



Neutronic and thermo hydraulic analysis of a modeled subcritical uranyl nitrate aqueous reactor driven by 30-MeV protons



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ABSTRACT

Recently aqueous homogeneous reactors have been taken in the spotlight again because of their good potential for radioisotope product since ARGUS exhibits such successful experience. Accelerator-driven types of such aqueous homogeneous reactors are being investigated and designed due to some their prominent advantages than the used ones by operation in critical level. The present work discusses neutronic and thermo-hydraulic performance of a modeled accelerator-driven aqueous homogeneous reactor. 30 MeV protons with 150 μA current were considered to induce fission process inside the core involving uranyl nitrate solution. Monte Carlo-based computational code was used to model such subcritical system. FLUENT code was used to determine the temperature distribution inside the operated subcritical core with an offered simplified geometry. The carried out calculations showed that the modeled system with external beam window positioning experiences 4.683 kW fission-related power. A neutron flux of the order of 10^{11} n/s. cm^2 is available inside the subcritical core. Enough negative reactivity coefficient of the modeled core moreover its subcritical level operation assures the core inherent safety. The system can be used to produce an adequate yield of different radioisotopes per week. Thermo-hydraulic system used for the modeled core could efficiently remove the aqueous core produced heat and keep the core temperature below 70 $^{\circ}\text{C}$.

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1. Introduction

The aqueous homogeneous reactor (AHR) was one of the first reactors built at the end of 1943, running on a uranyl sulfate solution. In 1944, the LOPO reactor went critical using a uranyl sulfate solution of 565 g ^{235}U dissolved in 13 L of light water in a sphere of 30 cm diameter. Between 1940s up 1980s, many AHRs were built and operated such as SUPO, HYPO, HRE, ARGUS, SILENE and so on (Bunker, 1983; Lane, 1958).

Currently, these aqueous reactors have been extensively taken in account for production of the radioisotopes. Some countries are developing subcritical accelerator-driven type of these aqueous reactors (IAEA-TECDOC-1601, 2008; Stichelbaut, 2011; Chemerisov et al., 2011). Ion Beam Applications (IBA) Company already started in 1995 to think about new ways to produce ^{99}Mo based upon accelerators instead of nuclear reactors (Stichelbaut, 2011).

Some accelerator-driven subcritical reactors with solid fuels are being designed by some countries. ADONIS is one project of this type, which is being run by Belgium and relies on a high-energy

high-current cyclotron coupled to a subcritical assembly. The proton beam impinges on a conical spallation target made in Tantalum to generate a high-intensity neutron flux. The Ta target is surrounded by 4 cylindrical layers of HEU targets interleaved with Beryllium moderation rings. The targets have a cylindrical shape with an outer radius of 1.1 cm and a length of 20 cm. They are identical to those used in production today in nuclear reactors, contain 4 g of ^{235}U each, and immersed in heavy water (D_2O) for cooling. The subcritical core contains 150 HEU targets surrounded by a thick Be reflector (Stichelbaut and Jongen, 2011).

In addition, Advanced Medical Isotope Corporation licensed a hybrid accelerator-based technology from the University of Missouri to provide a minimum of 50% of the United States ^{99}Mo demand from a subcritical solution of Low-Enriched Uranium. An electron beam device strikes a high-density tungsten target. The produced photons emerge into a stainless steel tank holding D_2O and low-enriched uranium salt. The photons eject neutrons from deuterium atoms, initiating fission in the LEU target material, which provides fission reaction in the LEU target (Stevenson et al., XXXX).

High proton-fission cross sections on $^{235/238}\text{U}$ isotopes, even at low proton energies with an $E_{\text{threshold}} \sim 10$ MeV (EXFOR, 2016) motivates us to investigate application of low energy proton

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accelerators for providing an external neutron source in a subcritical-designed aqueous homogeneous reactor. Hence, in this work potentially investigation of an aqueous subcritical reactor containing uranyl nitrate fuel for producing the ^{131}I , ^{89}Sr and ^{99}Mo radioisotopes was proposed. Proton accelerator of CYCLONE 30 was used to drive the subcritical core.

The CYCLONE30, from Ion Beam Applications Belgium, commercial accelerator utilizes an ion source technology similar to a LBL style ion source dating from the early 1980s. The Cyclotron is a compact device created with the purpose of accelerating positive or negative ions of H, D or α particles whose beam is extracted and conveyed toward to the specific targets. This device can generate beams of protons with energies up to 30 MeV and current up to 350 μA (Leung et al., 1986; Rabelo and Campo, 2011).

2. Material and methods

In this work, MCNPX 2.6.0 has been used as a powerful particle transport code with the ability to calculate steady-state reaction rates, normalization parameters, neutronic parameters, as well as fuel burn up using CINDER90 to calculate the time-dependent parameters (Pelowitz, 2008; Fensin, 2008).

A cylindrical aqueous reactor was modeled using the MCNPX 2.6.0 code. Light water flowed inside the considered coils was selected as a coolant for the fuel solution. Heavy water was used as a reflector. First A 40 cm thickness was selected for the reflector and its adequate thickness was determined in next steps. A 3D neutronic model was set up using the MCNPX 2.6.0 code in cold zero power situations by means of ENDF/B-VI continuous-energy cross section. The cross sections of $S(\alpha, \beta)$ was used for fuel solution, heavy water and light water. KCODE card was used for neutronic parameter calculations. The modeled core specifications are presented in Table 1.

A fixed dimension ($36 \times 38 \text{ cm}$) was selected for the modeled cylindrical core and the uranium concentration of 190 g/l was selected to achieve an effective multiplication factor (k_{eff}) less 0.96. The core volume was selected as 35.3 lit. The core dimension is chosen in such a way to keep the geometry buckling ($H = 1.84 \cdot R$ for a cylindrical bare core, H: core height, R: core diameter) and a k_{eff} less than 0.96. If higher-enriched uranium is used in uranyl nitrate solution, a more compact core is accessible. In the present work, 20%-enriched uranium (U) was considered to form uranyl nitrate solution. A fixed dissolved 190 g U/l was proposed for the aqueous solution; a lower uranium salt concentration in the fuel solution results in a larger Kd for Mo(VI) and therefore a more effective and efficient recovery of ^{99}Mo from such solutions (Bunker, 1983). Uranyl nitrate solubility in water is 660 g/L (Elgin, 2014). By enhancing the core radius, the core effective multiplication increases, then an optimized radius and height were determined with respect to the proposed k_{eff} . Because of huge numbers of the available figures, the optimization process report was ignored in the manuscript text.

Two beam window positioning options were investigated consist of 1-beam window positioning inside the fuel solution 2-beam window positioning outside the fuel solution (Fig. 1).

Table 1
Core material and dimensions modeled using MCNPX 2.6.0.

Core specifications	Value	Unit
Fuel solution 1: W%: ^{235}U : 3.1, ^{238}U : 12.61, O: 74.2, H: 8.2, N: 18.4	1.19	g/cm^3
Stainless Steel cover plate: W%: Fe:69.5, Cr: 19, Ni: 9.5, Mn: 2	7.95	g/cm^3
Heavy water reflector: W%: D: 66.67, O: 33.33	1.105	g/cm^3
Dimension (Diameter $\times$ Height)	36×38	cm

A 500- μm tungsten beam window was considered for the subcritical system. The 30 MeV protons with 150 μA current and 0.8 FWHM spatial distribution were regarded to irradiate the beam window. Light water was used to cool the beam window with 1.2 lit/min flow (Fig. 2).

Light water cooling coils with 1.2 cm diameter were used to cool the fuel solution during the subcritical reactor operation. Beryllium oxide reflector was considered to reflect neutrons through the fuel solution in the modeled geometry with outside beam window positioning. The BeO thickness and height was determined to maximize the fission rate due to the reflected neutrons toward the fuel solution.

Neutron spectra available in the different modeled geometries were calculated using the F4 tally card of the computational code. Axial and radial deposited power distributions were calculated using the mesh tally card for the geometries. Reactivity coefficients of fuel, coolant, and moderator were calculated using the TMP card and temperature-related cross section library of .71c from endf70 in MCNPX. The TMP cards provide MCNPX the time-dependent thermal cell temperatures that are necessary for the free-gas thermal treatment of low-energy neutron transport. This treatment becomes important when the neutron energy is less than about 4 times the temperature of heavy nuclei or less than about 400 times the temperature of light nuclei. The thermal temperature of a cell is denoted by kT in units of MeV. To apply a temperature for a cell, its temperature should be written on MeV at the end of the input line related to this cell and simultaneously the related library be used in the material card, which identifies the cell material. Other required libraries of endf70a, endf70b, endf70c, endf70i, endf70j, endf60, mcplib04, ENDF/B-VI and ENDF/B-VII-0 were used for the calculations. The void reactivity effect of the coolant and the solution fuel were calculated for the modeled cores. Delayed neutron fraction and effective delayed neutron fraction were calculated separately for the cores separately. The Burn-up calculation was performed in a power of 4.683 kW for 2 months using the BURN card for the selected geometry. Production rate of ^{99}Mo and ^{89}Sr radioisotopes after the burn-up time were investigated. The production rate of long half-life alpha emitter radioisotopes after the burn-up time was investigated. The average temperature of the modeled subcritical core driven by 30 MeV protons and a current of 150 μA was calculated using the FLUENT code (ANSYS, 2011).

3. Results and discussion

Initially, the proton beam window was considered in the aqueous homogenous fuel solution. The cylindrical subcritical core dimension was selected so that could achieve a multiplication factor of <0.96 when it contains 20%-Enriched uranyl nitrate solution. The computational calculations showed the modeled core effective multiplication factor is 0.93976 ± 0.00022 using 190 g/l of the enriched dissolved uranium in the aqueous reactor. Total deposited power inside the modeled core was calculated using F7 tally and nps mode of the used computational code. The 30-MeV proton beam produces neutrons inside the tungsten beam window, which could emerge inside the fuel solution and start fission process. The calculations showed 7.173 kW power was deposited inside the modeled subcritical core by the fission process. Another proposed geometry was investigated which the beam window was inserted outside of the fuel solution in order to the subcritical core simplification. However, empty place of the beam window is replaced by the fuel solution that increased the solution volume about 412 cm^3 , it is expected the fission rate falls down for losses of the produced neutrons in the external beam window. The calculations showed the second modeled core effective multiplication factor is 0.92116 ± 0.00022 using the same uranium concentration dis-

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