Instruments and Methods for Measuring ^{14}C
(*a Review*)

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Received June 17, 2021; revised June 28, 2021; accepted July 16, 2021

Abstract—The main methods for measuring ^{14}C based on a Libby counter, a proportional gas-filled counter, liquid scintillation counting, and accelerator mass spectrometry are considered. The measurement features, advantages, and disadvantages of each presented method are described. The radiocarbon concentration in annual rings of a pine tree grown in Akademgorodok of Novosibirsk has been measured using the accelerator mass spectrometry method. The measured ^{14}C concentration in the samples is in the range from 95.4 ± 0.2 to 191.5 ± 2.2 pMC.

INTRODUCTION

^{14}C is a radioactive carbon isotope with an atomic nucleus containing six protons and eight neutrons. There are several factors that make obtaining metrologically substantiated data on the radiocarbon activity in a sample a relevant task. First, the use of ^{14}C in radioisotope dating makes it possible to establish the age of a biomaterial sample of up to 60 000 years [1–6]. This dating technique is most often used in archeology, glacial geology, atmospheric physics, glaciology, and other sciences, but it can also act as a tracer in various natural processes. Second, ^{14}C is used in medicine, e.g., to perform breath tests for Helicobacter Pillory [7–9]. Third, ^{14}C contributes to the exposure of population, as it is present in the atmospheric emissions from nuclear reactors of various types and from fuel reprocessing plants [10, 11].

^{14}C was experimentally discovered in 1936 by W.E. Burcham and M. Goldhaber, British physicists who irradiated ^{14}N nuclei in a photographic emulsion with slow neutrons according to the ^{14}N$(n, p)^{14}$C reaction [12]. The half-life of ^{14}C is 5700 ± 30 years [13]. ^{14}C is a β emitter; it decays into ^{14}N:

$$^{14}_{6}\text{C} \rightarrow\, ^{14}_{7}\text{N} + e^{-} + \overline{\nu}.$$

The end-point energy of β particles is 156 keV, and their mean energy is 45 keV [14].

^{14}C is has both natural and anthropogenic origin. The natural origin is based on the interaction between the ^{14}N isotope contained in the Earth's atmosphere and neutrons produced in the reaction between cosmic rays and various substances of the atmosphere:

$$^{14}_{7}\text{N} + {}^{1}_{0}n \rightarrow {}^{14}_{6}\text{C} + {}^{1}_{1}\text{H}.$$
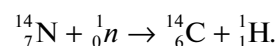
Approximately 1.4×10^{6} GBq of radiocarbon are annually produced in this way; the total amount of ^{14}C in the atmosphere is estimated at 1.4×10^{8} GBq. The largest amount of ^{14}C is contained in the oceans: it is approximately 1.0×10^{10} GBq [15].

Nuclear tests in the period from 1945 to 1980 were an anthropogenic source of radiocarbon; they were responsible for a release of 2.2×10^{8} GBq of ^{14}C into the environment [16]. Emissions from nuclear fuel
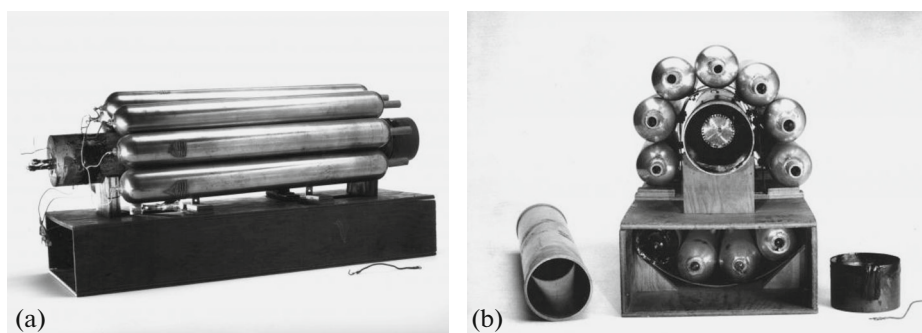
Fig. 1. The external appearance of the Libby counter with the lid (a) closed and (b) open [21].

cycle enterprises are another anthropogenic source of radiocarbon. During the operation of a nuclear reactor, ^{14}C is produced mainly by neutron activation of ^{17}O, ^{14}N, ^{13}C, and, to a lesser extent, of ^{15}N and ^{16}O, which are present in fuel elements, structural materials, moderator, and coolant; ^{14}C is also produced by triple fission reactions of uranium and plutonium in nuclear fuel [17]. According to estimates in [18], approximately 1.1×10^6 GBq of ^{14}C is annually generated by operating nuclear power plants around the world; 1.1×10^5 GBq of this amount of ^{14}C is emitted in gaseous form into the atmosphere per year, while approximately 3.7×10^5 GBq of ^{14}C are emitted per year from nuclear fuel reprocessing plants in both gaseous and liquid forms. The method for generating ^{14}C at nuclear power plants depends on the nuclear reactor type [19] or, more precisely, on the degree of uranium enrichment in the fuel and on the concentration of oxygen and nitrogen in the fuel, structural materials, moderator, and coolant.

Measuring the activity of ^{14}C is fraught with difficulties in detecting β particles, since their maximum energy is 156 keV. Conventional solid-state (scintillation and semiconductor) detectors are unsuitable for β particles with this energy. In addition, self-absorption of low-energy β particles makes it impossible to use large-volume samples for measurements. In this regard, methods for measuring radiocarbon should feature high detection efficiency for low-penetrating β particles, a low background component of instruments, and high stability of the equipment.

In this paper, we consider the main methods for measuring ^{14}C, starting with the first gas-discharge counters and finishing with the modern method of accelerator mass spectrometry. The advantages and drawbacks of each method are presented and the practical application of the accelerator mass spectrometry method is described.

METHODS FOR ^{14}C MEASUREMENTS

The Libby Counter

W. Libby, an American physicist, developed the first device for measuring ^{14}C in 1946. He used a special system in which a Geiger–Müller counter with brass walls was surrounded by other counters, which were connected in anticoincidence with it (Figs. 1 and 2). A pure carbon (soot) sample was deposited on a thin-walled cylinder and enclosed in a central counter. The use of external counters has made it possible to avoid the registration of false pulses, and the central counter detected only the particles that were emitted by the counting sample [20].

This method is not used today, since it has a number of disadvantages: low detection efficiency (~5.5%), difficulty in obtaining chemically pure carbon (free from radicals and impurities), and the high probability that a prepared counting sample will be contaminated with airborne radioactive substances.

Proportional Gas Counters

In the 1950s, the method proposed by Libby was replaced by a method for measuring ^{14}C using gas-filled proportional counters. Their main feature was that the $^{14}CO_2$ gas released from the sample was placed directly into the sensitive volume of the detector. Two concentric tubes with 11 equidistantly placed anode wires (Fig. 3) were used instead of the ring of Geiger–Müller counters [23]. The advantages of this design over the ring of individual counters are the stability of
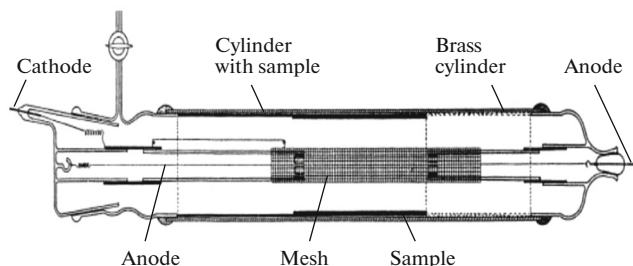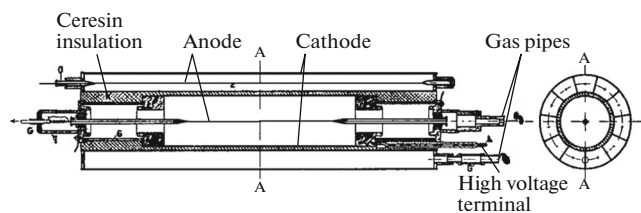

Fig. 2. The design of the Libby counter [22].

Fig. 3. A gas-filled proportional counter [23].

Fig. 4. The functional diagram of a liquid scintillation spectrometer [25]: (*PMT*) photomultiplier tubes and (*HV*) high voltage.

the system and the same trigger potential for all counters.

Cathodes of a proportional counter were most often made of materials containing a small amount of radioactive impurities, such as copper or stainless steel. In order to reduce the probability that a part of the sample will be deposited on the chamber walls, the inner cathode surface was coated with substances that did not react with gases. Either large counters (up to 10 L in volume) or counters with a higher working gas pressure (2−10 atm) can be used to increase the sensitivity of the method.

Although proportional counters have such advantages as high detection efficiency and low background, they are not free from drawbacks. It is difficult to eliminate radioactive impurities from the counting gas and remove deposited sample particles from the chamber walls for subsequent measurements. In addition, the use of large-volume proportional counters leads to an increase in the size and weight of the shielding components.

A 1-L counter filled with CO_2 at atmospheric pressure typically requires 0.5 g of carbon in a sample. This requirement is critical for many areas of research, especially for archeology, where milligram samples are usually available. Therefore, the development of micro-counters with a volume of a few milliliters, for which a few milligrams of carbon in the sample were needed, became a relevant task. The disadvantage of such counters was the data-acquisition time, which was as long as 1 month. Micro-counters have not been widely used due to the rapidly developing method of accelerator mass spectrometry [21].

The Liquid Scintillation Counting Method

The development of the liquid scintillation counting method began in the 1950s almost simultaneously with the development of the gas counter method. The principle of liquid scintillation counting is based on the detection of scintillations produced by charged particles (in our case, β particles). A liquid scintillator is a mixture of a solvent, a scintillator proper, and a wavelength shifter. The intensity of scintillations is proportional to the particle energy, which allows an electronic system to select the range of particle energies that is interesting to the researcher.
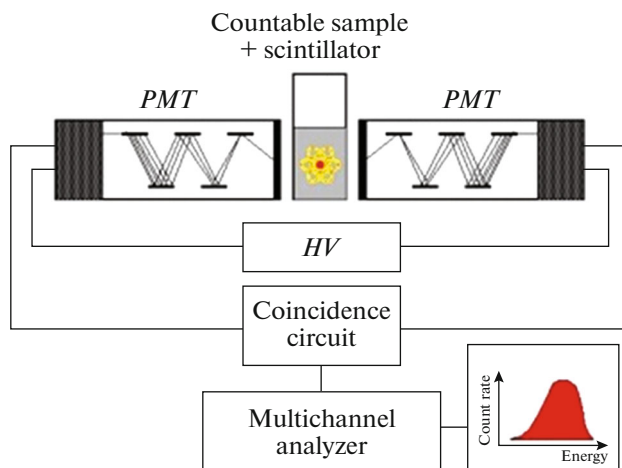
Radioactive samples and the scintillation mixture are enclosed in small transparent (often glass or plastic) vials, which are then placed in front of photosensitive devices, most often, photomultiplier tubes (PMTs). Many instruments have two PMTs connected in a coincidence circuit (Fig. 4), which cuts off the PMT noise. The counting efficiency depends on the measurement technique; for ^{14}C, it can reach a level of 95% or more. The disadvantages of the method include quenching effects, i.e., a number of effects that lead to energy losses during conversion of the decay energy into electrical pulses of a PMT [24].

The Accelerator Mass Spectrometry Method

The above-mentioned ^{14}C measurement methods imply detection of β particles from only a small number of decaying carbon atoms. Accelerator mass spectrometry, which appeared in the 1970s, provides individual counting of each radiocarbon atom in a sample. The operation of an accelerator mass spectrometer is based on measuring the intensity of ion beams with different masses. The sensitivity of this method makes it possible to separate a beam with an ultralow relative intensity (there are approximately 10^{12} [12]C atoms per ^{14}C atom) from neighboring beams with a very high intensity, such as ^{13}C and ^{12}C beams.

Since the half-life of ^{14}C is 5700 years (5568 years are used in radiocarbon dating calculations), merely one decay happens in 4 s in 1 g of carbon extracted from modern wood. Obtaining an accuracy of 0.1% requires acquisition of 10^6 decay events, which takes 4×10^6 s (almost 46 days). For 100 g of carbon, the time is reduced to 11 h. Accelerator mass spectrometry analysis calls for only 1 mg of carbon; this carbon will be converted into charged ions with an efficiency of 2%, and ^{14}C ions will be counted [26].
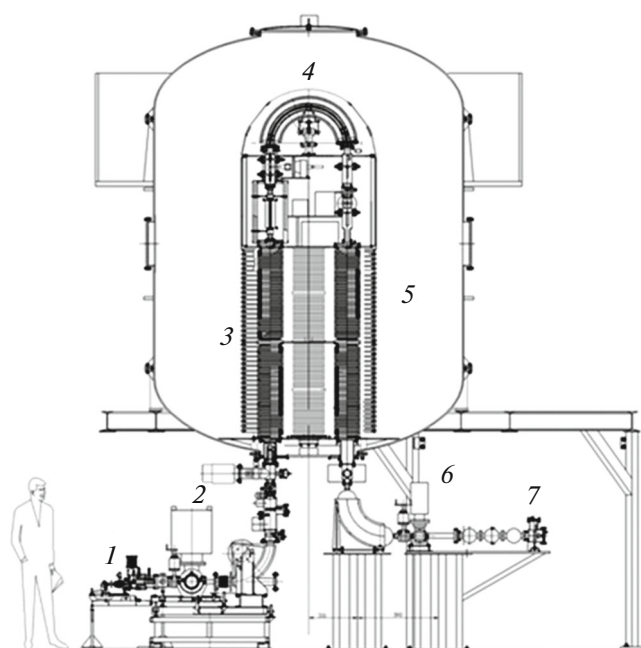
Fig. 5. The diagram of the accelerator mass spectrometer at the Budker Institute of Nuclear Physics: (*1*) ion source, (*2*) bending magnet, (*3*) the first accelerating tube, (*4*) magnesium vapor target, (*5*) the second accelerating tube, (*6*) magnetic-field filter, and (*7*) thin-film time-of-flight detector.

Figure 5 shows a diagram of an accelerator mass spectrometer that was created in 2011 and commissioned by the Budker Institute of Nuclear Physics in Novosibirsk [27]. The sensitivity of this setup is 10^{-15} (^{14}C/^{12}C).

Carrying out radiocarbon analysis using the accelerator mass spectrometry method consists of the following stages.

1. Generation of an ion beam. First, 23 graphitized samples are placed in a drum at a distance of 7 mm from each other. A small steel container filled with liquid cesium is heated to 600°C. Cesium vapor produced thereby is incident on a tantalum ionizer heated to 100°C and then, being accelerated by a voltage of 8 kV, hit the sample. A negative beam of ^{12}CH$_2^-$, ^{13}CH$^-$, and ^{14}C$^-$ ions with an energy of 25 kV is generated at the exit from the ion source. The negative charge state of the initial beam allows one to eliminate ^{14}N$^-$ ions, which are extremely unstable.

2. Focusing of the ion beam and its rotation through 90° by the magnetic field, as a result of which the beam enters the first accelerating tube, where it is accelerated to a voltage of 1 MV. The beam then hits a magnesium vapor target.

3. Ion charge exchange in the magnesium vapor target to the 3+ charge state, as a result of which molecular ions are eliminated. The resulting beam of ^{12}C^{3+}, ^{13}C^{3+}, and ^{14}C^{3+} particles is bent through 180°

and enters the second accelerating tube, where it gains an additional energy of 3 MeV. The beam of particles is bent by the magnetic field, their selection by the mass and charge is performed, and the ^{14}C^{3+} beam hits the counters.

4. Detection of ^{14}C^{3+} ions by a thin-film time-of-flight detector consisting of electrostatic and cycloidal sensors. There is a film in the sensor; in passing through it ions cause the emission of electrons. Electrons are then incident on a microchannel plate, in which their multiplication occurs. At this time, the time of flight between the sensors is measured.

An ion mass spectrum, i.e., the dependence of the measured current on the ion mass, is the final result of the analysis [28].

MEASUREMENTS OF THE ^{14}C CONCENTRATION IN ANNULAR RINGS OF A PINE TREE

Accelerator mass spectrometry makes it possible to determine the exact age of samples in archeology, geology, oceanology, and other scientific disciplines. In addition, it can be used to measure the ^{14}C concentration in various environmental objects in order to carry out both radioecology examination [29] and retrospective estimation of the radiocarbon concentration in the atmosphere by measuring the ^{14}C activity in tree rings [30].

Data on background values are needed for estimating the additional contribution to the natural radiocarbon content during operation of nuclear facilities. The accelerator mass spectrometry method is best suited for this purpose, since it is the most sensitive and precise.

Since cellulose, the main component of wood, does not interact with environmental carbon and the half-life of ^{14}C is 5700 years, each annual ring of a tree contains information about the ^{14}C activity in the atmosphere for each year during the entire life cycle of the tree. A retrospective estimate of the radiocarbon content in the atmosphere at different periods of time was obtained by studying wood rings in Akademgorodok of Novosibirsk, i.e., in a region located at a certain distance from operating nuclear facilities. An analysis of the annual rings of a pine that grew in Akademgorodok is shown in Fig. 6. The ^{14}C content is measured in terms of Percent Modern Carbon units (pMC), which were adopted in the second half of the 20th century. A value of 100 pMC = 227 Bq/g C corresponds to the hypothetical specific activity of ^{14}C in the atmosphere in 1950 without any human impact [31].

The age of the pine that grew in Akademgorodok was 113 years. The sampled core was divided into annual rings, from which cellulose was extracted. The cellulose extraction technique was as follows: a ring fragment was poured in 13 ml of a solution of catalyst
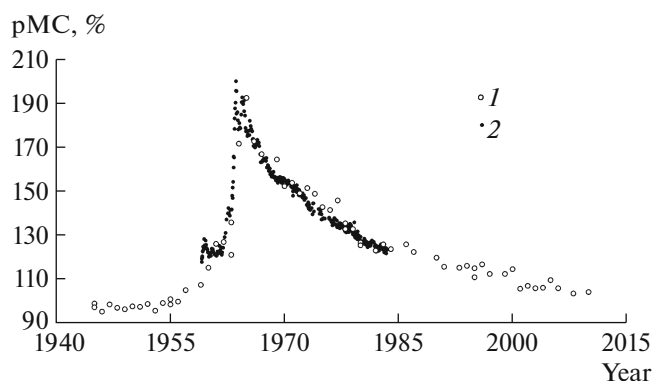
Fig. 6. The ^{14}C concentrations (*1*) in the annual rings of the pine tree that grew in Akademgorodok of Novosibirsk and (*2*) in the air at the Vermunt station, Austria [33].

and oxidizer and kept for 70 min at a temperature of 85°C. An oxidizing solution for six ring samples with a total mass of up to 1.5 g was prepared by dissolving 2.25 g of sodium tungstate dihydrate (analytical grade) in 38 ml of distilled water; 1.8 ml of concentrated sulfuric acid (high purity grade) was then added dropwise, after which the resulting yellow precipitate was dissolved by adding 40 ml of a 33% hydrogen peroxide solution (high purity). The thus bleached cellulose fibers were washed and dried. The resulting cellulose was completely burned and converted into graphitelike carbon at an absorption-catalytic facility that was designed to obtain targets for accelerator mass spectrometry [32]. One graphite target was produced from 5−7 mg of purified cellulose. The obtained samples were then analyzed using an accelerator mass spectrometer. According to Fig. 6, there has been an almost two-fold increase in the concentration of ^{14}C since the 1950s; this was caused by nuclear tests in the period from 1945 to 1980. Such an increase is characteristic of the entire northern hemisphere, which is corroborated by the data on the ^{14}C activity in the ambient air at Vermunt station in Austria [33].

CONCLUSIONS

Methods for measuring ^{14}C have been rapidly developed since the 1950s. The gas-discharge counter created by Libby revolutionized dating and had a huge impact on the development of many sciences, mainly archeology. The radiocarbon dating method made it possible to date objects with higher accuracy compared to earlier methods (e.g., stratigraphy) and, in addition, to compare and synchronize events at large distances in time.

The Libby counter, into which a solid carbon sample (soot) was placed for measurements, was quickly replaced by a gas-filled counter. This is explained by its drawbacks: the low detection efficiency, the laborious

sample preparation process, and the probability that a sample will be contaminated with airborne radionuclides.

Gas-filled proportional counters, in which a sample was placed in the form of a gas (most often CO_2), helped to improve the measurement sensitivity. However, the problem of selecting detector materials that should not contain radioactive substances was added to the problem of sample preparation (the transfer of a sample into a gaseous state).

Liquid scintillation counting and accelerator mass spectrometry are the most popular methods today. The first method is well suited for high-volume and high-activity samples. In Russia, the liquid scintillation counting method is widely used mainly for monitoring ^{14}C in emissions from nuclear facilities [34] and in the environment [35]. The quenching phenomena that reduce the detection efficiency are the disadvantages of this method. Accelerator mass spectrometry is the most sensitive of all the presented methods; it directly counts ^{14}C atoms in a sample. The drawback of this method is its cost. The analysis of ^{14}C in annual rings of a pine tree grown in Akademgorodok of Novosibirsk confirmed the published data on of the ^{14}C activity in the atmosphere throughout the northern hemisphere.

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Translated by N. Goryacheva