INSTRUMENTS AND METHODS FOR DAY-TO-DAY MONITORING OF TRITIUM AND CARBON-14 IN PRODUCTION

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Abstract

A complex of instrumentation and techniques for dosimetric monitoring at industrial enterprises using unshielded beta-emitters of T and ^{14}C is offered. Air-borne radionuclides are assayed by automatic monitors developed on the basis of gas counters. A portable device for detection of radioactive contamination of surfaces is worked out. Scintillation and gas counters have been used to measure ^{14}C and T content in personnel. The sensitivity of the methods is sufficient for monitoring of all categories of exposure.

Introduction

The problems of protection from tritium and ^{14}C radiation arise in the manufacture and application of tracer compounds, luminophores, tritium-titanium targets as well as in spheres involving the use of the energy of nuclear fission and fusion. Certain difficulties, as far as the dosimetry of these radionuclides is concerned, are due both to their specific radiation characteristics and insufficient knowledge of the radiotoxicity of their compounds[1,2].

This paper does not deal with the biophysical aspects of the indirect dosimetry of T and ^{14}C; it describes instrumentation and methods for detecting low-enrgy beta-emitters in the human body and environment.

Monitoring of Radionuclides in Air

Gas filling counters are employed as detecting devices in all the monitors developed. The instruments also incorporate devices for passive and active shielding of the counters from external radiation, systems for intake, processing and regulation of the flows of assyaed air and filling gas, electronic registering systems, control and signalling assemblies as well as high- and low-voltage power supplies for the counters and transistorized circuits, respectively. The instruments are designed as portable double-tier columns with detecting device at the bot-

tom and gas systems and electronic blocks at the top.

A mixture of aerosol-free air (0.1 l/min) and methane (0.5 - 1 l/min) is fed to the detectors of instruments A and B. The air piping and proportional counter bodies are made of Teflon. The operating volume of Counter A (Fig.1a) of 0.15 l is limited by a cylindrical cathode consisting of 22 metal wires.[3] The de-

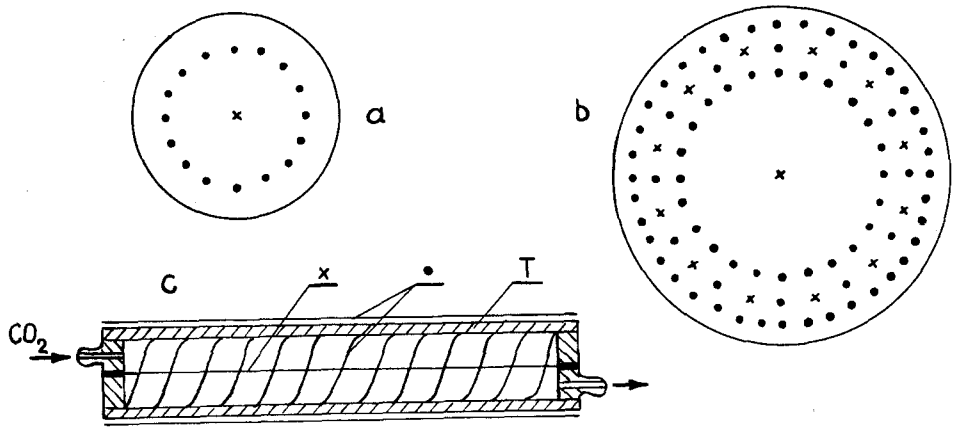

Fig.1. Schematic constructions of gas-flow counters.
x - anodes; . - cathodes; t - quartz tube.

tector and pre-amplifier are surrounded by a 5 cmethick lead shield. Pulses are registered by an integral discriminator with a sensitivity of 0.5 mV. To calibrate counters A and B, a flow of methane bubbler-saturated with T-ethanol vapours is passed in succession through the multi-wire counter and a calibration counter with known parametres. The counting efficiency is calculated with due regard to the ratio of the detector volumes and the known efficiency of the calibration counter.

The detection efficiency for T and ^{14}C beta-radiation for counter A operation in air-free methane is over 90%. A 500V - long counting plateau has no appreciable slope. Introduction of air affects the counting characteristics considerably. For a 5% admixture of air, the plateau length is reduced to 250V and its slope is 2%/100 V. The count curve for a mixture containing 10% of air in the region of 3.4 to 3.6 kV has a slope of 8%/100 V. The efficiency of beta particle detection in the working point (3.5 kV) is 85%. The count curve becomes still less stable as the air portion increases. The counter background of 150-180cpm remains stable up to 4.0 kV. The detector and gas system are cleaned from THO vapours and gaseous tritium exponentially (λ = 1 min^{-1}), from the moment radioactivity intake has ceased.

The 0.5 l counter B with a multi-wire cathode is enclosed in a ring of protection counters located in the same volume of gas (Fig.1b). A two-channel panel selects coincidence and anticoincidence pulses.

The counter B anticoincidence background is 35 cpm in methane, 70 cpm in 10% air mixture and 85 cpm at 20% content of air, respectively. The count curves for T and ^{14}C have a plateau slope of about 5%/100V at 10% and 20% air in the mixture. The counting efficiency is about 90%.

The assay of the energy composition of air-borne radioactivity is based on the correlation between the number of coincidence counts in the measuring and protection counters and the path of

primary ionizing particles[4]. Thus, the ratio of coincidence and anticoincidence counts for T is 0.1, whereas for ^{14}C - 1.6. The counter B measurement results may be used for estimating the ratio of nuclide concentrations in air.

Counter C (Fig.2) is intended for a selective determination

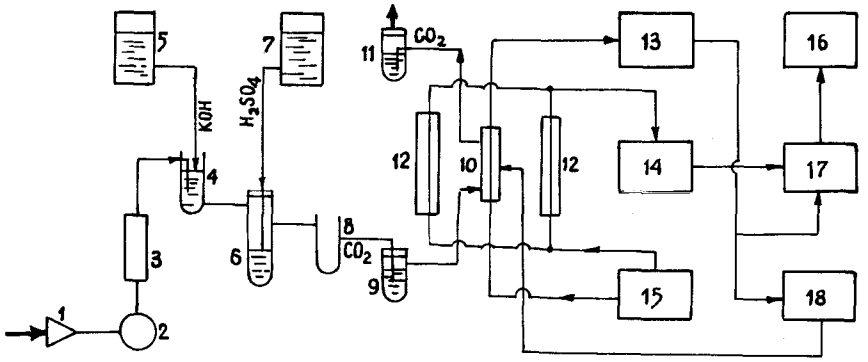

Fig.2. Block-diagram of air-borne ^{14}C monitor.
1 - air intake; 2 - compressor; 3 - oven; 4 - bubbler; 5 - vessel with KOH; 6 - reaction vessel; 7 - vessel with H_2SO_4; 8 - dessicator; 9 - bubbler with cyclohexan; 10 - CO_2-counter; 11 - liquid seal; 12 - schielding counters; 13,14-input blocks; 15 - high voltage block; 16 - recorder; 17 - measuring block; 18 - electronic suppressor.

of air-borne as carbon dioxide or carbonate aerosols. Air under assay is bubbled through a flow of a 10% solution of KOH to absorb CO_2. The solution of KOH and K_2CO_3 is passed to a reaction vessel with a 25% solution H_2SO_4. Carbon dioxide resulting from the reaction: $K_2CO_3 + H_2SO_4 \rightleftarrows K_2SO_4 + H_2CO_3$; $H_2CO_3 \rightarrow H_2O + CO_2\uparrow$ is passed through a desiccator and a quenching agent (cyclohexan) bubbler to the counter. CO_2 acts both as an operating gas in Geiger counter and a radiocarbon carrier.

The cathode of the CO_2 -counter is provided with a 0.3 mm-dia. platinum wire coil wound at a pitch of 5 mm inside the inner surface of a 18 mm-dia. quartz tube (Fig.1c). The counter volume is 30 cm^3; the anode diameter 0.1 mm.

The count curve in measuring crude CO_2 is quite good due to the incorporation of an external suppressor assembly with a pulse height of 1.5 kV, duration 2 msec. The plateau length is 400 V; the slope - 2%/100 V; the working point - 3600 V. The background of the counter surrounded by a 5 cm-thick lead shield at anticoincidence with a ring of shielding Geiger counters is 4-5 cpm. The counter radiometric characteristics for air monitoring are given in the Table. The minimal detectable concentration (MDC) in the atmospheric air is defined as a level capable of inducing a count rate equal to that of the background. The simplest monitor A ensures a reliable detection of maximum permissible concentrations for personal of THO vapours (5.10^{-9} Ci/l[6]) and ^{14}C compounds (3.5×10^{-9} Ci/l). The model B is capable of detecting concentrations lower than the above by an order as well as differentiating beta - radiation with respect to energy. The provision for the chemical selec-

Table of Parameters of Air-Borne Radioactivity Monitors

Counter	Gas to be assayed	Sample volume, cm^3	Background cpm	MDC, Ci/l of air	Response time min.
A	Air	15	150	$5x10^{-9}$	1
B	Air	100	85	$4x10^{-10}$	5
C	CO_2	30	4	$2x10^{-14}$	30

tion of CO_2 makes counter C applicable for monitoring of air-borne ^{14}C for all categories of exposure[6]. These instruments intended for operation under various conditions offer a still wider range of uses:

(a) automatic recording and signalling of emergency situations;
b) simultaneous sampling of air from several checking points;
(c) differentiation of gaseous tritium and THO vapours by means of the desiccator assembly;
(d) continuous catalytic combustion of ^{14}C and tritium-labelled organic compounds in air to form CO_2 and THO.

Monitoring of Radioactive Contamination of Surfaces

National standards treat of radioactive contamination of surfaces in terms of particle output from a unit of area regardless of radiation energy[6]. To comply with this, the efficiency of our portable instrument hardly depends on radiation energy. Detection is effected by a four-section gas-flow proportional counter with an open window (Fig.3). Anode wires are designed to run parallel to the examined surface. The outside cathode made of 0.6x0.6mm mesh brass net is separated by a 2mm clearance from the surface.

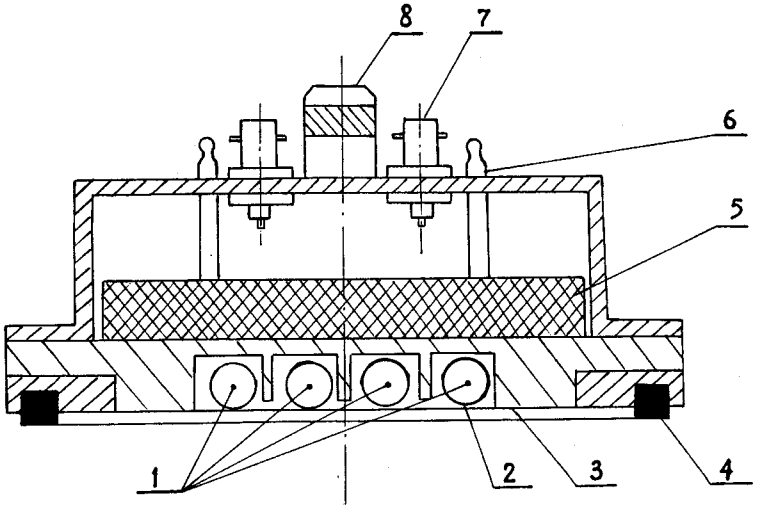

Fig.3. Surface contamination detector.
1-anodes; 2-insulators; 3-net; 4-rubber ring; 5-lead disk; 6-gas connection pipe; 7-plug connector; 8-grip.

The sensitivity area is 40 cm^2. The detector comprises a transistor pre-amplifier of signals. The gas (methane) flow rate is less than 0.5 l/min.

The measuring panel incorporates an integral discriminator,

a rate meter, a pointer indicator and a power supply.

Solid sources of beta radiation of tritium, ^{14}C and ^{137}Cs are used for calibration purposes. Plateaux of 200 to 300 V length with a slope of 5%/100 V have been obtained. The counting efficiencies at the point of operation are: 70% - for tritium, 80% - for ^{14}C and 90% - for ^{137}Cs. The proximity of the above values is due to the peculiarity of open-window counter operation, which detect a great portion of ionization electrons released in the gas layer bounded by the examined surface and the cathode The counter background is 6 cps. The instrument reliably detects contamination under 0.01 of the maximum permissible levels for operation surfaces (2.000 particle/cm^2 min[6]).

Measurement of Tritium and ^{14}C Content in Human Body

The internal contamination of the human body induced by such beta-emitters as T and ^{14}C may be determined only indirectly on the basis of the results of biological sample assays.

T and ^{14}C concentrations in the body liquid samples and exhaled air were measured by scintillation and gas counters. Samples of urine, saliva, blood and exhaled vapour condensate were mixed with a liquid scintillator containing 8 g PPO + 0.2g POPOP + 100 g naphthalene per 1 l dioxane. Different methods of preparation of tritium-containg samples with respect to the degree of purification, such as distilled, activated coal-treated (urine only) and untreated samples, were tested. Precipitates were separated by filtration.

The beta-radiometry of the samples was carried out on a scintillation coincidence counter with two venetian-blind photomultipliers ФЭУ-81А A 30 ml cuvette for samples is made of Teflon and provided with quartz windows. The electronic recording system selects time-coinciding (within 100 nsec) pulses fed from the two phomomultipliers and analyses them with respect to amplitude by means of a differential discriminator with a threshold ratio of 10:1. The measurements were carried out under balance conditions. The instrument was calibrated with the aid of a solid emitter of X-rays simulating tritium radiation[7].

The best sensitivity is ensured in measuring samples containing 20% water or 8 to 10% urine, with the tritium counting efficiency being 12 to 15%; the counter background is 50 cpm. The sensitivity threshold to tritium in water is $3 \cdot 10^{-9}$ and in untreated human urine - $7 \cdot 10^{-9}$ Ci/l, respectively (measurement time -30 min; relative error -25%).

An analysis of the instrument background shows that 65% is contributed by internal processes occurring in the photomultipliers and generating light impulses; 30% - by radioactive and cosmic radiation, and 5% - by random coincidence of dark current pulses and the phosphorescence of samples.

The minimal detectable concentration of tritium is four orders below the "initial levels of the examination" of personnel in contact with tritium oxide[8]. The scintillation counter sensitivity to ^{14}C concentation in urine or water samples is 10^{-9} Ci/l which is quite sufficient for the purposes of industrial dosimetry.

Another modification of the indirect dosimetry method is provided by the measurement of THO vapours and $^{14}CO_2$ in exhaled air by means of gas monitors of types B and C. The air to be assayed is continuously sampled from a through-flow vessel where exhaled air comes. The CO_2 counter is capable of measuring ^{14}C concentrations up to $2 \cdot 10^{-12}$ Ci/l in exhaled air (CO_2 content -3%). This makes it possible for ^{14}C excretion to be reliably monitor-

ed during one week after one "initial level of the examination" of $NaH^{14}CO_3$[8] incorporation. During the first days after exposure this level can be detected by instrument B as well. For a similar THO incorporation the body fluids tritium concentration is 35 μ Ci/l[8], whereas this parameters for exhaled air is $1.5 \cdot 10^{-9}$ Ci/l at the body temperature. However, when air is transferred to the detector, the air temperature is equalized to that of the device parts. This leads to the condensation of some vapours and the residual concentration of THO in the assayed air is determined by the ambient temperature. Thus, $5 \cdot 10^{-10}$ Ci/l of exhaled air corresponds to the "initial level of the examination" at 20°C. This concentration is reliably detected by counter B.

So, the above complex of instrumentation and techniques guarantees an all-round monitoring of exposure conditions of operation in contact with unshielded tritium and radiocarbon compounds Depending on the process used and scale of production of different radioactive substances, the dosimetrical service may be supplied with some of the instruments of this complex or a suitable combination of them. The counters have been tested for a long period of time at the works of manufacturers specializing in the large-scale production of various radionuclide items, such as labelled organic and inorganic substances, luminous compounds and devices, metallic targets with tritium, etc. The test results have shown the described equipment and methods to comply with the requirements of day-to-day dosimetry.

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