INDUSTRIAL APPLICATIONS OF THE KARLSRUHE COMPACT CYCLOTRON

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Summary

The Karlsruhe isotope production facility consists of a compact H-cyclotron and four sophisticated target stations. The cyclotron is variable in energy between 15 MeV and 42 MeV. Extracted currents up to 120 μA can be produced reliably. This facility is dedicated to the production of isotopes for medical applications and for the irradiation of machine part for wear measurements in industry and engineering research laboratories. The routine production of ultra-pure iodine-123 and rubidium-81 is discussed as well as the Rb-81/Kr-81m-Generator system. The radionuclide technique for wear measurements developed at KfK is presented and some examples are given.

Introduction

In the frame of a technology transfer project the KfK has over the past five years built-up an accelerator facility for the production of important radioisotopes for medical- and industrial applications. On the medical side the facility concentrates on the production of very short-lived, high-purity radioisotopes not available before in Germany. This program has now been carried out for two years at the Karlsruhe compact cyclotron, which is an H-machine variable in energy between 15-42 MeV.

It could be proved that a dedicated machine is capable of being very reliable, which is important particularly for the production of short-lived, radioisotopes like $I-123(t_{1/2} = 13.2 \text{ h})$, and $Rb-81(t_{1/2} = 4.6 \text{ h})$.

Radioisotopes for medical application 1.1 Ultra-pure iodine-123

The most interesting accelerator-isotope produced at present at the Karlsruhe cyclotron is iodine-123 (half-life: 13.2 hours), which has recently begun to replace the reactor-isotope iodine-131 (half-life: 8.05 days) used until now.

Iodine-123 has the following important advantages:

- lower radiation dose for the patient (a factor 60-100 less than for iodine-131).
- lower radiation dose for the hospital personnel,
- markedly better scintigraph quality and hence a higher diagnostic power,
- lower environmental contamination (a factor 5900 lower less than for iodine-131).

All the known iodine-123 production methods produce large amounts of undesirable contaminations in the form of iodine-124 or iodine-125. Both the isotopes possess disadvantages comparable to those of iodine-131.

With an aim to produce iodine-123 with as low an impurity as possible, a new production method via the Xe-124(p,2n)Cs-123 reaction was taken into operation.

Iodine-123 is created by the subsequent decay:

Cs-123 5.9 min > Xe-123 2.08 h > I-123

The production of ultra-pure iodine-123 requires a very highly enriched Xe-124 gas astarget material. In the procedure described here, Xe-124 with an enrichment



Fig. 1: Automatic production system for ultra pure iodine-123. The set-up is composed of a high pressure gas target with an automatic foil changer, a sophisticated beam diagnostic system and gas transfer and target wash-out equipment.

> 99.8 % (natural abundance 0.1 %) is used, with the consequence that the end product iodine-123 has only a contamination $< 5 \times 10^{-5}$ (12 h after EOB). As the cost of a litre of enriched Xe-124 is very high it is important to keep the target volume as small as possible and at the same time be able to control the beam power, 50 µA x 30 MeV = 1.5 kW, generated during irradiation. The target arrangement consists of a special entrance foil and safety volume, in order to prevent losses of the target gas. Because of the limited life-time of the entrance foil a robot was installed to carry out automatic foil changes. Apart from that an integral part of the target arrangement is a beam diagnostic system, aligned with the target, which makes sure that the production gas is always irradiated optimally by the 30 MeV proton beam (Fig. 1).

After an irradiation cycle the xenon gas is cryopumped out of the target and subsequently the iodine-123 deposited on the cooled walls is washed out by means of 40 ml of water. The aqueous iodine-123 solution is then transported to a hot cell via a stainless steel pipeline, where it is concentrated down to 0.5 ml by means of highly specialized ion exchangers (Fig. 2).

The essential advantages of this method are:

- highest purity of the iodine-123 produced
- high production efficiency (300-500 mCi/accelerator hour).
- low production costs by the operation of a relatively small accelerator facility and microprocessor-controlled production.

The arrangement, which has operated extraordinarily reliably right from beginning has a present capacity of 2-3 Ci iodine-123 per batch.



Fig. 2: Chemical unit for the concentration of the iodine washout from 40 ml to 0.5 ml (0.05 n NaOH). Four ion exchangers with a special resin are used. The loading and reloading of the ion exchangers is carried out by HPLC pumps which allow precise control of the flowrates. In-process measurement of the iodine activity is possible with small GM counters.

1.2 <u>The Rb-81/Kr-81m Generator System for Lung</u> Diagnostics

The radionuclide Kr-81m (half-life: 13 s), which is the daughter of Rb-81 (half-life 4.65 h), has gained increasing importance for lung function diagnosis over the past few years.

The essential advantages of this radioisotope compared to the other radioisotopes used in this field today are:

- minimal radiation dose for the patient and hospital personnel,
- static and dynamic ventilation studies,

- no exhaust problems and also no risk of contamination.

The Kr-81(p, 2n)Rb-81 reaction is used for the production. During irradiation the Kr-81 gas (pressure: about 20 bars) is contained in a pressure vessel, which is sealed by a thin stainless steel entrance window. The target arrangement is similar to the Xe-target.

The Rb-81, produced through the irradiation, deposits on the cooled walls of the target and is removed after the end of bombardment by means of sterile water. The Rb-81 is then pumped 15 m via a stainless steel pipeline to a hot cell located outside the irradiation room. There the Rb-81 dissolved in water is purged through a special ion exchanger which traps the Rb-81. This socalled generator is then fitted into a transportable lead shield and a complete quality test is carried out. The KfK, possesses a pharmaceutical manufacturing permit for the Rb-81/Kr-81m generator according to paragraph 13 AMG.

For the application of the generator in the hospitals it was imperative to develop a simple application system. The "Kryptovent II" system, which has been developed at KfK, is offered with the generators as a complete inhalations system.

1.3 Ultra-pure Rubidium-81 for IV-Injection

Ultra-pure Rb-81 represents a unique way of measuring an organ's blood flow using the relationship between the radioisotope Rb-81 and its short lived daughter Kr-81m (13 sec).

In contrast to the I-123 process via ultra-pure xenon described above, ultra-pure Rb-81 cannotbe produced in a single step; using a (p,2n) reaction a small impurity is always present due to the (p,n) reaction, which cannot be suppressed at all.

For this reason an electromagnetic isotope separator for routine production of isotopically pure Rb-81 for medical use has been constructed (Fig. 3).



Fig. 3: Total view of the isotope separator for the production of ultra-pure Rb-81. The ion source (surface ionization type) is shown on the righthand side, the catcher for the purified Rb-81 is on the left hand side.

The radioactive rubidium is produced by irradiating natural krypton with 36 MeV protons using the existing facility for charging Kr-81m generators (see 1.2).

The isotopical composition i.e. the activity ratios one hour after EOB is Rb-81: Rb-82m: Rb-83: Rb-84 = 1:1.3: 0.02:0.02. This rubidium activity, fixed onto a small ion exchanger column, is washed from the resin with 1.5 ml 1 n HNO₃. The solution is evaporated on a small tantalum plate, which can be introduced into the ion source of the mass separator. The separated Rb-81 is implanted into sodium chloride crystals. After the routine separation process the radionuclide purity 4 hours after EOB is: Rb-82m < 2 . 10^{-4} ; Rb-83 < 5 . 10^{-6} ; Rb-84 < 5 . 10^{-6} . Since, the beginning of the routine operation (twice a week) in 1984, the mass separator and ion source have

1.4 Future developments

turned out to be very reliable.

Since there is a proposal to install a PET diagnostic centre at KfK, target and process developments will be started with the aim to produce biomolecules labelled with short-lived positron emitters like C-11 (20 min), N-13 (10 min), O-15 (2,2 min) and F-18 (110 min).

2. Radionuclide Technique in Mechanical Engineering The Thin-Layer Activation Technique

In order to supply industrial and engineering research world-wide with thin-layer activated machine components for wear and corrosion measurements at machines and plants during operation, a new powerful irradiation facility has been installed at KAZ. The construction and the technique are based upon the 15-year experience in thin-layer activation. The fully automatic series irradiation facility is shown in Figure 4. Shown are the beam head of the Cyclotron with various devices to adjust and align the machine parts with the beam; in front of that is the positioning unit, which is a 3-dimensional cross-table system with the parts to be irradiated.

The supply system can be seen in the background. This friction roller conveyer can take up to 6 standard plates, upon which a total of up to 50 smaller or 6 large machine parts together with their accompanying movements apparatus per series can be mounted. The maximum weight per part is 6000 N. The parts can be positioned in the beam with a precision of 0.01 mm. Three inter-communicating computers control and monitor the supply, positioning and irradiation, respectively, so that a high degree of reliability can be achieved. The present capacity of series irradiation facility consists of 500 machine parts per year, which can be increased by a factor of three with an additional low investment within a short period of time in order to cover the world-wide demand.



Fig. 4: The fully automatic series irradiation facility for Thin-Layer-Activation of machine parts in service for industry and research laboratories. From the left: Beam head of the cyclotron with various devices to adjust and align the machine parts with the beam; in front of that is the positioning unit: the supply system for standard plates in the background capacity: 500 machine parts per year, 6 large or up to 50 smaller parts per series. Max. weight per part: 6000 N Accuracy in positioning: 0.01 mm With the operation of the high-precision irradiation facility a certain termination of the systematic development of Radionuclide Technique in Mechanical engineering (RTM) at KAZ has been reached. The motivation to commence this work were the urgent demands of the R&D departments at universities and in industry for a reliable diagnosis technique, which would enable wear and corrosion measurements of critical parts in a machine or a processing plant under real operating conditions.

The basic principle and the technique of the RTM system are:

- radioactive labelling (thin-layer-activation),

- the measuring methods,
- the measuring equipment.

These were fully developed and applied in numerous cases in industry. The working mode and main advantages of RTM are briefly described again:

Working Principle: On the critical surface area of the machine part subject to wear a thin layer of radioactive atoms (radionuclides) is produced by irradiation with a beam of charged particles from a cyclotron. The thickness of this layer can be varied between 0.02 mm and 1 mm according to the measuring problem. The position and the size of the wear zone on the engine components is marked with an accuracy of 0.1 mm. The machine is then assembled and operated and the removal of material from the activated region is followed sensitively by a radiation measuring instrument outside the machine. The relationship between surface loss and activity is linear. The activity levels which occur are verylow. The technique can be applied to all industrial iron and

steel grades (low-alloy steels up to high-alloy steels), non-ferrous metals and alloys:

Al, Co, Cr, Cu, Mo, Ni, Pb, Sn, Ti, V, W, Zn, sintered and hard metals.

<u>Advantages:</u> In Situ, On-line Technique for detecting wear in vital parts of a machine in operation.

<u>High Sensitivity</u> in discovering even minute surface losses of the order of micrograms or tenths of micrometers as a function of time and of various operating conditions of a machine.

<u>Selectivity</u> in covering areas of interest inside a machine (sizes between a few tenths of a millimeter and several ten centimeters).

 $\underline{\text{No Impact}}$ of the measurement on the operating conditions or the engine part of interest.

<u>Cost and Time Savings</u> up to $80 \ \%$ of the costs and up to $90 \ \%$ of the time are saved compared to conventional methods, e.g. for optimizing combustion engine parts, according to reports from industrial users.

The practical applications of RTM will now be demonstrated with several photos. In Figure 5 the irradiation setup of a railway brake disc at the KAZ beam head is shown. Through a concentric arrangement of the marked zones on the brake disc, not only the time-dependent, but also the local wear behaviour can be determined. The form and material development of brake discs and wheels of the high-speed train have been performed with RTM.

In Figure 6 the irradiation setup of a pipe elbow is shown. A radioactive label is deposited at a depth of 0.25 mm at the inner wall of the high-load bend region. After the installation of the irradiated pipe elbow material loss in the critical zone through corrosion, cavitation and erosion is determined by an external monitor. The application of RTM enables a precise service schedule and a considerable reduction of production stoppage and material costs.



Fig. 5: Railway brake disc in irradiation position infront of the cyclotron beam head. RTM-diagnosis technique has been applied with reasonable success in development of the components for the high-speed trains.



Fig. 7: Irradiation of cylinder bores in an engine block to monitor wear of the cylinder wall.



Fig. 6: Irradiation set up of a pipe elbow. A radioactive label is deposited at the inner wall of the high-load bend region. The material loss of the critical zone is monitored continuously and enables precise service schedule.

Another region of application is the development of combustion engines, for which two examples are shown in the following photos. Figure 7 shows a cylinder block irradiation. The block is rotated around the axis of the bore to be activated. The ion beam produces a uniform ring-shaped labelling on the cylinder wall in the TDC of the piston ring and enables the wear measurement in this life-time-critical zone.

The irradiation of a camshaft is shown in Figure 8. A special irradiation technique has been developed for the homogeneous activation of the irregular surface of the cams.



Fig. 8: Irradiation of a cam shaft for measuring the wear of the cam heads at different stages of load and revolutions. The very localized labelling enables the separate detection of the wear behaviour of the high-loaded regions on the top and flank of the cams.