

UTILIZATION OF RADIOISOTOPE FOR INDUSTRY

by

Hiromu Fushimi

Prof. Waseda University, Japan.

I. Fundamental Review

I have the honor to explain you today about the R I (Radio Isotope) Application for Industry in Japan through the good offices of President Choi & other friends.

We have presented 60 papers in the 9th Japan Conference on Radio Isotopes at Tokyo in the middle of May this year.

Conference papers have presented from many divisions; Science, Engineering, Medicine, Biology, Agriculture, Shielding, Basic Research, Problem of X-Ray Fluorescence Analysis, Radiatry, etc.

The papers presented at the engineering division were as follows.

Actual R I application reports	12 papers
R I experiment reports	14 papers
Chemical Analysis	13 papers
New Application Methods of R I	2 papers

Now, I am going to explain the summaries of some of these papers.

1. Activation Analysis in Japan

Works on activation analysis are considered to have started in 1957 when the first reactor JRR-1 became available. The research on activation analysis in Japan has been chiefly concerned with the technique of neutron absorption process, but new techniques R I sources are being developed today.

A commercial small-type neutron generator producing 14 MeV neutrons has been popular since 1963. On the other hand, R I source generators have made it possible to conduct a comprehensive analysis of cement elements and have been improved by introducing direct readout system which makes rapid calculation of analytical results possible.

2. Isotope Applications in Japan Industry

For the past several years the industrial applications of radioisotopes have been increasing at a fairly rapid rate. The statistics says that the number of gamma radiography equipments and isotope instruments used in industrial firms is on a sharp increase. The total number as of March 1968 is further added by 460. if those used in educational facilities and research institutes are included. In addition, it should not be overlooked that smoke detector using alpha sources has just recently been put into practical use in tall buildings as a fire-monitor.

2. 1 Gauges

Highly reliable radioisotope instruments are widely used as signal sources for the central control system of an automated plant. Especially, the level gauges of both on-off output and continuous output types are very popular.

Among the radioisotope gauges, level gauge and density gauge have been in practical use since more than ten years ago. Practical use of moisture gauge, snow meter and radioisotope instrumental analyzer has been delayed, however, for several years. Recent growth in the number of these devices in use is so remarkable. This is mainly due to the technical improvements in their sensitivity and accuracy, and to much improved stability over an extended time period.

With regard to the thickness gauge, a precision gamma thickness gauge capable of measuring the profile of steel plate of 0.3 to 4.0 mm thick and 1600 mm wide has been developed, where the error is 5 μ m in case the measuring time lasts for 40 sec. The gauge is now in routine use and contributing very much to the rolling technique of steel plates.

In addition, the use of gamma thickness gauge is also spreading widely. Particularly, the gauge using Am-241 source is suitable to measure medium thickness because of the low energy, and hence its wide use in this country is highly expected in the near future.

(1) Level gauge

The use of level gauge extends to the most fields of industry. As well known, the level gauge is classified into two types, ON-OFF type and continuous indication type. In the earlier period the former had been used much more than the latter, because of its superior accuracy in indicating level. However, as the accuracy of the continuous indication type has recently been improved to the extent of ± 2 to 5%, it is increasingly replacing the ON-OFF type.

As to the continuous indication type gauge, the linearity between level and response of the detector is essential. For this purpose, the measures have been so adopted as to have

many gamma sources such as Co-60 or Cs-137. Further, in connection with the ON-OFF type gauge, the so-called fail-to- safe system (radiation relay) has been introduced.

(2) Density and moisture gauges

A remarkable increase in their application to the measurement of density and moisture of soil is the recent tendency. Consequently, several achievements in research and practice concerning these kinds of gauges will be mentioned in the following.

Density gauge: In measuring the density of soil or coal using the depth type density gauges, there can be an incidental error and it is believed to have been caused by the non-uniformity of density distribution in sensitive volume which is in the vicinity of the measuring head and by the void in the uniform gap along the access tube which effects error in such a rate as 2% per 1 mm gap length. With the surface type density gauge, the shape of leaden shield intervening between the source and the detector was elaborated and there by the gauge has turned out to be much improved.

Moisture gauge: When a matrix moisture content which is to be measured contains a considerable amount of elements having large absorption cross section for thermal neutrons, such as chlorine, the output of neutron moisture content due to such a condition, a method of correcting the results obtained is proposed. However, it is necessary to know the moisture content in the various matrix by use of R I Am-Be neutron source.

2.2 Radiography

Radiography is now a very popular technique in this country. Among the radiographic techniques, however gamma radiography is not yet in full use. All point sources for gamma radiography have been imported to date and they are mainly Co-60 of 1 to 3 curies. Besides Co-60 sources, Cs-137 sources are a little less than and Tm-170 and Ir-192, sources are found to be very much used in various fields of industry such as mechanical, metallurgical, electriccal, ship-building and civil engineering.

Electron linac, although being no radioisotopes, has been used extensively for radiography of thick plate welds. The linacs developed so far are small in size and thus portable but are of high output, consequently making quick radiography possible. The characteristic of an electron linac of 500 R/min at 1 mm is such that its X-Ray output is kept almost constant with the angle of elevation by adjusting the frequency of magnetron. Its performance covers JIS, ASME and MIL sepecifications for steel thicknesses of 30-450 mm, 100-300 mm and $1\frac{1}{2}$ -6", respectively. Inspection operation can be done within 10 min with a steel plate 350 mm thick.

2.3 Instrumental analysis

Absorptiometry: The radioisotope instrumental analyzers making use of absorptiometry, such as coal ash meter and sulfur meter, are now proving their usefulness through the routine use. Two R I source ash meter may be used at the online for the mixing ash control. And the sulfur meter introduced here is a continuous detecting apparatus.

The coal ash content is measured by an Am-241 source. Another source Cs-137 is used to observe the effect of the bulk density of the sample at the time of measurement and to correct the result of ash measurement by Am-241.

The principle is based upon the following equation, in which A means Cs-137, and B means Am-241.

$$\mu_B = \frac{\log I_{oA} - \log I_A}{\log I_{oB} - \log I_B} \mu_A$$

The results of the present study clarified the relation between the chemical composition of coal and the absorption coefficient of it. If the build-up factor is obtained for each coal seam, it will be useful in case estimating the coal quality from the penetration of radiations from two sources is very effective especially for the rapid determination in routine use. The progress of coal ash measurement in this direction will contribute to the optimization of coal treatment process.

For sulfur, by simultaneous use of X and beta sources, it is possible to eliminate errors due to density change with time. Its performance is such that the sulfur concentration can be measured up to 4% by weight with error of ± 0.05 . Thus, it plays a role as on-line stream analyzer and is now in routine use for monitoring the sulfur content of blended fuel oils.

X-Ray fluorescence analysis: This technique is now well developed and being put into practical use. An X-ray fluorescence analyzer for determining Cr and Mo contents in alloyed steel has come into practical use. With the aid of the instrument, it is easy to grade alloyed steels in accordance with the content of Cr or Mo ranging from 0.3% to 0.5%, with each measurement time requiring less than 45 seconds. The principles of operation of portable X-ray fluorescence instrument are the same as those of conventional equipment in which X-ray is generated electronically and the secondary fluorescent radiation characteristic of the element sought is selected and measured with a crystal spectrometer. In the case of the portable instrument, however, a radioisotope X-ray source replaces the generator and a pair of balanced absorption filters substitutes the diffracting crystal. Each element has a characteristic X-ray spectrum which bears a simple relationship to its atomic number, namely where λ is the wavelength of an emission line and Z the atomic number.

$$\frac{1}{\lambda} \propto Z^2$$

The successful use of balanced filters for energy selection depends on the availability of suitable low-energy X-ray and γ -ray sources. Theoretically, the optimum excitation of fluorescent radiation is achieved by the use of a monochromatic source with its characteristic energy just above that of the absorption edge of the element to be determined. However, not many of such sources are available and bremsstrahlung sources that emit a continuous spectrum of X-rays are used instead. Such sources tend to produce a higher background of scattered or scattered radiation. Suitable sources presently available are listed in Table I. Filter pairs suitable for selecting the fluorescent radiation of the elements that have so far been successfully measured with portable instruments are made either of metal foils or of a convenient compound encapsulated in a plastic or epoxy resin. The excitation sources and filter pairs appropriate for a few metals are indicated in Table II.

Experiments are currently being carried out by members of the staff of the Geochemical Division of the Institute of Geological Science to determine a number of additional elements. These are listed in Table III.

Portable instruments can determine all the main metals such as titanium, chromium, manganese, iron, cobalt, nickel, copper, zinc, tin, zirconium, niobium, molybdenum, antimony, barium, tantalum, tungsten, lead, bismuth, and uranium as well as several non-metals.

Activation analysis: Fast neutron activation analysis is coming to a common technique in industry. Under such circumstances, developed is a 14 MeV neutron generator of 10^{10} n/sec and compact size, and it has made special improvements in a few points in a few points, that is, longer life attainable by adopting an eccentrically rotating tritium target and its neutron yield being monitored by a plastic scintillator coupled to C R circuit. By using this machine several studies were carried out as follows: (1) In determining oxygen in steel, the error caused by segregation of oxygen was lowered by rotating test piece down to 1/5. (2) In order to make on-line analysis in production of barium ferrite the technique was applied because of its rapidity, where nuclear reactions, Ba-138 ($n_f, 2n$) Ba-137m and Fe-56(n_f, p) Mn-56, are available with respective accuracies required for the process control. (3) When sulfur in oil is analyzed by this method based on a reaction S-34 (n_f, p) S-34, the analytical value is affected by many interfering elements contained in oil.

In addition to the above, many industrial application research works are under way by means of reactor thermal neutron activation analysis.

Tracer techniques: Activable tracer techniques are popularly used in our country, particularly for field experiments and process analysis in factories. In connection with this,

activation analysis procedure requires rapidity and simplicity, excluding chemical treatments. Several spectroscopic research works on this line were being carried out.

Besides the above, it should be noted that some tracer techniques taking advantage of natural activities are now developed for the measurement of the pulp flow in flow in flotation cell, agitation CN ion, phenomenon of the cyanidation plant, action of flotation reagent and cement material motion in the kiln, etc.

Especially, kiln test by use of RPI have achieved much results in Poland and now, it has advanced for hydroengineering problems.

Heat and power use of mass radioisotopes: In Japan the first radioisotope powered generator was made in 1968. This is a thermoelectric power conversion type using ^{90}SrO as a heat source which was imported from the USA., and Bi_2Te_3 thermoelectric elements. The shielding system adopts the double biological shielding, with the primary surrounding, with the primary surrounding the fuel capsule being made of tungsten and the secondary constituting outer wall being made of lead, and the thermal shielding constitutes intervening layer. Its performance is as follows: 180°C and 50°C at hot and cold junctions respectively, 3 W of electric output at 9 V, 3.5% of efficiency and 450 kg in total weight. A new R I generator is under trial making in Japan.

Further development of this kind of use is being expected for the purposes of not only space development but also ocean development and for terrestrial uses.

In Japan, We have tried to make R I generator for the abovementioned purposes by use of ^{90}Sr source. But we have met some troubles regarding shielding system, I heard.

II. Development in Several Industrial Fields

1. Iron and steel industry

In this field the isotope techniques are very frequently applied to development research on new techniques and products and to process analysis of the early production stages. A well elaborated method was used for estimating the amount of residuat molten pig iron in blast furnace, and to observe the eroded condition of hearth bricks, under an assumption that there is some turbulenc in the molten pig iron. By measuring the activity of radioactive SiO_2 , the oxygen content in molten steel can be measured quantitatively.

To mention some other notable appliactions :

- (1) Study on segregation of in steel ingot
- (2) Study to trace the behavior in air bath during the heating of steel

(3) Study on corrosion cracking of high tension steel by the effect of sulfide

2. Non-ferrous metals industry

The use of radioisotopes in non-ferrous metals industry is also popular, as much as in iron and steel industry.

A basic study on the influence of such factors as casting speed, temperature cooling of mold and ingot itself on solidification process of copper and its alloys in the continuous casting process was carried out systematically, using Au-198. Radioisotopes are also used to investigate metal surface phenomenon by means of techniques of labeling K C N with C-14 on the washing efficiency and thermal diffusion behaviour of electrolyte in plating processes. Investigation by use of Cu-64, Mn-53, and Ga-72 clearly revealed the mechanism of corrosion caused by impurities in aluminum alloys.

In alumina manufacturing facilities of Bayer process, by using radioactive tracers, the process analysis was made successful with respect to flowing behaviours of solution, solid grains and powder in the processes of bauxite extraction, aluminum hydroxide precipitation, and moreover flow rate and flow pattern of metal and electrolyte bath in aluminum reduction cells were determined.

By means of 14 MeV fast neutron activation analysis, the oxygen contents of aluminum ingots were determined. The limit of the determination was 3-4 P.P.m. and the results of analysis were $20-30 \pm 3$ ppm.

3. Chemical industry

In the chemical and fiber industries, radioisotope instruments such as level gauge and thickness gauge are used and the demands for them are ever increasing. There are considerably many examples in which radioisotopes are utilized for industrial analysis, which, however, are mainly radioactivity analysis. More recently, gas chromatography is popular, in which tritium is used as a tracer.

The first case is an experiment of measuring distribution of traveling velocity in a cement kiln. 90 mc of La_2O_3 labeled with La-140 prepared by irradiation was mixed with 10 Ka of raw material, and the mixture was injected in pulses through the feed shoot of the material. The travel of radioactivity was followed with 18 scintillation probes by measuring gamma radiation penetrating through the kiln wall. Sampling was also made at the outlet. The information obtained included traveling time, distribution of velocity, longitudinal dispersion, movement in tangential direction and estimation of the points where dust originates.

In the fiber industry, in the study of the so-called yellowing or blackening of cleanliness after washing, by use of C-14 labeled fatty acids and cholesterol as a tracer, it was made

clear that polyester fibers are more easily cleaned than cotton and the glycerides of higher fatty acid are hardest to clean. And also, C-14 labeled compounds were used to investigate the diffusion of stain in film.

There are various methods of measuring unevenness in yarn, however, it is difficult for the conventional methods to measure unevenness in blending fiber in yarn.

In oil and fat industry, activation analysis was applied for the determination of minute amounts of As, Fe, Co, Ni, and Zn in refined bean oil. And also, the contents of As in sunflower seed oil and rape oil were determined.

A new test method using neutron & natural γ and N-N logging was applied for the hydraulic fracturing test, in which glass sand containing boron was forced into a well as a stable tracer. A test using I-131 to reveal the flow behaviour of water in an oil-bearing formation and mineral deposits fields has been reported. These two are still at the stage of model experiments.

Oil logging has been executed with Am-Be neutron source since several years ago. An application of pulsed neutron generator for distinguishing salt water from oil just begun. These fundamental studies are much advanced now.

4. Machinery and electrical industries

The principal use is gamma radiography in these fields, but every kind of isotope technique is now used popularly. As most of development research works recently carried out in these fields were mentioned in the preceding chapter, only some typical works tracer and radiation curing techniques are advanced day by day. Using short lived radioisotopes, Cu-64 and Au-198, which were produced by gold in case of making them slide on steel ring lubricated with spindle oil.

One of the developments under way is radiat curing of paint by electron beam. Joint research by paint makers and users, and machine manufacturers is now being performed.

5. Civil engineering

Neutron moisture meter and gamma density meter are the most popular radioisotope instruments being used in civil engineering. The investigation by the Japan Atomic Industrial Forum points out that it is necessary to standardize the measuring procedures in order to stimulate the use of these instruments and the gamma penetrating density meter of surface type will be more widely used in the future.

Many limitations tend to be put on a large use of radioactive tracers in the field because of special conditions of the natural and social geography in our country.

Accordingly, an estimation of the amount of groundwater solely from pumping tests may give rise to error and a preliminary test using radioisotope flow meter is necessary. Now, we have made a new flow meter for measuring the velocity and direction of ground-water with radioisotope, which can be used in a small diameter boring.

III. Review of R I Techniques in other countries

Recently, industrial application of R I is expected to be more and more spread when it is proved to be more economical than the conventional measures.

I would like to present some topics of other countries on R I research processes in spite of considerable gaps among them. The USA, England, France have made top developments of atoments of atomic energy use. But the best radioiso tope application is made in the USSR.

I would like to explain some good works done in these countries. (slide):

- (1) England: Wantage Isotope Division
- (2) France: Saclay, Centred Eludes Nucleairesde
- (3) Italy: ENI
- (4) USA: Bookhaven Lab., Michigan University, Nuclear Chicago, Standard Oil Research Lab.
- (5) USSR: Inst. of Meakhanobr
- (6) Finland: Outokumpu Co. (Pyhasalmi Mill)
- (7) Sweden: Isotope Techniques Lab.
- (8) Poland: Inst. of Nuclear Research, Center for Nuclear Equipment, Institute of Nuclear Research, Hydroengineering of Polish Academy
- (9) Czechoslovakia: Institute for Ore Research

As you know from the pictures, many countries are making efforts to use radio-active energy for peaceful industrial application.