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Short Research Article

Production of radioisotopes in the BR2 high-flux reactor for applications in nuclear medicine and industry†

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Abstract: The BR2 reactor, put into operation in 1963 and refurbished in 1995–1997, is a 100 MW high-flux 'materials testing reactor' which provides thermal neutron fluxes up to 10^{15} n/cm^2 s. The production of radioisotopes, characterized by high specific activities, is carried out in dedicated in-core devices within standard irradiation cycles of 3–4 weeks and in accordance with a 'quality system' that has been certified to the requirements of the 'EN ISO 9001:2000' in November 2003. Due to its operating flexibility, its reliability and production capacity, the BR2 reactor is considered as a major facility for a routine supply of radioisotopes such as ^{99}Mo (^{99m}Tc), ^{131}I, ^{133}Xe, ^{192}Ir, ^{186}Re, ^{153}Sm, ^{169}Er, ^{90}Y, ^{32}P, ^{188}W (^{188}Re), ^{203}Hg, ^{82}Br, ^{79}Kr, ^{41}Ar, ^{125}I, ^{177}Lu, ^{89}Sr, ^{60}Co, ^{169}Yb, ^{147}Nd, etc. It will continue to play this key role with the possibility of an extra operating cycle from 2006 and the realization of various 'test' irradiations to develop the production of new radioisotopes. Some irradiation devices allow the loading and unloading of irradiated targets during the operation of the reactor. Hot cells and storage facilities are available to prepare and organize the shipment of the irradiated targets to dedicated processing facilities.

Keywords: BR2 high-flux reactor; radioisotopes; production

Introduction

The BR2 reactor (Figure 1) is a 100 MW$_{th}$ high-flux 'materials testing reactor' which first became operational in 1963 and has since been refurbished in 1995–1997. It is operated by the Belgian Nuclear Research Center in the framework of programmes concerning the development of structural materials and nuclear fuels for fission and fusion reactors. The availability of thermal neutron fluxes up to 10^{15} n/cm^2 s allows an important routine production of radioisotopes characterized by high specific activities for medical and industrial applications. The BR2 reactor is also a major supplier of NTD-silicon worldwide. Currently, a standard irradiation cycle consists of 3 or 4 weeks operation at an operating power between 55 and 70 MW$_{th}$, depending on the core configuration, which is easily adapted to the experimental load.

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Core configuration

BR2's present annual operating regime is based on five irradiation cycles, i.e. 110–120 operating days per year. The reactor uses 93% ^{235}U-enriched uranium as fuel and is moderated by light water and beryllium. The core is composed of beryllium hexagons with central irradiation channels of 200, 84, 50 or 33 mm diameter. The cooling water is pressurized at 12 bar and has a temperature of 40–45°C. The aluminium pressure vessel (Figure 1) is located in a pool filled with demineralized water. The actual core configuration 20G (Figure 2) is characterized by 32 fuel elements, seven control rods and a regulating rod. This is arranged around the central beryllium plug H1 in order to provide peak thermal neutron fluxes up to 10^{15} n/cm^2 s in its seven irradiation channels for the production of radioisotopes of high specific activities as ^{192}Ir, ^{89}Sr, ^{188}W, ^{32}P, ^{169}Yb, etc. Other irradiation positions are located in peripheral reflector channels (peak thermal neutron fluxes from 1 to 3.5×10^{14} n/cm^2 s) for the production of ^{99}Mo, ^{131}I, ^{133}Xe, ^{153}Sm, ^{186}Re, ^{90}Y, ^{177}Lu, ^{125}I, ^{169}Er, ^{203}Hg, ^{82}Br, ^{41}Ar, ^{79}Kr, ^{147}Nd, etc. The thermal neutron fluxes of 4.5×10^{14} n/cm^2 s

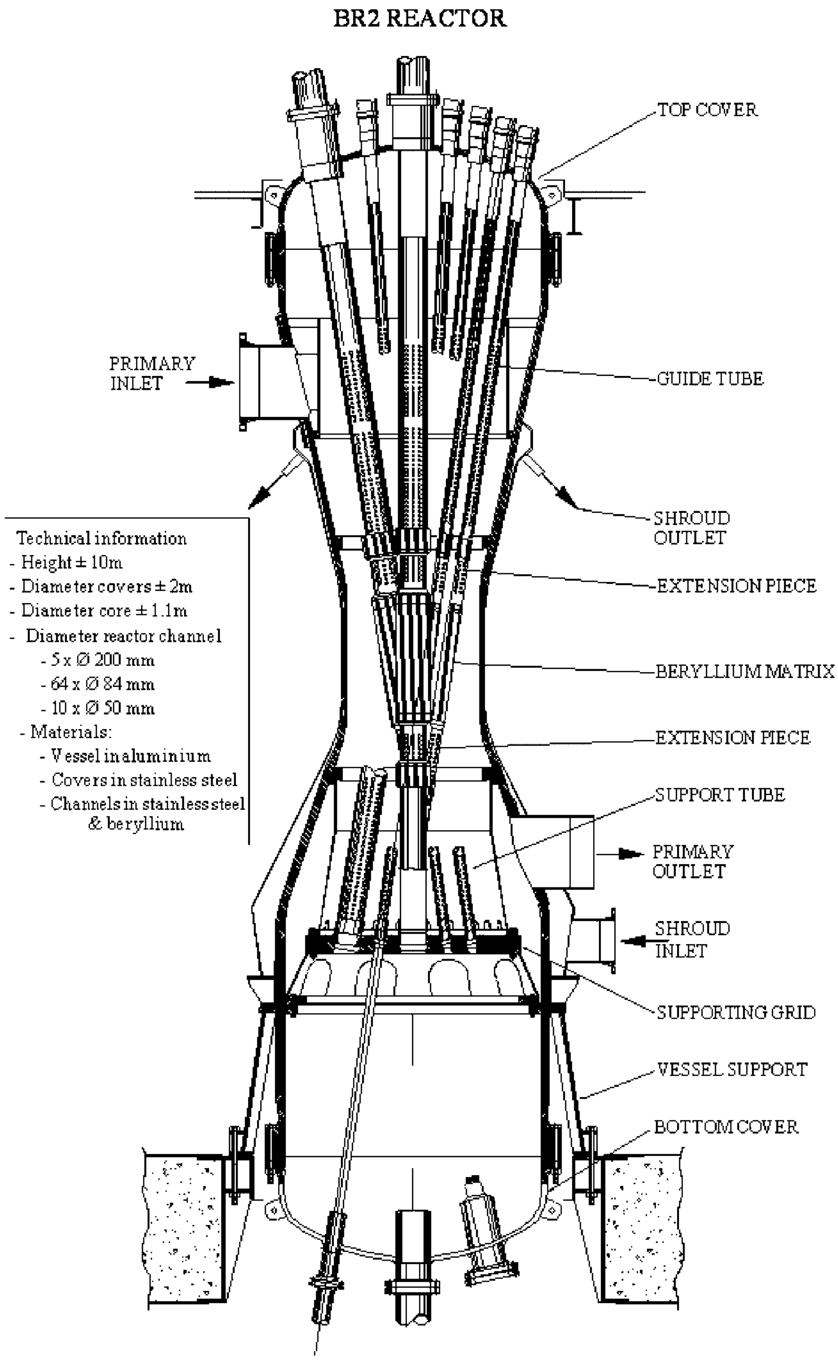

Figure 1 BR2 reactor.

available at the bottom of the control rods are used for the production of ^{60}Co. If fast neutron fluxes are required, for the production of ^{117m}Sn, ^{67}Cu, etc., for example, the targets are loaded into an irradiation basket inside a BR2 fuel element where the neutron flux above an energy of 0.1 MeV is between 1 and 5×10^{14} n/cm^2 s, depending on the burn–up and the location of the fuel element in the core.

Irradiation capsules and devices

The targets for the production of radioisotopes are supplied by the customers and loaded into suitable irradiation capsules. After commissioning tests, the capsules are loaded and irradiated in dedicated in-core devices; some of them allow their loading and unloading during the operation of the reactor. After

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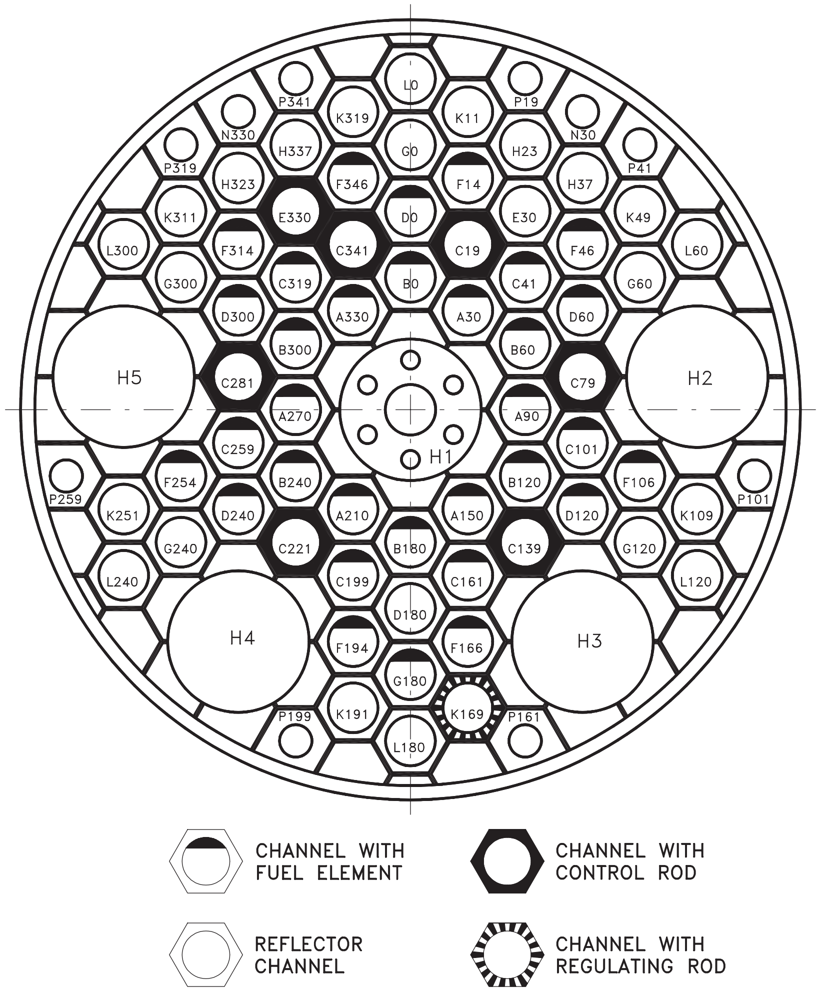

Figure 2 BR2 configuration (20G).

irradiation, temporary storage for cooling and, if required, passage through the BR2 hot cells for decanning, the targets are loaded into suitable shipping containers in order to be sent back to the customers for processing. The routine production of radioisotopes is carried out in standardized cold-welded aluminium '*CSF*' capsules which contain an inner graphite or aluminium cylinder and helium gas. If necessary, the target material is loaded into quartz ampoules (Figure 3). Other type of capsules are manufactured according to the dimensions of the specimens to be loaded, the space available in the irradiation device, and parameters as heat production, chemical composition, physical characteristics, irradiation time, activity to be produced, etc. The irradiation capsules are loaded into aluminium irradiation baskets characterized by a diameter of 25 mm in the reflector and 15 mm inside a standard six plates fuel element. Loading and unloading of the irradiation baskets are only possible during the shutdown periods of the reactor.

The four aluminium thimble tubes '*DG*' ('Doigt de Gant') provide a total irradiation capacity of 36 'CSF' capsules at the same time. They are loaded in peripheral reflector channels as E30, H37, K191 or L180 (Figure 2), allowing the loading and unloading of the 'CSF' capsules during the operation of the reactor.

The two '*DGR*' ('Doigt de Gant Refroidi') devices were built in order to improve the heat evacuation capacity of the thimble tubes 'DG' by the installation of an individual closed circuit of forced water cooling. They are dedicated to the production of fission radioisotopes like ^{99}Mo (^{99m}Tc), ^{131}I and ^{133}Xe by the irradiation of 93% ^{235}U-enriched uranium targets. They provide a total irradiation capacity of six targets of 4 g ^{235}U at the same time. The targets can be loaded and unloaded during the operation of the reactor. The four '*PRF*' (Primary Reloadable water-cooled device for Fissile

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targets) devices are also dedicated to the production of fission radioisotopes like ^{99}Mo (^{99m}Tc), ^{131}I and ^{133}Xe by the irradiation of 93% ^{235}U-enriched uranium targets. They are loaded in reflector channels as G0, G60, G120, G240 and G300 (Figure 2) and provide a total irradiation capacity of 50 targets of 4–5 g ^{235}U at the same time. The targets are cooled by the primary water of the reactor during irradiation and in the cooling position before unloading; they can be loaded and unloaded during the operation of the reactor.

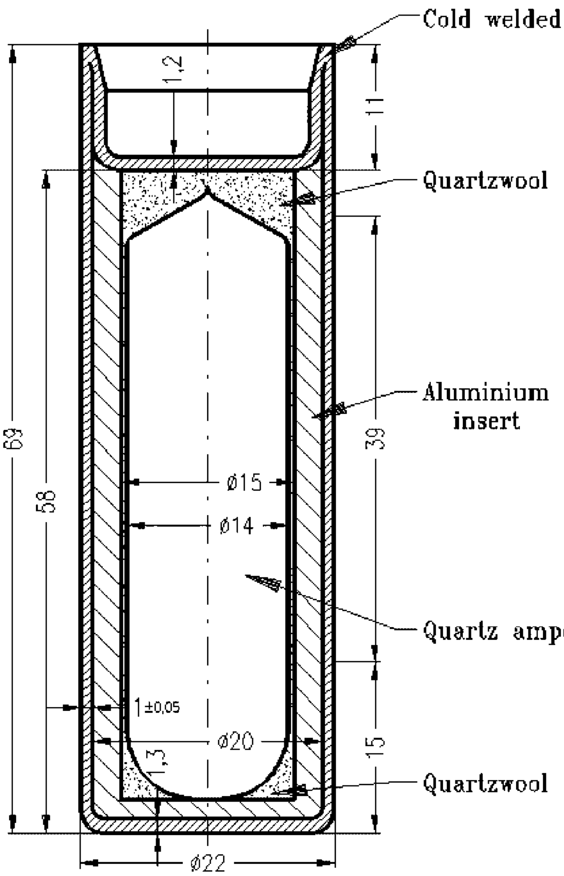

Figure 3 Typical 'CSF' cold-welded irradiation capsule loaded with a quartz ampoule.

Major radioisotopes produced in the BR2 reactor

The major isotope produced in the BR2 reactor is **^{99}Mo** ($T_{1/2} = 66$ h). It is obtained by the irradiation of high enriched uranium targets (93% ^{235}U) of 4–5 g ^{235}U. Six irradiation devices are routinely loaded in reflector channels as G120 (Figure 2), providing a total irradiation capacity of 56 targets at the same time, i.e. 168 targets per reactor cycle. A typical irradiation of 150 h in a thermal neutron flux of 2.5×10^{14} n/cm^2 s yields up to 1000 Ci per target at the end of the irradiation 'EOI,' i.e. 220 Ci per target at calibration date ('EOI'+6 days), process yield not taken into account. In their processing facilities, the customers recover the ^{99}Mo activity – in order to manufacture **^{99}Mo/^{99m}Tc** ($T_{1/2} = 6$ h) generators, the most important diagnostic tool in nuclear medicine – and other fission isotopes as **^{131}I** ($T_{1/2} = 8.02$ days) for thyroid cancer therapy and **^{133}Xe** ($T_{1/2} = 5.24$ days) for diagnostic imaging of the lung function.

The production of **^{192}Ir** ($T_{1/2} = 74$ days) for industrial applications (mainly testing of welds) consists in the irradiation of *natural iridium* discs in the six peripheral channels of the central beryllium plug H1, characterized by thermal neutron fluxes of up to 10^{15} n/cm^2 s. The maximum irradiation capacity is 48 'CSF' capsules loaded with 5 g iridium each; in this case, an additional fuel element can be loaded in H1/central in order to compensate the large antireactivity effect of the

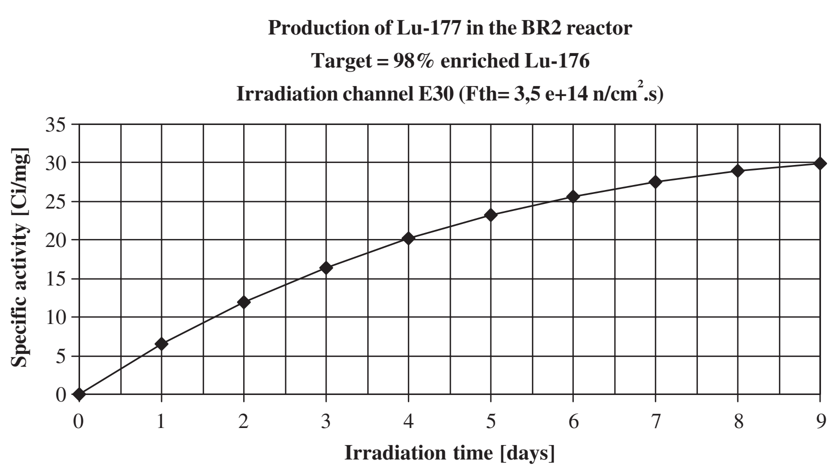

Figure 4 ^{177}Lu specific activity as a function of the irradiation time.

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irradiation baskets. The specific activities achieved after one irradiation cycle of 21 days are 500–750 Ci/g 'EOI,' depending on the size of the discs, the loading of the capsules and the axial position in the basket. Furthermore, for medical applications (brachytherapy) and to provide BR2 with more flexibility to satisfy the demand of the customers during its shutdown periods, *enriched iridium* targets (80% ^{191}Ir) are irradiated to supply specific activities up to 1500 Ci/g 'EOI.'

A considerable effort has been made to take advantage of the high thermal neutron fluxes available in the central beryllium plug H1 to produce other radioisotopes than ^{192}Ir. Irradiations of high enriched (97%) ^{186}W targets in 'CSF' capsules have been performed in collaboration with ORNL (Oak Ridge, USA) for the production of $\mathbf{^{188}W}$ ($T_{1/2} = 69.4$ days) and the manufacture of $\mathbf{^{188}W/^{188}Re}$ ($T_{1/2} = 17$ h) generators.[1] Specific activities of 1.1 Ci ^{188}W/g of ^{186}W were obtained after one irradiation cycle of 21 days in the central beryllium plug H1. $\mathbf{^{188}Re}$ is one of the most attractive radioisotopes for a wide variety of therapeutic applications in nuclear medicine, oncology and cardiology because of its availability from a generator, its attractive radionuclidic and chemical properties. The generator system itself has a very long useful shelf-life of several months, which makes it cost effective and attractive.

The productions of $\mathbf{^{89}Sr}$ ($T_{1/2} = 50.5$ days) for metastatic bone pain palliation and $\mathbf{^{32}P}$ ($T_{1/2} = 14.3$ days) for cardiology are also carried out in 'CSF' capsules loaded in the central beryllium plug H1. Specific activities of 0.3 Ci ^{89}Sr/g of ^{88}Sr and 50 Ci ^{32}P/g of ^{31}P are achieved after one irradiation cycle of 21 days.

The productions of $\mathbf{^{186}Re}$ ($T_{1/2} = 3.8$ days), $\mathbf{^{153}Sm}$ ($T_{1/2} = 46.7$ h) and $\mathbf{^{90}Y}$ ($T_{1/2} = 64$ h) for metastatic bone pain palliation are carried out in 'CSF' capsules loaded in 'DG' irradiation devices. Specific activities of 1400 Ci ^{186}Re/g of ^{185}Re, 4500 Ci ^{153}Sm/g of ^{152}Sm and 60 Ci ^{90}Y/g of ^{89}Y are achieved by irradiation of enriched targets during, respectively, 5, 4 and 9 days in a thermal neutron flux of 3.5×10^{14} n/cm^2 s.

The productions of $\mathbf{^{177}Lu}$ ($T_{1/2} = 6.71$ days) for therapy applications are carried out in 'CSF' capsules loaded in 'DG' irradiation devices. The irradiation of 98% enriched ^{176}Lu during 7–9 days in a thermal neutron flux of 3.5×10^{14} n/cm^2 s yields specific activities of 27.5–30 Ci/mg of ^{176}Lu (Figure 4).

Conclusion

The extensive refurbishment programme of the BR2 reactor, performed in 1995–1997 after more than 30 years utilization, provided a lifetime extension of more than 20 years. A serious effort has also been made to perform all the commercial activities (production of 'radioisotopes' and 'neutron transmutation-doped silicon') in accordance with a 'quality system' that has been certified to the requirements of the 'EN ISO 9001:2000 – quality systems – model for quality assurance in production, installation and servicing' (3 November 2003). The BR2 reactor will thus continue to contribute to a reliable supply of a wide range of radioisotopes to meet global demands for many years.

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