ON POSSIBILITY OF ALPHA-RADIOACTIVE THICK-LAYER WASTE ANALYSIS USING HIGH-VACUUM TECHNIQUE METHODS

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A practical possibility of specific alpha-activity measurement of the radioactive waste (RAW) as much as thousands tons placed in the 4-th Chornobyl wrecking unit is demonstrated by means of determination of the helium content (on the account of Pu, Am decay) in the hermetic containers filled with the RAW. The method using application of helium filter is acceptable but filterless one seems to be more economical. The latter is based on a constraint of all gases into choused gaseous medium but helium. The appraisal of necessary time for passive helium accumulation up to partial pressure of ~10^{-7} Pa (a value to be measured by mass-spectrometer) is of ~1000 hours using a scheme with real helium filter and of ~10 hours in the case without filter for specific alpha-activity of ~10^5 Bq/kg. (a threshold level for the RAW disposition).
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1. INTRODUCTION

Investigation of thousands tons of the 4-th Chernobyl unit radioactive waste (RAW) for the purpose of determination of their activity group can be carried out by means of measurement of the helium making rate from the α radiators (Pu, Am, U). The disposable facilities for measurement of the low partial pressure of gases and the use of helium accumulation in a closed volume allows to determine sufficiently low specific activities including conditionally secure ones, when the RAW does not demand of especial deposition [1,2]. Two methods and two schemes of measuring plant concerning this problem are proposed and analyzed in the report.

2. METHOD WITH USAGE OF HELIUM FILTER

It is possible to represent a measuring plant supplied with a helium filter as a hermetic container having gas volume V_1 (Fig.1). The latter is filled with studied material and connected to measuring volume V_2 through a filter having conductivity for helium G [3].

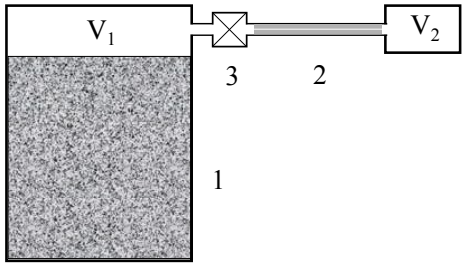

Fig. 1. *Simplified scheme of measuring plant (for calculation): 1 – container with RAW; 2 – helium filter; 3 – filter valve*

In case when the helium flux from the RAW is B, and the filter is opened after a time of helium accumulation (only in the free volume V_1) t_0 then an increase of the helium pressure P in the measuring volume as a function of the time **t** from filter opening can be described by expression

$$P(t) = \frac{B}{V_1 + V_2}\left[t + (t_0 - \tau)\left(1 - e^{-t/\tau}\right)\right] \quad , \qquad (1)$$

where $\tau = \dfrac{V_1 \cdot V_2}{G(V_1 + V_2)}$. (2)

So, when the plant parameters as well as the times t_0, τ and a material mass are known it is possible to calculate the helium flux and specific α activity of the RAW by means of single measurement of helium pressure. It is important to know precisely and to be sure of temporal stability of two parameters: a sensitivity of the mass-spectrometrical pressure measurer and a filter conductivity. A static mass-analysing magnet seems to be most suitable for a real application [4]. A filter maid of fused quartz has the most stable conductivity for helium. These assemblies can be calibrated previously at a routine plant with a sufficient guarantee of their parameters keeping. Taking into account a gas type it should be assumed also for the larger accuracy, that the helium pressure must to be more than 10^{-7} Pa.

To estimate a necessary time for helium accumulation let us consider a case of the RAW with the specific α activity $1 \cdot 10^5$ Bq/kg. – a mean value in the region of particle interest [6]. The container of 200 l. volume has been filled with a destructured concrete having the apparent density 1.6 kg./l. and solid mass 320 kg. So, the volume of solid matter is 140 l. and the pore volume – 60 l. Let us assume also that V_1=70 l. including some technological vessels. The measuring volume let be V_2=5 l. (together with the mass-spectrometer and helium purification devices). The helium filter (coaxial quartz tubes: ∅100x1 and ∅96x1 of 60 cm length) has conductivity $G = 8 \cdot 10^{-6}$ l./s. under quartz temperature 800 °C according to the known data [5], so τ is equal to 186 h. Total α activity of the material in the container is $3.2 \cdot 10^7$ Bq, that causes the helium flux $B = 1.33 \cdot 10^{-10}$ l.·Pa/s. Dependence of the helium pressure P in measuring volume on the time from filter opening calculated from the expression (1) is represented in Fig. 2.

It is seen that the total time (t_0+t) to achieve a pres-

sure to be measured is the least when a helium filter is previously opened. However, taking into account the energy for the filter heat, it seems to be profitable to keep a number of prepared containers with closed filters. A choice of the measuring regime is obviously compromise. In this example the time of accumulation and measurement can not exceed of 500 h. A real plant will be, of course, sufficiently complicated in comparison with the scheme described above and a measuring procedure will be rather labour-consuming. In particular, it is necessary to provide a material drying, a measurement of gas volume in the filled container (for instance, using the method of controllable compression), a removal of the atmospheric background helium (obviously, by means of pumping out using a powerful oil-diffusion pump), preliminary gassing of quartz from the atmospheric helium.

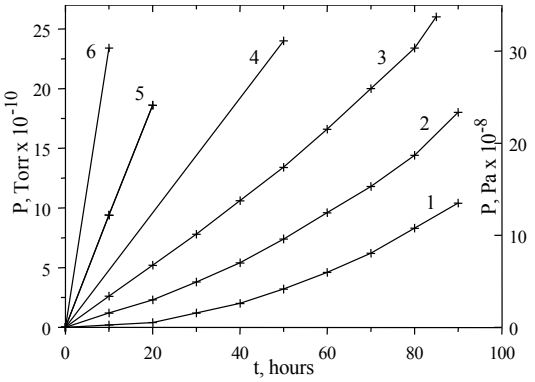

Fig. 2. *Helium pressure in measuring volume vs time from filter opening for several values of helium preliminary accumulation time t_0 in container of V_1 volume only (in conditions of example).* ***1*** *- t_0=0 ;* ***2*** *- t_0=0.1, τ=18.6 h.;* ***3*** *- t_0=0.2, τ=37.2 h.;****4*** *– $t_0=\tau$ =186 h.;* ***5*** *– $t_0=2\tau$=372 h.;****6*** *– $t_0=5\tau$=930 h*

The gas pressure in the measuring volume in presence of acceptable size filter is much less than in the container. There is the expression for this pressure and for the overfall of the pressure through the filter:

$$\Delta P(t) = \frac{B\tau}{V_1}\left(1 + \frac{t_0 - \tau}{\tau} e^{-t/\tau}\right) \quad . \qquad (3)$$

In the particular case of $t_0=\tau$, the pressure overfall does not depend on the time, $\Delta P=V\cdot\tau/V_1$. The pressure overfall is approaching to this value in time under $t_0<\tau$ or $t_0>\tau$ from the side of less or more values correspondingly. In the adduced example ΔP is about $1.3\cdot10^6$ Pa. It is possible to rise sufficiently an increase of the measured helium pressure by application of a thin filter with a large surface manufactured of some organic materials having under indoor temperature more conductivity relative to helium than a hot quartz [5]. However, this fact causes a grave doubt, generally, because a possible instability of the organic filters conductivity as well as taking into account a large gassing of them in a vacuum and their large penetrability for a number of light organic compounds.

3. FILTERLESS METHOD

An analysis of the RAW can be speed up (and, may be, cheapen) by usage of a filterless method. In conditions of the sample mentioned above, the partial pressure in the volume V_1+V_2=75 l. is increasing at the rate of about $6.6\cdot10^{-9}$ Pa/h. (~$5\cdot10^{-11}$ Torr/h.). The pressure ~ $1.3\cdot10^{-6}$ Pa (~$1\cdot10^{-8}$ Torr) will be achieved for the accumulation time of 200 h., that allows to increase the accuracy of the pressure measurement and to reduce the demands to completeness of evacuation of the atmospheric background helium. It is eliminated also the uncertainty caused by filter conductivity. The measuring volume is connected to the container for a short period and then cut off for a measurement. However, "the purification facilities" of the measuring volume must have by far more gas capacity for absorption of predominant component of the gassing in the container and can be more complicated in comparison with a plant having a helium filter. It should be noted that the measuring volume must be small because a mass-spectrometer itself has a sufficient rate of the pumping out. As the estimation shows, on ionizer of the mass-spectrometer (electron current of 5 mA at the distance of ~2 cm) isolated before measurement from the volume and working under ultrahigh vacuum (~10^{-9} Pa) can provide the helium ion pumping at the rate up to $5\cdot10^{-3}$ l./s. For the measuring volume of 5 l. the ion pumping time constant is about 1000 s. So, a measurement have to be carried out for a time of ~10 s. A realization of the filterless method is facilitated by preliminary evacuation of the studied material. A specific α activity I of the material with the mass of M kg, gas volume of (V_1+V_2) l. and partial pressure of the helium P (for the accumulation time of t s.) is given by expression

$$I = 2.49\cdot10^{17}\frac{P(V_1+V_2)}{Mt}\ Bq/kg., \qquad (4)$$

where $2.49\cdot10^{17}$ – the number of gas atoms in 1 l. under pressure of 1 Pa and temperature of 25°C.

4.CONCLUSION

This report is represented as a some technical proposal. Not all issues are finally resolved. A rate of the helium production in the grains of materials of the 4-th Chernobyl unit and going out into gas environment has reached of equilibrium during 15 years after the wreck. Therefore, one can consider a helium flux from the grains of the RAW as directly proportional to their α activity. However, it is not clear how to use helium inside the grains containing a good deal of information and which can be released by especial treatment of the material. It should be excluded a possibility of helium loss under its cleaning, properly, by maintenance of the total gas pressure not above of 10^{-2} Pa, that is necessary for a normal work of the mass-spectrometer. A number of issues demands of experimental study.

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