}Mo production at IPPE, Russian Federation

In general all the wastes generated by the ^{99}Mo and ^{99m}Tc generator production processes at IPPE are trapped, treated and stored on site. Therefore, no transportation of the wastes out of IPPE is required.

Gaseous waste

The main gaseous and aerosol waste released during the processing of short-cooled irradiated targets are noble gases (mainly xenon) and iodine radionuclides. To avoid their uncontrolled release into the in-cell area, all process steps of ^{99}Mo production are carried out in hermetically constructed devices. Xenon and other noble gases released during dissolution step are trapped into pre-evacuated stainless steel tanks. Additional trapping of noble gases is done by cryogenic adsorption (at the temperature of liquid nitrogen) on activated charcoal (see Figs 9 and 14). Iodine is trapped by alkaline absorption and adsorption on silver coated

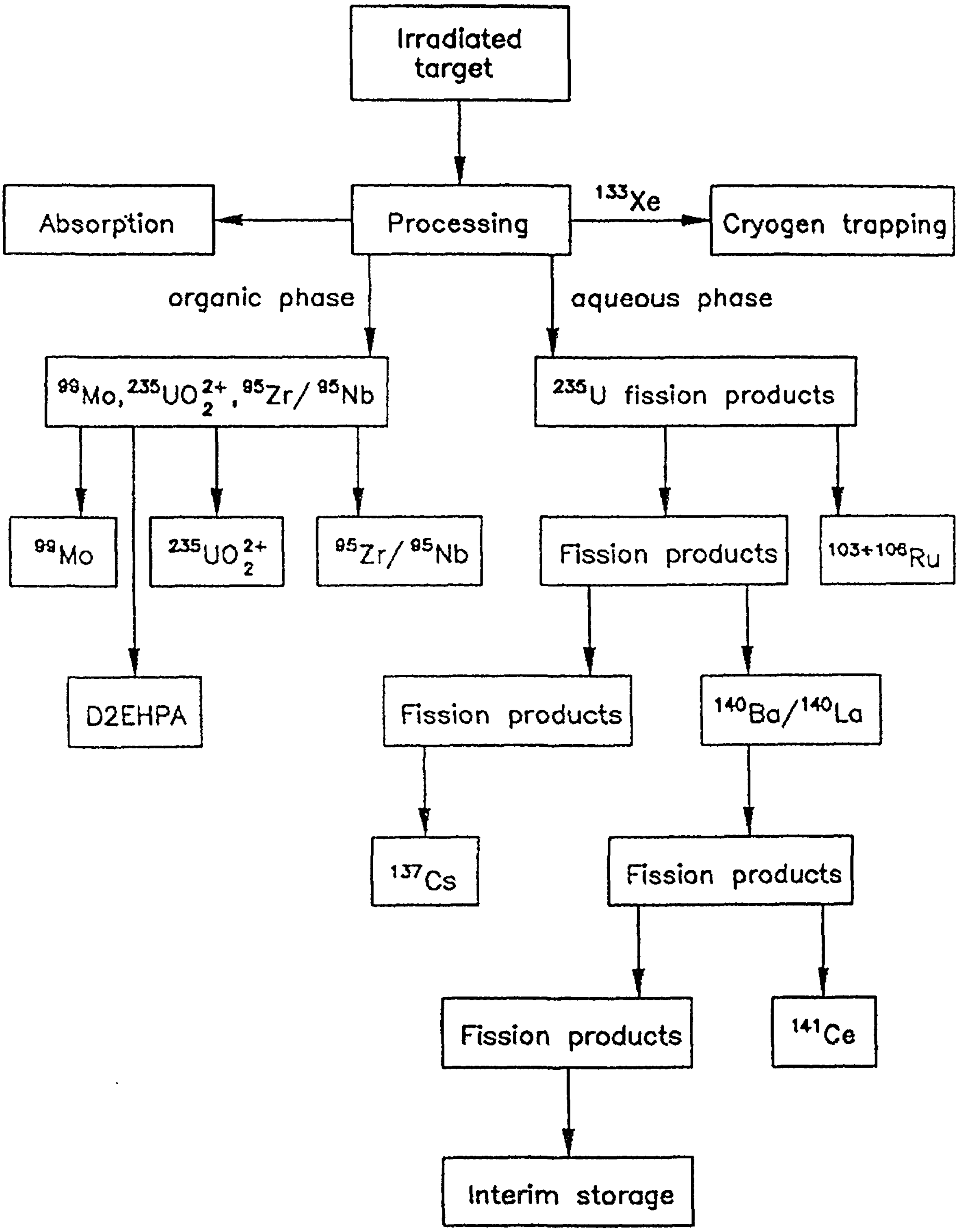


Fig. 14. Materials and waste flow diagram of ^{99}Mo recovery process at IPPE, Russian Federation.

charcoal. To prevent release of iodine and β particulate aerosol to the environment additional absolute filters (or aerosol filters) are used. After trapping the radionuclides of xenon and iodine, they are stored in a cell for 6 months to allow for decay. After this time the holding tanks are emptied through the ventilation system. Iodine and xenon can be additionally purified and sent to customers upon request.

The isotope production facility is equipped with several types of measuring monitors for noble gases, ^{131}I and β particulate located after filters and in the exhaust system. The measurements are carried out in a continuous mode. It has never exceeded the approved strict limits implemented by the regulatory body for the radioactive gases and aerosol release to the environment from the start of ^{99}Mo production up to now.

Liquid waste

During the extraction and re-extraction steps high level aqueous and organic wastes are formed. After the processing these are stored separately in special vessels in a cell for 2 months to permit decay of short lived radionuclides. The aqueous solution is then collected in a high level aqueous waste stainless steel tank for interim storage and future reprocessing. The organic solution is collected in a critically safe vessel for future separation of ^{235}U. The saleable radioisotopes contained in those solutions (for example, ^{137}Cs, ^{106}Ru, ^{140}Ba/^{140}La, ^{95}Zr/^{95}Nb, etc.) can be extracted on request (see Fig. 14). The contents of the tank is monitored on a regular basis. Table XX gives the results of such measurements. The composition of the aqueous waste is typical not only for ^{99}Mo production; it shows the integral result (mix) of other kinds of activities which were carried out at the IPPE isotope production facility over a period of several decades.

TABLE XX. THE CONTENT OF HLW INTERIM STORAGE TANK (IPPE, Russian Federation)

Uranium content (sum of isotopes) – 480 mg/L; Nitric acid concentration – 0.8 mol/L;
Am and Pu concentration – about 6.5 mg/L; Solution volume – approximately 930 L.
List of radionuclide impurities:

Radionuclide	T2, days	Content, Ci/L
^{155}Eu	4.7 years	0.075
^{154}Eu	8.6 years	0.011
^{141}Ce	32.5	0.242
^{144}Ce/^{144}Pr	285	1.02
^{140}Ba/^{140}La	12.8	0.206
^{103}Ru	39.3	0.137
^{106}Ru/^{106}Rh	374	0.137
^{137}Cs	30.2 year	0.825
^{134}Cs	2.07 year	0.055
^{95}Zr/^{95}Nb	64	0.025
sum of γ-emitters	-	2.5
sum of β-emitters	-	6.8
sum of α–emitters	-	1.8-10.3

Intermediate level aqueous waste formed during other process steps of ^{99}Mo production are collected in other stainless steel tanks for storage for 2 years, followed by transfer to the IPPE waste processing facility for concentration and further interim storage. To avoid the precipitation of magnesium salts and for preventing of the tank material corrosion waste solutions are stored in acid form (without neutralization).

Solid waste

Solid waste includes target cladding, used chromatographic columns, and several types of filter materials from the off gas trapping system and other consumable materials used in the hot cell process. The targets cladding and used chromatographic columns are packed into 10 L cans and they are stored in a special concrete-shielded shallow land burial located under the IPPE isotope production facility. The filters are disposed of in the same method without containerization.

5.2.6. Interim storage and proposed conditioning of waste from ^{99}Mo production in Australia

The primary liquid waste received from ^{99}Mo production facility is collected in concrete-shielded, stainless steel tanks each having a capacity of 1100 liters. The secondary waste of lower activity is accumulated in glass lined steel tanks (also shielded) each with a capacity of 2200 liters. In total, ANSTO has about 6000 liters of these liquid wastes.

Volume reduction of the waste solutions by means of evaporation to a dry solid (predominantly uranium nitrate hexahydrate) has been considered, but the presence of ammonium nitrate in the waste constitutes a safety hazard. Research is in progress on a process to destroy the ammonium ion prior to evaporation.

In the longer term, it is proposed that the waste be converted to a leach-resistant form. Two options are under consideration i.e. incorporation into cement or a ceramic waste form (such as Synroc).

5.3. OFF-SITE PROCESSING OF ^{99}Mo PRODUCTION WASTE

5.3.1. Management of ^{99}Mo production waste at CRL, Canada

In general, all the wastes generated by the ^{99}Mo production process at CRL are treated and stored outside the production facility at CRL site. Therefore, no transportation of the wastes is required, however this treatment is considered as an off-site.

Liquid waste

The fissile liquid waste generated by the ^{99}Mo production process is temporarily stored in a 24 m^3 storage tank. The uranium level in the solution is continuously monitored. A facility is planned to recover the enriched uranium using the PUREX process. The waste from the uranium recovery process is expected to be vitrified for final disposal. Research work has been performed to define vitrification process. An alternative to vitrification is extraction of saleable radioisotopes such as ^{137}Cs, ^{90}Sr, ^{144}Ce, ^{147}Pm and ^{106}Ru. The residual waste would be suitable for bituminization in the existing facility at CRL.

The intermediate level non-fissile liquid waste is temporarily stored in a storage tank and the waste is planned to be solidified using cement or bitumen depending on the chemical and radioactive composition of the waste at the time of solidification.

The low level liquid waste from the facility is discharged to the existing active liquid drainage system and treated in the CRL Waste Treatment Center. The treatment process involves microfiltration, reverse osmosis, evaporation and bituminization. The bituminized waste is stored in 210 L drums and stored in either the bunkers or the tile-holes in the WMA at CRL.

Solid waste

The intermediate level wastes from the hot cell are packed in 20-liter containers and the containers are put in the top-loaded, bottom-release type shielded flasks for shipment. The flasks are shipped to the CRL Waste Management Areas (WMA) and the 20 liters containers are placed in specially built underground concrete cylinders (tile holes) for storage. These wastes are labeled using a bar-code system and can be removed from the tile holes for final treatment and disposal.

The low level solid wastes, such as out-side hot cell waste and laboratory waste are collected in plastic bags and shipped to the CRL Waste Treatment Center for compaction. These wastes are combined with other wastes generated at CRL. The compacted wastes (bales) are transferred to the WMA for storage in large circular concrete bunkers located underground.

The spent molecular sieves and activated carbon are transferred into 210 L drums and stored in the CRL WMA. The HEPA filters are also stored in the WMA after a decay period.

5.3.2. Off-site processing of waste from the IRE, Belgium

Liquid waste

In the early 1980s the ILLW (intermediate liquid level waste up to 1 Ci/L gross β) from IRE was sent to the waste treatment facilities of the CEN Mol. After a temporary cooldown period the basic ILLW streams were mixed up with other hot waste streams (up to 1 mCi/L gross β) and treated by the flocculation technique. The residual sludges were incorporated into distilled bitumen Mexphalt 20–30 at the "Mummy" installation. Characteristics of this bitumen are presented in Table XXI.

TABLE XXI. CHARACTERISTICS OF BITUMEN MEXPHALT 20/30 USED IN "MUMMY" ON SITE 2 OF BELGOPROCESS

	Limit values
Penetration value (10^{-1} mm) (at 25°C)	20–30
Softening point (°C)	59–69
Density	1.02–1.07
Flash point (°C min) (Cleveland Open Cup)	>275
Solubility in Tri (wt%)	>99
Elasticity (cm)	>30
Loss on heating (ASTM D-6) (%wt max)	>0.2
Drop in Penetration after Heating (% max)	165
Radiation resistance (rad)	$>10^{10}$

The “Mummy” installation is a batch process. The sludges are continuously introduced into a known volume of molten bitumen with the mixture maintained at temperatures ranging from 180 to 200°C. The water is evaporated and the remaining solid particles are incorporated into the bitumen. After a predetermined amount of waste has been added to achieve the required waste form composition, the feed is stopped. The process continues until the residual water has been evaporated and then the mixture is discharged into 400 L drums and allowed to cool. Before entering the installation, the blended sludge undergoes a freeze/thaw cycles. This cycles is necessary for obtaining a better filtration capacity.

Since 1985 all ILLW from IRE has been received at the waste facilities of Belgoprocess. Part of these waste streams, mixed up with other ILLW solutions are treated and incorporated in bitumen (Mexphalt 85–40) in the Eurobitum installation. Characteristics of this bitumen is presented in Table XXII.

TABLE XXII. CHARACTERISTICS OF BLOWN BITUMEN MEXPHALT 85/40 USED FOR THE BITUMINIZATION OF INTERMEDIATE LEVEL LIQUID WASTE AT THE EUROBITUM PLANT

Penetration at 25°C(10^{-1} mm)	35–45
Softening point (°C)	80–90
Density at 25°C (g/cm^3)	1.01–1.05
Weight loss during 5 h at 163°C (%) (ASTM D-6)	0.2
Flash point COC (°C min)	240
Solubility in CS_2 (%)	>99
Ductibility at 25°C (cm)	>3
Average pumping temperature (°C)	165
Radiation resistance (rad)	$>10^{10}$

A reception station is equipped for the reception of the alkaline waste stream solutions. The solutions are pumped into a reception vessel and transferred to either the storage vessel or to the Eurobitum facility for incorporation into bitumen.

The ILLW waste is at first analyzed as a control step and in view of the concentration step which is foreseen in the NCP (Nuclear Chemical Plant)-evaporator (Step 1). After the concentration step, the concentrate is sent to the Eurobitumen facility.

Another concentration step takes place in a reaction vessel (Step 2). This concentration step has a triple aim, i.e. reducing the volume to reach a final salt concentration of $\leq$40 wt% with a maximum of 350 g/L $NaNO_3$, eliminating the eventual presence of NH_3 to exclude any explosion risk during the incorporation step, and finally reducing the exothermic behavior of the sludges.

After the evaporation step, depending on the presence of some isotopes, addition of chemicals take place. On the final slurry batch DTA/DSC (differential thermal analysis/differential scanning calorimetric) analysis is carried out to ensure that the end product is within safe limits.

The bituminization of the slurry results in an extruder product (Step 3) which contains 40 wt% salts and 60 wt% bitumen with a β/γ activity of 9.15 10^9 Bq/L (max <3.7 10^{10} Bq/L) and an alpha activity of $1.13 \cdot 10^7$ Bq/L (max $Pu^{235+240}$ $<1.11 \cdot 10^8$ Bq/L). These maximum limits are established to not exceed the radiation limits of $9 \cdot 10^{10}$ rad during 1000 years of storage. Typical composition in the product streams at different steps are summarized in Table XXIII.

TABLE XXIII. CHEMICAL PRETREATMENT AND BITUMINIZATION STREAM DIAGRAM OF THE ILLW FROM FLEURUS

Step	**1**	**2**	**3**
Bitumen wt%			60
Volume L	1350	691	
Density kg/L	1.14	1.28	1.26
Weight kg	1539	881	
Tot. salts g/L	261	509.91	40 [wt%]
Tot.salts wt%		39.99	
Act.gross beta Bq/L	4.77E+09	9.32E+09	9.15E+09
Act.gross alpha Bq/L	5.91E+06	1.15E+07	1.13E+07
pH	8–9	8–9	
Temperature °C		25	185

Step 1: chemical pretreatment: input.
Step 2: chemical pretreatment: output for the bituminization.
Step 3: extruder product.

After processing, this waste is stored in a specific bunker at the Belgoprocess site. The packaging is carried out in accordance with the quality assurance handbook which is confirmed to the IAEA Code of Practice 50-C-QA [37]. To guarantee the final quality of the conditioned waste package (particular care is taken to strictly observe the operational manual and to execute during production the specific controls and tests). Final disposal in deep underground clay is foreseen.

End product specifications of bituminized waste is presented in Table XXIV.

Solid waste

Low and intermediate level solid wastes from the IRE are sent to the Cilva plant at Belgoprocess for treatment.

The filters are precompacted and put in 200 L drums; the 200 L drums are sent to supercompaction and the resulting pellets are loaded in 400 L drums for cementation. The general practices for solid waste management at Belgoprocess are:

- The 200 L drums are supercompacted, the pellets loaded in 400 L drums and cemented.

TABLE XXIV. END PRODUCT SPECIFICATIONS OF BITUMINIZED WASTE PRODUCTS AT BELGOPROCESS

Installation	EUROBITUM	MUMMIE	Unit
Drum drawing nr.	3-E-21968b	2201 DE-001A	
Volume in BWPdrum	180	400	dm^3
Density BWP	1.33	1.33	kg/ dm^3
Weight BWP in drum	230	520	kg
Maximum drum weight	400	600	kg
Contact dose rate drum	< 2	< 0.02	Sv/h
Alpha surface contamination	< 3.7	< 3.7	kBq/m^2 drum
Beta surface contamination	< 37	< 37	kBq/ m^2 drum
Spec.Beta-gamma activity	< 3.7 E+10	< 3.7 E+08	Bq/ dm^3
Spec.Pu239+Pu240 activity	< 1.1 E+08	< 3.7 E+06	Bq/kg
Bitumen type	Mexphalt R 85/40 blown bitumen	Mexphalt 20/30 Distilled bitumen	
Salt/bitumenratio in BWP	40/60	40/60	
Water content in BWP	< 1	< 0.5	%
Self ignition point	> 300	>300	°C
H_2-radiolyse production/drum	< 25	< 0.6	cm^3/day
Alpha leachrate in H_2O	< 1 E -06	< 6 E-09	g/ cm^2/day
Beta leachrate in H_2O	< 2 E-04	< 4 E-07	g/ cm^2/day
Max.integrated dose (∞)	< 1 E+10	< 3 E+08	Rad
Max.integrated dose (1000Y)	< 1 E+09	< 3 E+07	Rad
Porosity at 1 G Rad	< 3	< 3	%
$NaNO_3$ in BWP	25	< 0.5	W%

- The 200 L drums with non compactible waste are loaded in 400 L drums for cementation.
- The burnable waste is sent to the incinerator. The resulting ashes are collected in 200 L drums which are supercompacted, loaded in 400 L drums and cemented.
- The conditioned waste is brought into an interim storage building at the site of Belgoprocess. Final disposal by shallow land burial will be realized as soon the site is prepared.
- To guarantee the quality of the process the whole process is covered by a ISO-9001 certificate.

For the treatment of La Celeine containers filled with medium and high level waste the following options are available. Containers with a dose rate <0.2 Sv/h are compacted and loaded in 400 L drums for cementation. Containers with α dose rate >0.2 Sv/h can be loaded either in 400 L, 200 L drums or in glass canisters (type La Hague) for cementation.

5.3.3. Off-site processing of waste from fission ^{99}Mo production at COVRA, Netherlands

The shipment of the waste from ECN/Mallinckrodt site to the site of COVRA takes place by road using a COVRA-owned truck according to IAEA recommendations for transport of radioactive materials [36]. The first transport with solid waste took place in 1996 and the first liquid waste transportation in 1997.

Liquid waste

The low level liquid wastes from ECN/Mallinckrodt are treated at COVRA together with other LLW liquids. The activity is concentrated by means of chemical coagulation and co-precipitation [38]. The precipitates are separated and conditioned with cement. After control of the activity, the purified water is discharged into a nearby river. The liquids with the higher activity are first handled in a dedicated reception-installation for emptying the liquid holder. This liquid is then stored in 2 stainless steel buffer tanks of 3000 L awaiting immobilization by cementation in 200 L drums with a mobile cementation unit, which will take place in the waste treatment building. The maximum activity within the waste packages should comply to the A-2 transport requirements. The 200 L drums will be placed in a 1000 L concrete overpack.

Solid waste

For the treatment of filled Syntacs containers the following routes are available. Syntacs-containers with a dose rate above 10 mSv/h on surface will be stored in a shielded interim storage bunker for a period of approximately two years [38]. Depending on the dose rate of the Syntacs' containers from the interim storage bunker after two years of storage the containers will be placed and embedded in cement directly in 200 L drums or will follow the supercompaction route.

The Syntacs-containers with a dose rate below 10 mSv/h on the surface will be pre-pressed and put in a standard 100 L drum for solid waste. The 100 L drums will be treated by supercompaction whereafter the resulting parcels are embedded with cement in 200 L drums. The conditioned waste packages are placed in an interim storage building for a period of 100 years [38].

5.3.4. Waste conditioning and interim storage at the AEC, South Africa

Liquid waste

Low active liquid waste is normally treated chemically (e.g. precipitation) to remove any possible hazardous (inactive) elements. It is then discharged according to licensing requirements and various other regulatory and legislative conditions. If required, for example if the discharge conditions regarding radionuclide content cannot be met, LLW is concentrated up by evaporation, and the concentrate immobilized in cement.

Solid waste

If the radiation levels of solid waste received from the ^{99}Mo production facility is too high to handle without shielding, it is stored in concrete containers for up to two years to allow for the short lived nuclides to decay. It is then compacted, the compacted waste is put into drums, the empty space in the drums filled with cement and it is then disposed of by shallow land burial.

A licensed pipe-storage facility for the interim storage of small steel containers (8 liters capacity) is available at Pelindaba. This can be used for storage of uranium residue or small volumes of other waste which cannot be handled in the short term due to high radiation levels. In this way, hot cells are not used for the storage of waste.

5.3.5. Waste management and interim storage at IPPE, Russian Federation

Liquid waste

After two years of storage, low and intermediate active aqueous wastes is transferred from the IPPE isotope production facility to the IPPE waste treatment facility by underground pipeline. The aqueous waste is concentrated by an evaporation process. The factor of aqueous waste volume reduction is about 300–400.

The concentrate with the content of mineral salts about 600 g/L^{-1} are pumped to the special designed concrete-shielded stainless steel tanks having a capacity of 125–300 m^3 for further interim storage. After installation of a special cementation device these aqueous concentrates will be solidified and disposed of at a shallow land burial site located not far from IPPE waste treatment facility. The initially purified condensate is passed through the series of ion-exchange columns for final treatment. After monitoring for contents of radioactive impurities it is released to the nearest river.

Solid waste

Low and intermediate level solid waste from ^{99m}Tc generators production is packed into 200 L drums. Then they are compacted and transferred into concrete shielded steel tanks for solid waste interim storage.

5.4. TREATMENT OF WASTE RELATED TO ^{99}Mo PRODUCTION

5.4.1. ^{99m}Tc generator production and spent generators

The solutions with ^{99}Tc are treated as low level liquid waste. From the spent generators the columns containing ^{99}Mo are treated as low level solid waste and follow the route of supercompaction and cementation.

5.4.2. Waste resulting from decommissioning

The range of waste from decommissioning of ^{99}Mo production facilities depends on the scale/capacity of the facility and its specific features. As for many others nuclear facilities most of waste from decommissioning of ^{99}Mo production facility will belong to low and intermediate level categories. Waste from decommissioning will be sorted out and will follow the route for treatment, conditioning and disposal that is available for this kind of waste. More information on management of waste from decommissioning and decontamination could be found in Refs [39–41].

5.5. REQUIREMENTS FOR WASTE DISPOSAL

Conditioning of waste have to be done in such a way that requirements for transport, interim storage and final disposal will be fulfilled [36, 42–45]. In some countries with a large nuclear power programme the criteria for waste disposal and capacities for to fulfill these criteria already exist. In other countries these criteria are still being formulated. The main characteristics of solidified waste forms to be considered when developing the waste acceptance criteria for solidified waste are summarized below:

- For cemented waste the main criteria for the waste matrix are:
  - homogeneity;
  - strength;
  - water resistance;
  - leach resistance.
- For bituminized waste the main criteria for the waste matrix are:
  - homogeneity;
  - density;
  - mechanical properties:
    - hardness;
    - softening point;
  - chemical and physico-chemical properties:
    - $NaNO_3$ loading;
    - flammability (flashpoint and self ignition point);
    - radionuclide release by leaching (e.g. ISO/TC85/SC5/WG5N22);
    - swelling;
    - heat production;
    - resistance to irradiation (total integrated dose <E7 Gy);
    - water content;
    - filling rate.

Requirements for disposal of radioactive wastes generated during ^{99}Mo production are not specific for this particular waste types. In general this requirements depends not on waste source, but are defined by available disposal option. These requirements and options are described and discussed in detail in many publications [46–48].

5.6. QUALITY ASSURANCE, LICENSING AND SAFEGUARDS

Due to stringent purity specifications for ^{99}Mo, production processes are normally carried out under a quality code of practice such as ISO 9000, ASME NQA-1 or specific national requirements [3, 4]. Example of these requirements is presented and discussed in Section 2.6 of this report.

From a waste management point of view it is imperative that all related practices are also covered by the same quality assurance system. This will ensure that control over nuclear and radiation safety aspects of all waste management processes, as well as the recording of data regarding the composition of waste streams and packages, will be carried out properly. In accordance with the international practice regulatory organizations in many countries are continuously enforcing more strict requirements concerning the management of radioactive waste. A comprehensive quality assurance system should be one of the licencing requirements as the only mechanism to ensure compliance to these requirements. More specific the subject of quality assurance for different waste management practices is covered in special reports [49– 52].

When producing ^{99}Mo by fission of HEU safeguards requirements must be fulfilled by very strict material accountability measures. Aspects such as the keeping of records, traceability, etc., as prescribed by nuclear materials safeguards agreements, are closely related to quality assurance. For these reasons it is imperative that ^{99}Mo production as well as management of the resultant waste should be supported by a proper quality assurance approach.

5.7. WASTE MINIMIZATION AS A STRATEGY FOR ^{99}Mo PRODUCTION

The concept of waste minimization should be considered already at the stage of planning, design and licensing of ^{99}Mo production facilities. Special care has to be taken to all aspects and all stages of waste management, starting from source of generation, proper collection and segregation, interim storage, treatment, conditioning, transportation, long term storage and/or disposal. During the design stage of the plant future decommissioning plans and associated waste generation have to be taken into consideration.

During ^{99}Mo production, segregation of the waste at the source of generation based on their chemical, physical and radiochemical composition is very important because it can have a strong influence on the volume of waste for further processing and in many cases on selection of the processing technologies and all other stages of waste management [53]. Proper implementation of the segregation of radioactive waste leads to simpler waste treatment methods, less complicated facilities and reduces risk of incidents.

Another important aspect of waste minimization is a prevention of unreasonable spread of contamination. This very much depends on general waste management culture, appropriate design of the facility, technology selection and operational experience of the personnel.

Recycling and reuse of valuable radioactive and non-radioactive materials and components from the potential waste streams is another important option of waste minimization. It is very important during ^{99}Mo production to consider thoroughly all possibilities of recovery other radionuclides which may have commercial value for medical, industrial or research application. Recycling of non-radioactive components of different waste streams, like recovery and recycle of solvents, acids and other process components may substantially reduce the volume of waste to be treated, stored and disposed of. In the case of fission ^{99}Mo production it is especially a challenge to recycle of enriched uranium, which is an expensive raw material for the process.

Minimization of radioactive waste leads mainly to considerable cost reduction and, more importantly, to the reduction of radiation doses. It is practical to minimize:

- the activity content of different waste streams, e.g. gaseous, liquids and solid (this can be reached also by optimization of irradiation time to reduce generation of fission products);
- the volumes of different waste streams;
- the amount of material classified as radioactive.

Finally, total waste volumes may be substantially reduced during processing of waste. Selection of proper waste management strategy and waste processing technologies, both on- and off site, very much define the final quantity and quality of waste packages prepared for long term storage or disposal. Possible processing options and approaches are discussed in Sections 5.2 and 5.3 of the report.

6. CONCLUSIONS AND RECOMMENDATIONS

6.1. GENERAL

Radioactive waste from the production of ^{99}Mo, which is the most important radionuclide for medical application, must be managed in such a way that the protection of people and the environment is ensured, now and in the future.

The volumes and types of radioactive wastes generated by ^{99}Mo production are largely dependent on the production method. Most methods used for ^{99}Mo production are based on nuclear fission of targets containing ^{235}U. Processing of these targets generates considerable amounts of radioactive wastes containing long lived fission products and $\propto$-emitters. The presence of fission products and actinides in solid and liquid radioactive wastes complicates all stages of waste management, especially in countries without existing waste management infrastructure. The treatment and conditioning of fission ^{99}Mo production wastes require more progressive technology and equipment, hot cells remote handling, shielded waste packages, and capacity for interim storage. The waste will be generated as solids, liquids, and/or gases, and will include material in the low, intermediate and even high level radioactive categories. Initial treatment of waste streams is usually required at the production site, prior to short or long term storage. The treatment required is dictated by both the form of the waste and its activity level. This technology is established, and generally available in countries with an existing nuclear industry. In some instances storage facilities may need to be constructed. Final disposal of waste should be considered right from the beginning of process development.

Only small amounts of radioactive waste are generated from ^{99}Mo production by means of neutron activation of ^{98}Mo targets, but this method is only applied on a small scale.

The new ^{99}Mo production ways using an aqueous homogeneous nuclear reactor and/or accelerators of charged particles are considered at the early research stage and there is no reliable information about the radioactive waste production associated with use of these technologies.

6.2. STATUS OF THE WASTE MANAGEMENT ACTIVITIES

Although the volume and composition of waste produced during the production of ^{99}Mo might be regarded as less serious than waste arising from other nuclear operations, this does not mean that waste can be regarded as easy to handle, process and dispose.

Countries operating nuclear power plants or having an extensive programme in the nuclear operations (e.g. reprocessing of spent fuel elements), should not experience difficulties with the management of radioactive waste generated from the processing of irradiated HEU or LEU targets for ^{99}Mo production. In these countries a complete system exists for the safe management of solid and liquid radioactive wastes, from the segregation of waste types to collection, transport, treatment, conditioning, interim storage and final disposal. These facilities could be (or are) used for processing of waste generated during ^{99}Mo production.

The treatment and elimination of gaseous waste can be solved by implementing an efficient off-gas system, and ^{133}Xe and iodine isotopes are often trapped on special materials for decay or commercial use.

Solid wastes do not present serious difficulties. Normally, compaction methods for LLW and ILW can be applied for volume reduction and subsequent conditioning in special containers. Shielding is normally needed for a short period and interim storage facilities assist in allowing decay to take place before further handling to reduce radiation doses.

Liquids or solids containing enriched uranium should preferably be recycled for refabrication of targets. It is highly recommended that this approach be implemented in the production process. Other waste solutions, containing all fission products and actinides, should be mixed and adjusted to pH 7 to 8 and immobilized with cement. This method is a well proven technology and should be available also in developing countries.

As a result of international regulations, standards and requirements suitable for waste management systems for ^{99}Mo production are very expensive. Developing countries with the ambition to operate research reactors and the production of radioisotopes for medical purposes (including ^{99}Mo) do not always notice the burden and difficulties connected with these high aims. The establishment of a waste management infrastructure and the operation of a complete safe and proper waste management system is extremely difficult and expensive in the long term. Therefore economical and ecological investigations should be undertaken before a decision is made to operate a research reactor with its accompanying infrastructure for radioisotope production, rather than to purchase the necessary radioisotopes from commercial suppliers with products of proven quality.

6.3. RECOMMENDATIONS

- An optimal strategy for waste management should be adopted during the first stage of development of a ^{99}Mo production process.
- All phases of planning, construction, operation and decommissioning of ^{99}Mo production and associated waste processing facilities should be licensed by the competent national authorities.
- Before starting up a ^{99}Mo production facility, existing waste management facilities must be available. However, such facilities cannot be justified only for ^{99}Mo waste.
- The practice of a long term accumulation of waste for "future treatment" should be avoided.
- The concept of waste minimization and optimization of all procedures should be followed thoroughly. In this case the IAEA recommendations could be followed [45]. Avoidance of cross-contamination and secondary wastes arising is especially important.
- Exemption levels should be taken into account in order to minimize the volume of radioactive waste to be treated and disposed of.
- Consideration and implementation of advance waste management developments is recommended to ensure the best waste management practices, especially for a new ^{99}Mo production facilities.

- The final product after waste treatment, conditioning and packaging must fulfill the acceptance criteria for final disposal, which can differ from site to site and should be accepted by the competent national authority.
- All actions related to the use of radioactive materials and waste must keep the ALARA principle in focus. Waste must be conditioned in the best possible way with the least possible environmental impact.
- When producing ^{99}Mo by fission route recycling of unfissioned uranium is highly recommended; preferably the uranium should be segregated from the waste streams within the production facility.
- Safeguards requirements should be fulfilled, especially when using highly enriched uranium targets.
- The economic aspects of ^{99}Mo-related waste management are important and can have a substantial influence on the final price of ^{99}Mo and by-products.

REFERENCES

[1] INTERNATIONAL ATOMIC ENERGY AGENCY, Fission Molybdenum for Medical Use, IAEA-TECDOC-515, Vienna (1989).

[2] INTERNATIONAL ATOMIC ENERGY AGENCY, Alternative Technologies for ^{99}Tc^{m} Generators, IAEA-TECDOC-852, Vienna (1995).

[3] HELUS, F., COLOMBETTI, L.G., Radionuclide Production, Vol. II, CRL Press, Inc. (1983) 126–152.

[4] INTERNATIONAL ATOMIC ENERGY AGENCY, Radioisotope Production and Quality Control, Technical Reports Series No. 128, IAEA, Vienna (1971) 698–701.

[5] TUCKER, W.D., GREEN, M.W., WEISS, A.J., MURRENHOFF, A.P., BNL 3746, American Nuclear Society Annual Meeting, Los Angeles, CA, 1958. Trans. Am. Nucl. Soc., v. 1 (1958) p. 160.

[6] TUCKER, W.D., GREEN, E.M.W., MURRENHOFF, A.P., Die Produktion von trägerfreiem Tellur-132, Jod-132, Molybdan-99 und Technetium-99m aus neutronenbestrahltem Uran durch fraktionierte Sorption an Aluminiumoxide, Atompraxis, v. 8 (1962) 163–167.

[7] STANG, Jr. L.G., Manual of Isotope Production Processes in Use at Brookhaven National Laboratory, BNL-864 (1964).

[8] HARPER, P.V., ANDROS, S., LATHROP, K.A., Semiannual Report to the Atomic Energy Commission. (Jacobson, L.O., Ed.) ACRH-18 (1962) 76–87.

[9] HARPER, P.V., BECK, R., CHARLESTON, D., LATHROP, K.A., Optimization of a scanning method using ^{99m}Tc, Nucleonics v. 22, N1 (1964) 45–60.

[10] SEABORG, G.T., WAGNER, J.H.N., Nuclear medicine. 100 years in the making. 1896–1996. Amer. Soc. Nucl. Medicine (1996) 20 p.

[11] SHIKATA, E., IGUCHI, A., Production of ^{99}Mo and its application in nuclear medicine, Journal of Radioanalytical and Nuclear Chemistry, Special Report, Vol. 102, No. 2 (1986) 533–550.

[12] TREVENA, I., “Production and supply of radioisotopes with reactors in North America and Europe: Current status and future prospects”, Proc. 21st Conf. for the Radiation and Radioisotopes, Sect. B-5, p. B520, 1–6, Japan (1994).

[13] DE VILLIERS, W. Van Zyl., “The production of fission molybdenum-99 and management of the resultant waste”, IAEA Regional Training Course on Management of Low Level Radioactive Waste from Hospitals and other Nuclear Applications, Pretoria, South Africa, 27 June–13 July 1995.

[14] BOYD, R.E., Technetium-99m generators — The available options, Int. Appl. Radiat. Isot., v. 33, N 10 (1982) 801–809.

[15] HARDY, C.J., Research on Medical Applications of Radioisotopes and Radiation in Australia, Trans. Am. Nucl. Soc. v. 56(1) (1988) 386–392.

[16] BOURGES, J., MADIC, G., KOEHLY, G., NGUYEN, T.H., BALTES, D., LANDESMAN, C., SIMON, A., On the French project developed in the 1980s for the production of ^{99}Mo from the fission of ^{235}U, Nucl. Technol. 1996 Vol. 113, N2 (1996) 204–220.

[17] BALL, R.M., “Use of LEU in the aqueous homogeneous medical isotope production reactor”, Int. Mtg on Reduced Enrichment for Research and Test Reactors, Williamsburg, VA, 1994.

[18] LAGUNAS-SOLAR, M.C., KIEFER, P.M., CARVACHO, O.F., LAGUNAS, C.A., CHA, Ya. P., Cyclotron production of NCA ^{99m}Tc and ^{99}Mo. An alternative non-reactor supply source of instant ^{99m}Tc and ^{99}Mo6^{99m}Tc generators, Appl. Radiat. Isot., Int. J. Radiat. Appl. Instrum. Part A. Ver.42, N7 (1991) 643–657.

[19] SALACZ, J., Production of fission ^{99}Mo, ^{131}I and ^{133}Xe, Revue I.R.E., Vol. 9, N3, (1985) 3–6.

[20] MARCHAND, L., Irradiation of ^{235}U in OSIRIS reactor for the production of ^{99}Mo, ^{131}I, ^{133}Xe radioisotopes, Review I.R.E. Vol. 9, N3 (1985).

[21] SAMEH, A.A., ACHE, H.J., Production techniques of fission molybdenum-99, Radiochimica Acta, Vol.41 (1987) 65–72.

[22] CHENG, W.L., LEE, C.S., CHEN, C.C. WANG, Y.M., TING, G., Study on the separation of ^{99}Mo and recycling of uranium to water boiler reactor, Appl. Radiat. Isot., Int. J. Radiat. Appl. Instrum. Part A, Vol.40, N 4 (1989) 315–324.

[23] COHILIS, P., JONGEN, Y., LANNOYE, G., "Recent advances in the design of a cyclotron-driven, intense, subcritical neutron source", Ion Beam Applications S.A. (IBA), Louvain-La-Neuve, Belgium (1996).

[24] JONGEN, Y., "A proton-driven, intense, subcritical, fission neutron source for radioisotope production", Int. Conf. on Accelerator-Driven Transmutation Technologies and Applications, Las Vegas, NV (1994).

[25] LEVINS, D.M., et al., ANSTO's Radioactive Waste Management Policy — Preliminary Environmental Review, ANSTO/E728, Australian Nuclear Science and Technology Organization, Lucas Heights, Australia (1996).

[26] SAMEH, A.A., Produktion von Spaltmolybdän für die Nuclearmedizin aus bestrahlten Uran nidriger Anreicherung, KfK-Nact z., Jahrg 25, 2/93 & 83-89 KfK Nachr., vol. 25, N2 (1993) 83.

[27] HUTTER, J.C., SRINIVASAN, B., VICEK, M., VANDEGRIFT, G.F., "Production of ^{99}Mo using low-enriched uranium silicide", 1991 Int. Mtg on Reduced Enrichment for Research and Test Reactors, Williamsburg, VA (1994).

[28] VANDEGRIFT. G.F., CHAIKO, D.J., HEINRICH, R.R., KUCERA, E.T., JENSEN, K.J., POA, D.S., VARMA, R., VISSERS, D.R., Preliminary Investigations for Technology Assessment of ^{99}Mo Production from LEU Targets, 1986 Int.Mtg on Reduced Enrichment for Research and Test Reactors (RERTR), Gatlinburg, TN, ANL/RERTR/TM-9, CONF-861185 (1986) 64–79.

[29] VANDEGRIFT, G.F., HUTTER, J.C., SRINIVASAN, B., MATOS, J.E., SNELGROVE J.L., "Development of LEU targets for ^{99}Mo production and their chemical processing. Status 1993", 1993 Int. Mtg on Reduced Enrichment for Research and Test Reactors, Ibaraki, Japan (1993).

[30] VANDEGRIFT, G.F., KWOK, J.D., MARSHALL, S.L., VISSERS, D.R. and MATOS, J.E., "Continuing investigations for technology assessment of ^{99}Mo production from LEU targets", 1987 Int. Mtg on Reduced Enrichment for Research and Test Reactors (RERTR), Buenos Aires, Argentina (1987).

[31] MARSHALL, S.L., REDEY, L., VISSERS, D.R., VANDEGRIFT, G.F., MATOS, J.E., "Continuing investigations on electrochemical preparation of LEU targets for ^{99}Mo production", 1988 Int. Mtg on Reduced Enrichment for Research and Test Reactors (RERTR), San Diego, CA (1988) 470–486.

[32] COHILIS, P., et al., "Recent advances in the design of a cyclotron-driven, intense, subcritical neutron source", EPAC'96 (1996).

[33] CROFF, A.G., ORIGENZ: A versatile computer code for calculating the mileage compositions and characteristics of nuclear materials, Nucl. Technol. 62, 335 (1983).

[34] VLCEK, J., Problemu likvidaci naplni generatoru ^{99}Mo/^{99m}Tc Jaderna Energie, v. 28 No.7 (1982) 264–265.

[35] LE, V.T., The Molybdenum-99 Production Facility, Annual Safety Review, AECL-MISC-303-94 (1994).

[36] INTERNATIONAL ATOMIC ENERGY AGENCY, Advisory Material for the IAEA Regulations for the Safe Transport of Radioactive Material (1985 Edition), Safety Series No. 37, IAEA Vienna (1990).

[37] INTERNATIONAL ATOMIC ENERGY AGENCY, Quality Assurance for Safety in Nuclear Power Plants, A Code of Practice, Safety Series No 50-C-QA, IAEA, Vienna (1978).

[38] CODIE, H.D.K., KASTELEIN, J., "Commissioning a new facility for the treatment and storage of low and medium level waste in the Netherlands", paper presented at SPECTRUM'96, Seattle, WA, 1996.

[39] INTERNATIONAL ATOMIC ENERGY AGENCY, Decontamination and Decommissioning of Nuclear Facilities, IAEA-TECDOC-716, IAEA, Vienna (1993).

[40] INTERNATIONAL ATOMIC ENERGY AGENCY, Decommissioning of Nuclear Facilities other than Reactors, Technical Reports Series No. 386, IAEA, Vienna (1998).

[41] INTERNATIONAL ATOMIC ENERGY AGENCY, Decommissioning of Nuclear Facilities: Decontamination, Disassembly and Waste Management, Technical Reports Series No. 230, IAEA, Vienna (1983).

[42] INTERNATIONAL ATOMIC ENERGY AGENCY, Chemical Durability and Related Properties of Solidified High Level Waste Forms, Technical Reports Series No. 257, IAEA Vienna (1985).
ISO 6961-1982 (E) Long term leach testing of solidified radioactive waste forms.

[43] INTERNATIONAL ATOMIC ENERGY AGENCY, Conditioning of Alpha Bearing Wastes, Technical Reports Series No. 326, IAEA, Vienna (1991).

[44] INTERNATIONAL ATOMIC ENERGY AGENCY, Acceptance Criteria for Disposal of Radioactive Wastes in Shallow Ground and Rock Cavities, Safety Series No. 71, IAEA Vienna (1985).

[45] INTERNATIONAL ATOMIC ENERGY AGENCY, Regulations for the Safe Transport of Radioactive Material, IAEA Safety Standard, Safety Series No. 6, Vienna (1990).

[46] INTERNATIONAL ATOMIC ENERGY AGENCY, Report on Radioactive Waste Disposal, Technical Reports Series No. 349, IAEA, Vienna (1993).

[47] INTERNATIONAL ATOMIC ENERGY AGENCY, Review of Available Options for Low and Intermediate Level Radioactive Waste Disposal, IAEA-TECDOC-661, Vienna (1992).

[48] INTERNATIONAL ATOMIC ENERGY AGENCY, Operational Experience in Shallow Ground Disposal of Radioactive Waste, Technical Reports Series No. 253, IAEA, Vienna (1985).

[49] INTERNATIONAL ATOMIC ENERGY AGENCY, Quality Assurance for Radioactive Waste Packages, Technical Reports Series No. 376, IAEA, Vienna (1995).

[50] INTERNATIONAL ATOMIC ENERGY AGENCY, Inspection and Testing in Conditioning of Radioactive Waste, IAEA-TECDOC-959, Vienna (1997).

[51] INTERNATIONAL ATOMIC ENERGY AGENCY, Characterization of Radioactive Waste Forms and Packages, Technical Reports Series No. 383, IAEA, Vienna (1997).

[52] INTERNATIONAL ATOMIC ENERGY AGENCY, Requirements and Methods for Low and Intermediate Level Waste Package Acceptability, IAEA-TECDOC-864, Vienna (1996).

[53] INTERNATIONAL ATOMIC ENERGY AGENCY, Minimization and Segregation of Radioactive Wastes, IAEA-TECDOC-652, Vienna (1992).

CONTRIBUTORS TO DRAFTING AND REVIEW

Basmanov, V.	IPPE, Russian Federation
Baehr, W.	Speyer, Germany
Efremenkov, V.M.	International Atomic Energy Agency
Holub, J.	NYCOM, Czech Republic
Luycx, P.	Belgoprocess, Belgium
Maton, F.	Institute National des Radioéléments, Belgium
Tsarenko, A.F.	International Atomic Energy Agency
Vera Ruiz, H.	International Atomic Energy Agency
Van Kleef, R.	COVRA, Netherlands
Van Zyl de Villers, W.	Atomic Energy Corporation of South Africa Ltd, South Africa
Wong, P.	AECL, Chalk River Laboratories, Canada

Consultants Meetings

Vienna, Austria: 4–8 December 1995, 23–27 September 1996