

THE MAPLE-X CONCEPT DEDICATED TO THE PRODUCTION OF RADIO-ISOTOPES

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ABSTRACT

MAPLE is a versatile new Canadian multi-purpose research reactor concept that meets the nuclear aspirations of developing countries. It is planned to convert the NRX reactor at Chalk River Nuclear Laboratories into MAPLE-X as a demonstration prototype of this concept and thereafter to dedicate its operation to the production of radio-isotopes. A description of MAPLE-X and details of molybdenum-99 production are given.

INTRODUCTION

MAPLE is a new Canadian multi-purpose reactor concept (1) resulting from an in-depth review by Atomic Energy of Canada Limited (AECL) to determine the role played by research reactors in national nuclear development. A research reactor business opportunity has been identified and a business organization is now in place to develop this opportunity.

The MAPLE (Multipurpose Applied Physics Lattice Experimental) concept is, in essence, an open-tank-type reactor assembly within a light-water pool. Its purpose is to combine the characteristics of an H₂O-cooled and moderated, enriched (MTR-type) reactor with the technical advantages of using D₂O as a reflector. This is achieved by utilizing a variant of AECL's SLOWPOKE (2) technology in combination with a scale-down and simplification of the basic NRX concept. (3)

AECL is a world leader in the supply of radio-isotopes for both nuclear medicine and industry; the primary production sources being the CANDU reactors, for medium specific activity cobalt-60, and the NRU and NRX research reactors, at Chalk River Nuclear Laboratories (CRNL), for the majority of the rest. However, the demands made by isotope production on operating schedules are often in conflict with the requirements of research and development programs for the NRU and NRX reactors; a situation that will be aggravated when the NRX reactor is retired in 1987.

It is planned to replace NRX with a reactor called MAPLE-X that can be dedicated to the production of radio-isotopes following demonstration of the MAPLE concept as a multi-purpose reactor.

The intent is to replace the NRX calandria with an open-tank, pool-type, prototype MAPLE reactor utilizing as many of the NRX reactor utilities as is economically practical. Thus MAPLE-X would serve the dual role of demonstration reactor and primary supplier of radio-isotopes.

The purpose of this paper is to describe MAPLE-X and to define a mode of operation for it as a dedicated isotope production facility (DIPF) that meets AECL Radiochemical Company's projected

requirements for fission product Mo-99 from 1988 into the next century. This isotope is the source of one of nuclear medicine's most powerful diagnostic isotopes Tc-99m and over 60% of the world market for this product is currently supplied from the NRU reactor. The Mo-99 fission product is extracted chemically from irradiated U-235 targets.

Technical and economic considerations dictate that any changes to established CRNL methods for U-235 target manufacture and post irradiation processing be kept to a minimum. Operation as a DIPF requires that the ratio of target power to reactor fuel power be maximized. Together these translate into MAPLE-X using a minimum practical critical mass of driver fuel, and U-235 targets, similar to the NRU design, placed in the highest neutron flux available at a reactor power level of around 10 MW. Details of the U-235 targets and outlines of both the initial and equilibrium MAPLE-X core loading are described. The results of reactor physics simulations using the 3-dimensional diffusion code 3DDT (4), together with a 5 neutron energy group cross section library derived from WIMS (5), are given. These simulations are used to estimate optimum Mo-99 production based on the assumption of a 100% reactor duty factor.

DESCRIPTION OF MAPLE-X

General

The basic MAPLE reactor concept consists of an open-tank-type reactor assembly within a light-water pool as shown in Figure 1.

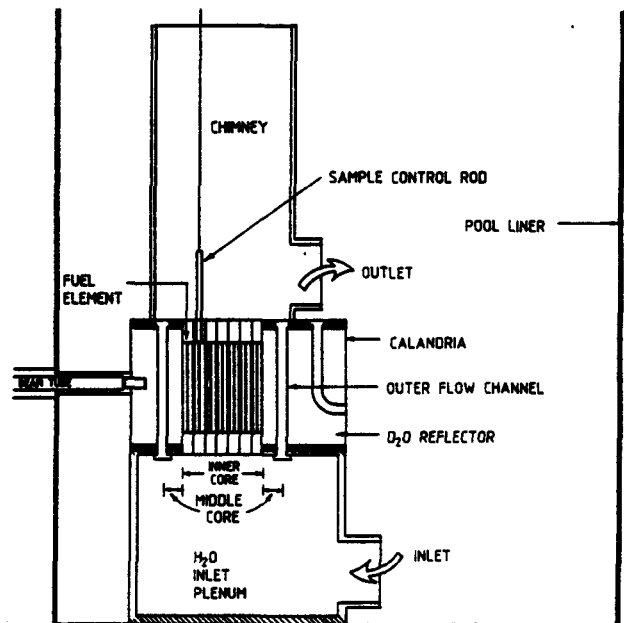


FIGURE 1: MAPLE SCHEMATIC OUTLINE

The reactor assembly consists of three parts: the inlet plenum, the calandria assembly and the chimney. The light-water primary coolant enters the plenum, flows upwards through the calandria assembly and exits from the chimney via an outlet nozzle. The calandria assembly is a cylindrical zirconium-alloy vessel penetrated by vertical flow tubes and horizontal beam tubes. The arrangement for MAPLE-X is shown in Figure 2.

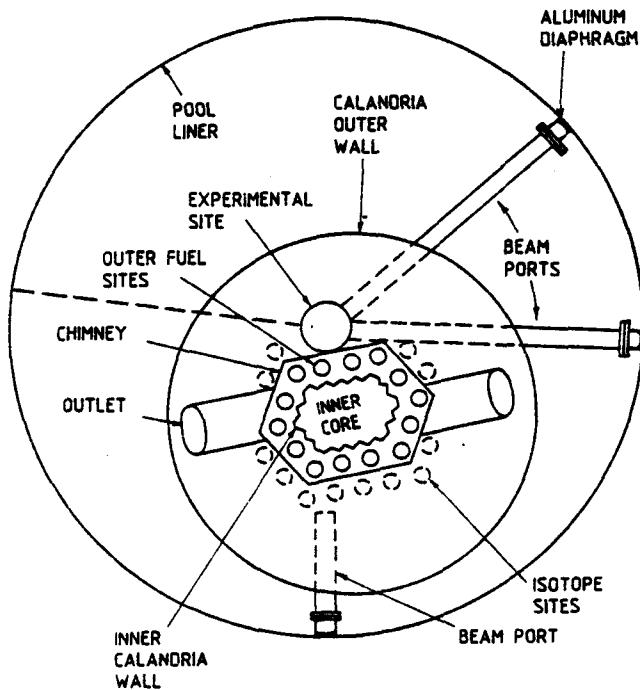


FIGURE 2: MAPLE-X CALANDRIA ARRANGEMENT

The vertical flow tubes are arranged to define three regions within the calandria:

- ° a central H₂O-cooled, H₂O-moderated core region
- ° an intermediate H₂O-cooled, D₂O-moderated core region
- ° an outer D₂O-filled reflector region

This combination of H₂O and D₂O moderated lattices within a D₂O calandria tank enables the MAPLE concept to achieve the flux advantages of a basic H₂O-cooled and moderated reactor while retaining the flexibility and space of a NRX-type reactor.

The MAPLE-X concept is to put a MAPLE core, suitable for isotope production, into a pool created in the NRX biological shields by lining the cavity left when the NRX calandria and upper shields are removed.

Inlet Plenum

The inlet plenum is a cylindrical stainless steel vessel 1.8 m diameter by 1.2 m high. The plenum is fixed at its base to the bottom of the pool liner. Loads from the outlet chimney and calandria vessel are transmitted through the plenum wall and into the pool liner.

The plenum serves as a coolant header with two 0.3 m diameter nozzles on the lower part of the plenum providing connections to the primary heat transport system.

Calandria Vessel

The calandria vessel is an annular, cylindrical, heavy-water filled, zirconium alloy vessel 1.8 m diameter by 0.8 m height. The inner annular wall of the vessel is in the shape of an elongated hexagon with fluted sides that penetrate and are welded to 10 mm thick upper and lower tube sheets. The central cavity thus formed is the boundary of the central core region which can accommodate 29 flow tubes on a hexagonal pitch of 80.1 mm.

Surrounding the central core region are 14 cylindrical flow tubes, 60 mm inside diameter, which penetrate directly through the calandria and are attached to the upper and lower tube sheets. They form the intermediate core region and are located on a hexagonal pitch of 160.3 mm.

A number of flow tubes with large radius 90 degree bends are attached to the upper tube sheet, outside the chimney, and to the outer shell wall. These positions, which are cooled by natural circulation of pool water, are non-fuel irradiation sites in the D₂O reflector region.

Outlet Chimney

The chimney assembly consists of a hollow elongated hexagonal structure welded to a 1.8 m diameter tube sheet. The chimney is a minimum of 0.8 m across the flats and is 2.6 m high.

Two 0.3 m diameter nozzles located about 1.2 m above the chimney tubesheet are provided for the connection of the primary heat transport piping. The chimney confines the upward flow of coolant from the core region, collects a downflow from the pool equivalent to the core bypass flow, and directs the mixture to the primary heat transport system for purification and recirculation, thereby suppressing N-16 gamma levels at the pool surface.

Near the top of the chimney, brackets and guides are attached for guiding and locating the reactivity control mechanisms.

Fuel Assemblies

Three types of fuel assembly are required for MAPLE-X. These are shown in Figure 3 and detailed in Tables 1 and 2.

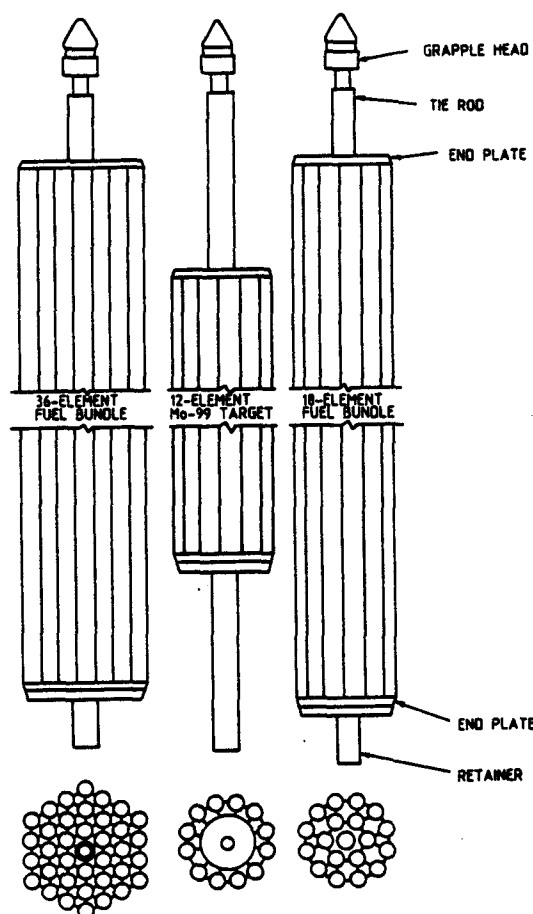


FIGURE 3: FUEL ASSEMBLIES

The 36-element bundles are installed in sites that have semi-permanent hexagonal flow tubes. Both the 18-element bundles and the 12-element Mo-99 targets can be installed either in central core region sites that have semi-permanent cylindrical flow tubes, or in the permanent flow tubes of the 14 intermediate core sites. The semi-permanent flow tubes of the core region are not removed during normal fuel management operations but all sites in the inner core can accommodate either type of flow tube thus permitting maximum flexibility of core loading arrangements.

All three types of fuel assembly have similar central Zircaloy support shafts. The fuel elements in the 36-element bundles are arranged on an 11 mm pitch. Those of the 18-element bundles are arranged in two concentric annuli of 6 and 12 elements, respectively. These two bundles use low enrichment uranium (LEU, less than 20% U-235 in total U) as driver fuel for MAPLE-X. The Mo-99 fuel targets use highly enriched uranium (HEU, 93% U-235 in total U) compatible with current NRU practice. A Mo-99 target assembly consists of a ring of 12 elements surrounding an aluminum can attached to the central support shaft. This can, which contains essentially stagnant H₂O coolant, defines the coolant flow path over the target fuel pins.

TABLE 1: MAPLE-X FUEL ELEMENT AND BUNDLE ASSEMBLY SPECIFICATIONS

<u>Fuel Element</u>	
Fuel Meat	61 wt% U ₃ Si-39 wt% Al
Enrichment	20 wt% U-235 in U
Density	5.43 Mg/m ³
Diameter	6.35 mm
Length	600 mm
Mass of LEU	60.14 g
Mass of U-235	12.03 g
<u>Cladding</u> Al (co-extruded)	
Thickness	0.76 mm
Fins	8
Height	1.02 mm
Width	0.76 mm
Diameter over Cladding	7.88 mm
Diameter over Fins	9.91 mm
<u>18 Element Fuel Assembly</u>	
Shape	Cylindrical
<u>Flow Tube Diameter</u>	
Inner	61.1 mm
Outer	62.8 mm
Mass of LEU	1.083 x 10 ³ g
Mass of U-235	216.5 g
<u>36 Element Fuel Assembly</u>	
Shape	Hexagonal
<u>Flow Tube Diameter (Flat to Flat)</u>	
Inner	74.4 mm
Outer	76.6 mm
Mass of LEU	2.165 x 10 ³ g
Mass of U-235	433.0 g

TABLE 2: MAPLE-X TARGETS FOR MO-99 PRODUCTION

<u>Fuel Element</u>	
Fuel Meat	21 wt% U-79 wt% Al
Enrichment	93 wt% U-235 in U
Density	0.68 Mg/m ³
Diameter	5.49 mm
Length	300 mm
Mass of U-235	4.5 g
<u>12 Element Target Assembly</u>	
Shape	Cylindrical
<u>Flow Tube Diameter</u>	
Inner	61.1 mm
Outer	62.8 mm
Mass of U-235	54.0 g

Control and Shut-Off Rods

These rods are actually hollow cylinders which slide over the semi-permanent cylindrical flow tubes of 18 element driver fuel rods in the central core region as indicated in Figure 4.

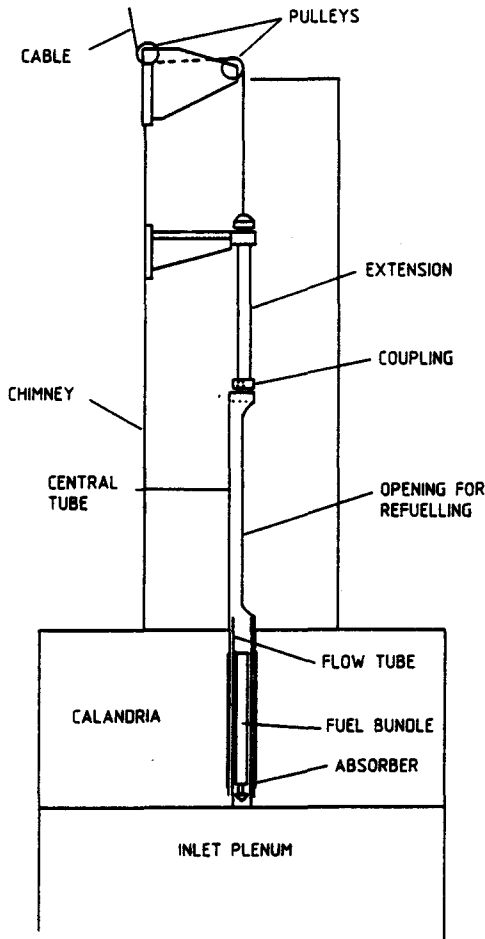


FIGURE 4: CONTROL ABSORBER UNIT

Each rod includes a 1.8 m long stainless steel central tube with a wide cut-out extending over the upper portion to permit fuel changing. Neutron absorbing material, either boron carbide or hafnium, is suitably attached to the outside of this tube over the bottom 600 mm section of the rod and is aligned with the driver fuel on full insertion into the core.

Fuel Handling

All fuel handling is done with the reactor in a shut-down state using hand tools from a bridge that can be driven over the pool.

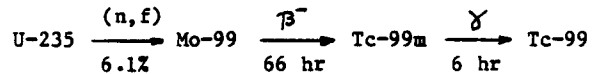
PRODUCTION OF MO-99/TC-99m

General

Uranium-235 fission produces molybdenum isotopes Mo-95 to Mo-100 inclusive. All of these isotopes are stable except for Mo-99 which decays, with a 66 hour half-life, to a temporary isomer of technetium

(Tc-99m). This isomer subsequently decays to the stable ground state (Tc-99), with a 6 hour half-life, and emits a soft (140 keV) gamma ray in the process. The combination of short half life and soft gamma ray emission makes Tc-99m one of the most powerful diagnostic sources known to nuclear medicine.

The production sequence is summarized as follows:



Secular equilibrium of Mo-99 and Tc-99m is reached in approximately two weeks. Percentage production levels resulting from irradiation in a thermal neutron flux of $10^{14} \text{ n.cm}^{-2}.\text{s}^{-1}$ are shown in Figure 5.

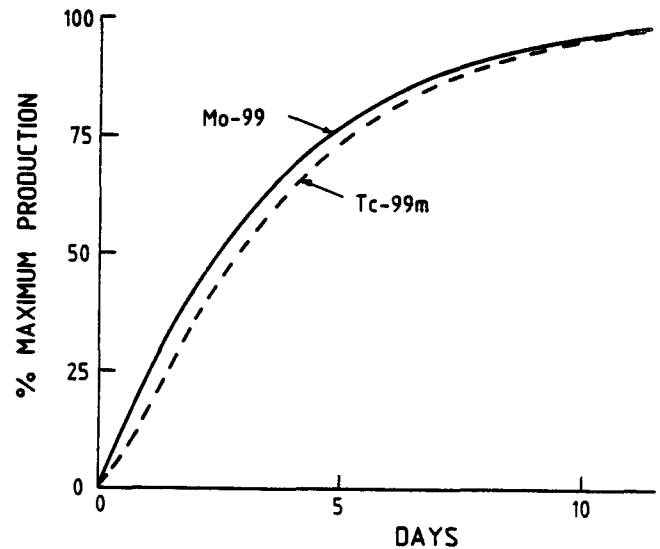


FIGURE 5: MO-99/TC-99m IN-REACTOR PRODUCTION

On discharge from the reactor the molybdenum fission products are chemically extracted from the U-235 targets at CRNL and shipped to AECL Radiochemical Company for final purification, packaging and world-wide distribution. The 66 hour half-life of Mo-99 gives the product a useful shelf lifetime of 3 to 4 weeks.

The production rate of Mo-99 atoms at equilibrium is directly proportional to the U-235 target power and, under these conditions, 5.15×10^4 Curies of Mo-99 per megawatt of target are produced at the end of irradiation (E.O.I.). Time delays incurred to purify and distribute the final product world-wide are considered by defining a "6-day Curie" measure as the market standard. This is simply the E.O.I. Curies remaining at the time of measurement, of the purified Mo-99 product, discounted by a further 6 days of decay.

Secular equilibrium activity levels of Tc-99m are dictated by the quantity of Mo-99 present at a given time and by the half-life of each isotope. If the isotopic balance between parent and daughter is chemically altered, then the 6 hour half-life of Tc-99m rapidly restores it as shown in Figure 6.

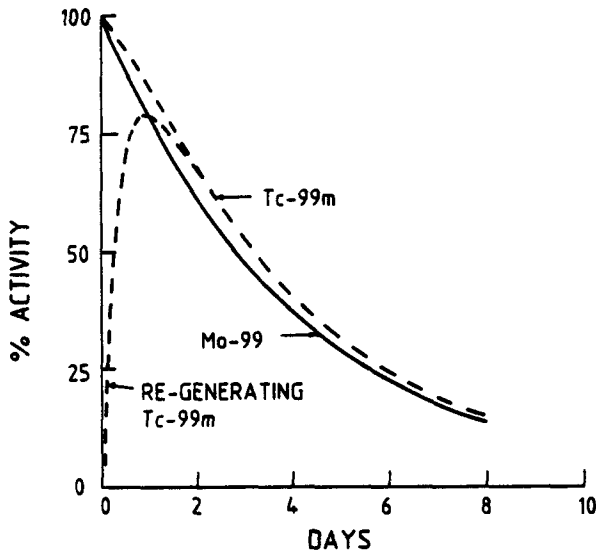


FIGURE 6: Mo-99/Tc-99m DECAY OF PRODUCT

Thus molybdenum "cows" are shipped regularly to hospitals around the world where they are "milked" daily for technecium.

Uranium-235 Target Irradiations

To meet the Mo-99 market requirements efficiently, a certain minimum number of U-235 targets must be irradiated simultaneously at as high a duty factor as possible. Some fraction (roughly a half) of these are replaced and processed each week ensuring that each target is irradiated for a minimum of 9 days. Longer irradiation times have negligible effect on the quantity of Mo-99 produced but the other stable molybdenum fission products continue to increase linearly with irradiation dose. Therefore, the specific activity of Mo-99 per gram of molybdenum product (i.e. the radio-purity) decreases with irradiation time and is independent of the neutron flux as shown in Figure 7.

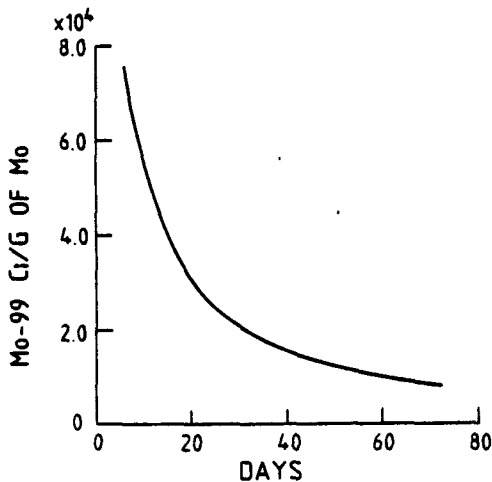


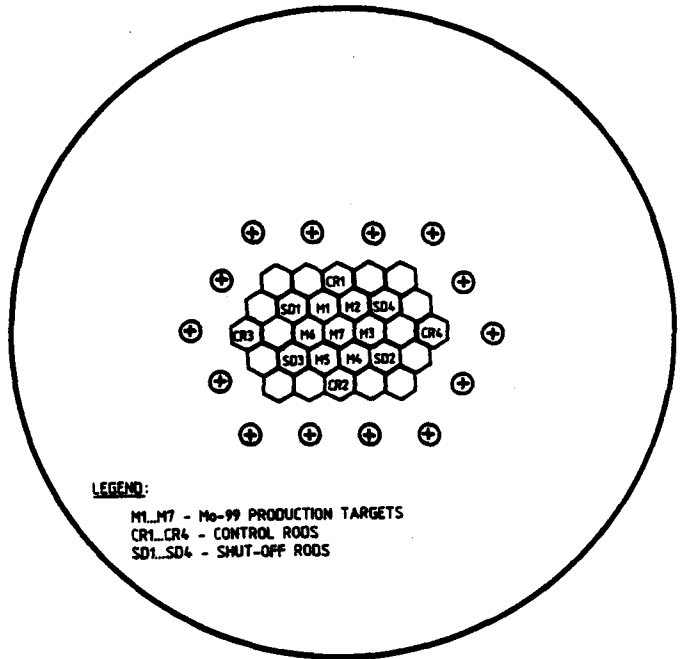
FIGURE 7: Mo-99 RADIO-PURITY VERSUS IRRADIATION TIME

The NRU reactor, with its on-power refuelling capability, is an ideal reactor for isotope production but its role as a research reactor necessitates planned shut-downs for the research program, which disrupt the Mo-99 supply schedule. At the present time, the NRX reactor acts as a back-up irradiator to NRU and maintains supply when necessary.

It is planned to use MAPLE-X as the primary Mo-99 production source in the future and MAPLE-X (like NRX) requires that the reactor be shut-down to change all fuel assemblies. Therefore, to maximize the duty factor for reactor operations, a design requirement for MAPLE-X is that a routine fuel change can be made during a short shut-down from full power without xenon poison-out of the reactor. The design minimum time to poison-out is 30 minutes and it is anticipated that Mo-99 production will require 4 or 5 target changes per week; hence, daily half-hour shut downs are anticipated.

MAPLE-X OPERATION AS A DEDICATED ISOTOPE PRODUCTION FACILITY (DIPF)

The proposed core loading arrangement for MAPLE-X operation as a DIPF is shown in Figure 8.



LEGEND:
 M1..M7 - Mo-99 PRODUCTION TARGETS
 CR1..CR4 - CONTROL RODS
 SD1..SD4 - SHUT-OFF RODS

FIGURE 8: MAPLE-X CORE LOADING

Uranium-235 targets for Mo-99 production occupy the seven central sites designated M1...M7.

MAPLE-X has 4 shut-off rods and 4 control rods which consist of cylindrical neutron absorbers operating outside the flow tube of an 18-element driver rod. The shut-off rods, together with control rods 1 and 2 are fully withdrawn during normal equilibrium operation at high power. Therefore, they form a ring of six, 18-element, fuel rods surrounding the seven central Mo-99 production sites. Control rods 3 and 4 are partially inserted to provide automatic control at equilibrium full power and xenon override capability.

Reactor shut-downs for fuel changes are effected by the simultaneous insertion of all four control rods, leaving the shut-off rods fully withdrawn. A reactor trip causes the shut-off rods to be dropped into the core.

On start-up, control rods 1 and 2 are initially the controlling rods and gradually withdraw to compensate for xenon build-up. Control is transferred to rods 3 and 4 only when rods 1 and 2 are fully withdrawn. The control system drives just one control rod out at a time but each rod in the control bank of two rods is always roughly in-line with the other, because the control system algorithm switches the drive signal alternately between the two rods every 50 to 100 millimetres of rod movement.

Fourteen sites remain to be accounted for in the MAPLE-X inner core and these contain 36-element driver fuel rods with an average burn-up of 105 MW.d (48610 MWd/Te) in the initial estimate for the equilibrium core loading. The corresponding estimated average burn-up for the 18-element fuel rod components of both the shut-off rods and the control rods is 45 MW.d (41677 MW.d/Te). This equilibrium core loading is maintained by a 28 day fuel change cycle involving the replacement of the oldest 36-element rod (172 MW.d) with a fresh 36-element rod and the oldest 18-element rod (75 MW.d) with a fresh 18-element rod.

Initial Core Loading and Transition to Equilibrium

The DIPF operation of MAPLE-X can be accomplished from day 1 following the initial commissioning program for the generic MAPLE concept. All that requires to be done is to load the core as described in Figure 8 but to replace all the 36-element fuel rods by fresh 18-element rods. Transition to the equilibrium loading simply requires individual replacement of those 18-element rods occupying 36-element rod positions by 36-element rods to match the fuel burn-up demands on reactivity.

REACTOR PHYSICS CALCULATIONS

The detailed reactor physics design calculations for the generic MAPLE concept were made by H.J. Smith, at AECL's Whiteshell Nuclear Research Establishment, using the 3-dimensional diffusion code 3DDT together with a 5 neutron energy group cross-section library derived from WIMS. These calculations, to establish the basic core design criteria, are outside the scope of this report which seeks only to investigate Mo-99 production capabilities for MAPLE-X operating at a power level of 10 MW. To do this the methods employed by H.J. Smith were adopted. WIMS cross-section sets were derived for proposed target assemblies and added to the 5-group library, and 3DDT simulations of possible core loadings were made. Results for a possible initial core loading are given in Table 3 and results for an equilibrium core loading are given in Table 4.

TABLE 3: INITIAL CORE

k_{eff} at beginning of core life	= 1.0163
Av. reactivity change from fresh 18-el rod to fresh 36-element rod	= +5.3 mk
Total power from 18-el rods	= 8.51 MW
Total Mo target power (7 targets)	= 1.49 MW
Average Mo target power/target	= 0.21 MW

TABLE 4: EQUILIBRIUM CORE

k_{eff} at equilibrium	= 1.0125
Reactor reactivity change due to burn-up	= 0.040 mk/MW.d
Exit burn-up of 36-element rods	= 172 MW.d
Exit burn-up of 18-element rods	= 75 MW.d
Reactivity change for 36-element (old/new)	= 7.7 mk
Reactivity change for 18-element (old/new)	= 3.4 mk
Total power from 36-element rods	= 5.86 MW
Total power from 18-element rods	= 2.70 MW
Total Mo target power (7 targets)	= 1.44 MW
Average Mo target power/target	= 0.21 MW
Driver rod cycle time	= 28 days
Reactivity worth of S/D rods	= 186 mk
Reactivity worth of CR#1 + CR#2	= 61 mk
Reactivity worth of CR#3 + CR#4	= 43 mk

DISCUSSION OF CALCULATIONS

The results indicate that each proposed 54 g U-235 target produces an average fission power of 0.21 MW, which corresponds to an equilibrium E.O.I. Mo-99 production of 1.1×10^4 Curies per target, in MAPLE-X operating at 10 MW. This result is comparable to NRU production and applies to both the initial core loading and to the equilibrium core configuration considered in this report. It is therefore expected to apply during the transition period between these two states and permits MAPLE-X to assume the role of a DIPF.

The values of k_{eff} quoted are 3DDT calculated values, corresponding to full power operation with equilibrium fission product concentrations, for the two cases. Some typical reactivity effects are given to indicate driver fuel management requirements for the equilibrium core. The fuel cycle requires alternately an 18-element or a 36-element rod change every two weeks, wherein the oldest rod in core is replaced with a fresh rod. During the transition from the initial core to the equilibrium core loading, 18-element rods occupying 36-element rod positions will be replaced with fresh 36-element rods on roughly the same two week schedule.

CONCLUSION

MAPLE-X can be operated at a power level of 10 MW as a dedicated facility for the production of Mo-99 and thus allow the NRU reactor to be dedicated to the research and development missions of AECL. However, the MAPLE concept is much more wide ranging than this limited objective implies and MAPLE-X is being designed to permit a future heat removal capacity greater than 10 MW. This could make Mo-99 and other isotope production from reflector sites a viable alternative to NRU in the future. It would also enable irradiations requiring high fast neutron flux levels to be performed in the MAPLE-X inner core and neutron physics experiments requiring high thermal neutron fluxes, cold sources and beam tubes to be accommodated in the D₂O reflector locations.

ACKNOWLEDGEMENTS

Most of the MAPLE-X descriptions given in this paper are summaries of the work of an AECL Company-wide design team. Any member of that team is likely to discover words and phrases in this paper which they may recognize as their own. If this happens, please accept my thanks for your unwitting contribution.

I also wish to thank E. Burke and E. Harrington for typing this paper.

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