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Research Article

Evaluation of the permissible ^{99}Mo activity in the KL-15 cask in the design of transportation and process scheme*

Vladimir V. Fomichev[1], Denis A. Pakholik[1], Oleg Yu. Kochnov[1], Nikita V. Kuznetsov[1], Mikhail V. Kharitonov[1], Vyacheslav V. Nichugovsky[2]

1 *JSC Karpov Institute of Physical Chemistry, 6 Kievskoye shosse, 249033 Obninsk, Kaluga Reg., Russia*

2 *Obninsk Institute for Nuclear Power Engineering, MEPhI, 1 Studgorodok, 249040 Obninsk, Kaluga Reg., Russia*

Corresponding author: Oleg Yu. Kochnov (kochnov@karpovipc.ru)

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Abstract

The demand for the use of radioactive isotopes in medicine is increasing with each coming year necessitating the increased output of radionuclide products. One of the most widely spread radionuclides used in medicine is technetium-99m (^{99m}Tc) (Feasibility of producing molybdenum-99 2015, NEA 2012, The Supply of Medical Radioisotopes 2015). The very short ^{99m}Tc life (6-hour half-life) requires its production directly on the site of medical treatment. This is achieved using molybdenum-technetium generators (Kodina and Krasikova 2014, Technical Reports No. NF-T-5.4. 2013, Technetium-99 Generator 2021) loaded with molybdenum-99 (^{99}Mo), which uninterruptedly decays (half-life of 66 hours) yielding ^{99m}Tc.

Close attention must be paid in the course of production of molybdenum-technetium generators to radiation safety during transportation of ^{99}Mo on the territory of the manufacturing facility. The main measure for ensuring radiation safety during transportation of ^{99}Mo is the application of special packaging kits. The Karpov Institute of Physical Chemistry JSC uses a wide range of packaging kits of types A andB for transportation of radioactive materials on the territory of the manufacturer with design features providing the required level of radiation safety.

In particular, the KL-15 shipping cask loaded/unloaded from the top is used for onsite transportation of ^{99}Mo for charging molybdenum-technetium generators. The maximum permissible activity of ^{99}Mo is not specified in the passport of the KL-15 cask. Planned construction of a radionuclide production shop in accordance with GMP requirements will require the increase of output of target radionuclides by several times. The above considerations necessitated the evaluation of the maximum permissible activity of ^{99}Mo planned to be transported in KL-15 casks. No other type of standard casks can be used because of their outside dimensions prohibiting the unloading of ^{99}Mo inside the "hot" chamber. Calculation and experimental evaluation of permissible ^{99}Mo activity during transportation inside the KL-15 cask wasperformed.

The paper presents the calculated evaluation of the maximum permissible activity of ^{99}Mo in a KL-15 cask to ensure the radiation exposure of personnel of group A working with the cask not exceeding the established level at the enterprise (80 µSv per shift) and not requiring the use of additional measures and means of protection.

The results of the work allow us drawing the conclusion that the KL-15 cask ensures the required level of radiation safety with up to 241 Ki of ^{99}Mo loaded in the cask.

Keywords

Molybdenum-99, technetium-99m, molybdenum-technetium generators, radiation safety during transportation, packaging kits, KL-15 shipping cask, maximum permissible activity

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Introduction

The goals and objectives of the Public Health national program and the regional cancer struggle programs dictate the need for increased production of radiopharmaceuticals. One of the radionuclide products by JSC Karpov Institute of Physical Chemistry is a molybdenum-technetium generator used for diagnosis and treatment of cancer. The ^{99}Mo nuclide is the key radioisotope used for these purposes. It decays to the short-lived ^{99m}Tc isotope that is used across the world for about 70% of diagnostic procedures in oncology and for up to 50% of those in cardiology.

An increase in production of molybdenum-technetium generators requires a great deal of attention to be given to radiation safety measures in transportation of ^{99}Mo (Specific Safety Guide. No. SSG-26 2014, NP-053-16 2017, Regulations for the Safe Transport 2019), including the radionuclide transport through the manufacturer's site. Radiation safety in the process of the ^{99}Mo transport is defined to a large extent by the design peculiarities of respective packaging kits.

Depending on the type of radiation emitted by radioactive materials (RM), there are three categories of packaging kits (Freiman et al. 1986):

- category I, used to transport gamma radiation sources; these kits include radiation safety devices (a shielded cask, a safety liner) made of lead, cast iron, steel, uranium and other heavy materials;
- category II, used to transport neutron sources; for protection against neutron radiation, such kits use paraffin or other water-containing substances with addition of boron or cadmium;
- category III, used to transport RMs that emit alpha and beta particles; kits of this type normally use light materials (aluminum and all kinds of plastics), as well as small-size lead safety liners.

In terms of the capability to retain the protective and sealing properties in conditions of external impacts, packaging kits for transportation of radioactive materials are divided into two main types (Freiman et al. 1986):

- type A is designed for normal conditions of transportation, that is, the kits are required to withstand the impacts encountered in normal transport of radioactive materials (fall from a small height, impact from neighboring goods, compression, rainstorm);
- type B is designed for service in potential transport emergencies (more specifically, in tests with simulation of normal and emergency conditions) without a change in the protective properties or with a very small decrease in the efficiency of the sealing and radiation protection systems.

Estimation and experimental evaluation of the permissible ^{99}Mo activity in a KL-15 cask

A broad range of type A and B packaging kits is used at the Karpov Institute of Physical Chemistry for handling of radioactive materials. One of these is the KL-15 shipping cask.

The KL-15 shipping cask (see Fig. 1 for its overall schematic view) has the following key characteristics:

- category – I;
- type – A;
- total cask height – 540 mm;
- minor diameter of truncated cone – 230 mm;
- cylinder diameter – 355 mm;
- shielding – lead (Pb);
- minimum shielding thickness – 150 mm;
- total cask volume – 46 000 cm;
- cask useful volume – 289 cm.

Figure 1. Overall view of a KL-15 laboratory cask.

To estimate the optimal conditions for operating a KL-15 cask, we shall determine the maximum activity of ^{99}Mo, with which the exposure for Group A personnel (SanPiN 2.6.1.2523-09 2009, SP 2.6.1.2612-10 2010) will not exceed the site specified level (80 μSv for the workday or 11.43 μSv/h).

For the calculation, we shall use the gamma radiation characteristics taken from (Kozlov 1977) (Table 1).

Table 1. Characteristics of gamma radiation

Isotope	Photon energy E_γ, MeV	Quantum yield for decay η, %	Differential gamma constant K_γ, R×cm²/(mCi×h)
^{99}Mo	0.960	1	0.051
	0.750	13	0.547
	0.180	4	0.037
	0.140	89	0.592

We shall interpolate the data for the linear attenuation factors for the Pb shielding taken from (Golubev 1986) (Table 2).

By estimating value μ*d* where μ ist the linear attenuation factor, a conclusion can be made that, for the lead

Table 2. Linear attenuation factors for Pb (d = 150 mm)

Isotope	Photon energy E_γ, MeV	Linear attenuation factor μ, cm^{-1}	μd
^{99}Mo	0.960	0.83	12.5
	0.750	1.02	15.3
	0.180	14.04	210.6
	0.140	25.93	389.0

shielding (d = 150 mm), gamma energies of 180 and 140 keV are absorbed practically in full in the cask's lead shielding and do not affect the total personnel exposure.

We shall determine the dose accumulation factor for a point isotropic generator in an infinite medium through the interpolation of data presented in (Mashkovich and Kudryavtseva 1995). We get the values shown in Table 3 as a result of the interpolation.

Table 3. Exponential dose accumulation factor for Pb (d = 150 mm)

Isotope	Photon energy E_γ, MeV	Dose accumulation factor Bd
^{99}Mo	0.960	3.55
	0.750	3.27

The gamma constant for a heterochromatic source for the Pb shielding (d = 150 mm) can have the form of the expression as follows

$$K_\gamma(d,Z) = \sum_i K_{\gamma_i} \cdot \exp(-\mu_i d) \cdot \delta_i \cdot B_{\infty i}\left(E_{\gamma_i}, \mu_i, d, Z\right), \quad (1)$$

where $\Sigma K_{\gamma i}$ is the sum of differential gamma constants, R×cm^2/(mCi×h); $E_{\gamma i}$ is the energy of gamma quanta, MeV; d is the lead shielding thickness, cm; Z is the atomic number of the shielding material; μ_i is the linear attenuation factor for a narrow gamma beam in the shielding material, cm^{-1}; δ_i is the ratio of the dose accumulation factor in a barrier geometry to the dose accumulation factor in an infinite medium; and $B_{\infty i}$ is the dose accumulation factor in an infinite medium.

We get the ratio of the dose accumulation factor in a barrier geometry to the dose accumulation factor in an infinite medium, δ_i, for the lead shielding by interpolating the data presented in (Mashkovich and Kudryavtseva 1995) (Table 4).

Table 4. Ratio of the dose accumulation factor in a barrier geometry to the dose accumulation factor in an infinite medium for the Pb shielding

Isotope	Photon energy E_γ, MeV	δ_i
^{99}Mo	0.960	0.9866
	0.750	0.9863

Using expression (1), we obtain value K_γ (Table 5).

Table 5. Gamma constant of a heterochromatic source for Pb (d = 150 mm)

Isotope	Gamma constant of heterochromatic source K_γ, R×cm^2/(mCi×h)
^{99}Mo	1.065×10^{-6}

The activity of the source is calculated by the following formula

$$A = P \times d^2 / K_\gamma, \quad (2)$$

where A is the source activity, mCi; P is the exponential dose rate, R/h; and K_γ is the gamma constant of the heterochromatic source, R×cm^2/(mCi×h).

In conditions of the charged particle ray equilibrium, as shown in (Mashkovich and Kudryavtseva 1995), an exponential dose of 1 C/kg is matched by an absorbed dose of 33.85 Gy in air or 36.9 Gy in the biological tissue; the arbitrary unit of 1 R is matched by an absorbed dose of 0.873 rad in air or 0.95 rad in the biological tissue. Therefore, with an accuracy of up to 5%, the exponential dose in R and the absorbed dose in tissues in rad can be considered coincident. The radiation quality factor with a photon energy of over 350 keV is equal to unity. The exponential dose in R, in this case, can be therefore approximately considered to be equal to an equivalent dose of 0.01 in Sv.

Using a conservative approach and by means of formula (2), we shall determine the maximum activity of ^{99}Mo in a KL-15 cask with which the personnel exposure dose does not exceed the specified level of 80 μSv (a dose rate of 11.43 μSv/h) for Group A personnel for a work shift (Table 6).

Table 6. Activity in cask

Isotope	Activity A, Ci
^{99}Mo	241

An experimental study was undertaken for the KL-15 protective properties to confirm the obtained estimation result for the permissible activity of ^{99}Mo. The dose rate was measured for different activity values of ^{99}Mo obtained as the result of the manufacturing process for production of molybdenum-technetium generators. An MKS-AT1117 dosimeter-radiometer was used for the measurements (Description of the Measuring Instrument Type 2013). The measurement point was selected with taking into account the discontinuity in the KL-15 lead shielding thickness caused by the geometrical shape of the cask (a cylinder with two adjacent truncated cones) and the centrally positioned radiation source (Fig. 2). With regard for these cask design features, a measurement point has been selected at which the shielding thickness relative to the radiation source position has the lowest value equal to 150 mm. The dose rate measurement results are presented in Table 7.

Table 7. Experimental dose rate values

Experiment No.	Activity of ^{99}Mo, Ci	Cask surface dose rate, μSv/h
1	238.7	8.97 ± 1.80
2	239.1	9.07 ± 1.81
3	240.6	9.71 ± 1.94
4	239.7	9.19 ± 1.84
5	240.2	9.65 ± 1.93

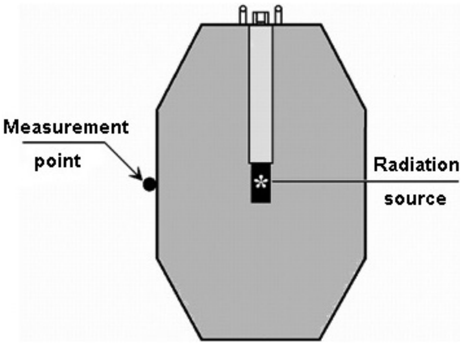

Figure 2. Measurement point and radiation source positions in the KL-15 cask.

The data in the table demonstrates that the KL-15 cask ensures proper personnel protection when containing the ^{99}Mo radionuclide with the activity as close to the estimated value as possible. Of the greatest interest is the result of measuring the dose rate with the cask containing molybdenum-99 with the activity 240.6 Ci. In this case, the dose rate value assumed for the permissible activity calculation exceeds the experimental value by 15%. This circumstance has been caused by the simplifications adopted for the calculations and the measurement uncertainty. However, the experimental study confirms that the undertaken calculation is conservative and allows a conclusion that the obtained result can be used in practice.

Conclusion

The calculations performed and the obtained experimental data allow a conclusion that the KL-15 cask fully ensures safety of its handling when containing ^{99}Mo with an activity of up to 241 Ci, this being especially important in ensuring safety of the manufacturing process with regard for the annual increase in the quantity of consumed and produced radionuclide products.

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