STUDY OF NUCLIDE FIELD BEHAVIOUR IN REACTORS WITH CONTINUOUSLY CHANGING CORE PARAMETERS

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ABSTRACT

This work first deals with the reactor concept INCOGEN, which is a small (about 40 MWth) advanced fission reactor for INherently safe CO-GENeration of heat and electricity. The reactor is based on the high-temperature helium-cooled pebble-bed reactor. For ease of operation and cost reduction, fuel is continuously added over a period of more than 10 years. Study of 3-D burnup fields in the INCOGEN reactor is the primary purpose of the current research. A new burnup option of the 3-D PROMETHEUS core analysis code system was used for this study. This option incorporates a number of unique features originating from the general purpose PROMETHEUS code system, the SAS6 module as added to the SCALE code system, and the ORIGEN burnup library updated on the basis of JEF2.2. As a result, detailed burnup data is obtained with higher accuracy and efficiency.

Subsequently, similar simulation tools are used to investigate the reduced volume, activity, and lifetime of radioactive waste in a fuel cycle strategy that incorporates on-site electrorefining of the spent fuel when it is compared to the more conventional once-through fuel cycle scheme. Significant reductions in the waste activity and the duration of its existence by recycling all but the neutron-absorbing fission products back into the reactor core allows for the possibility of on-site storage as an alternative to permanent disposal, while not isolating plutonium nor introducing any attendant increased proliferation risk.

I. THE INCOGEN REACTOR CONCEPT

A. Introduction.

In recent years, a systematic effort has been put into research of an innovative reactor design based on the high-temperature gas-cooled pebble-bed reactor. The fuel is in the form of coated particles of about 1 mm in diameter, packed together in graphite pebbles of about 6 cm in diameter. A reactor core is loaded with these pebbles in a tank with a graphite reflector. Although this reactor type has existed for some time, special designs are needed to match the desired safety characteristics. The reactor concept INCOGEN is a small (about 40 MWth) advanced fission reactor for INherently safe CO-GENeration of heat and electricity. The reactor is based on the high-temperature helium-cooled pebble-bed reactor. Besides its size, a major difference with the original design of the pebble-bed reactor is its fuel strategy. The period between refuelling of all fuel will be more than 10 years. Within such an operating period fuel will be continuously added to the reactor to compensate for burn-up effects and keep the reactor critical.

As the INCOGEN high-temperature design is totally different from conventional reactor designs, it requires important adaptations and verification of the methodology and tools for reactor calculations. This encourages exploration of new possibilities which were opened by the creation of a new integrated calculation system which covers the whole range of reactor core analysis, including thermal hydraulics, dynamics and depletion. In addition, nuclear data libraries were generated based on the most recently evaluated nuclear data file JEF2.2. The cross section generation system includes the module SAS6[1] for cell or assembly burnup based on the SCALE system, but with strongly reduced computation time. The library for burnup calculations, containing three-group cross sections, radioactive decay data and fission product yield data for about 750 light nuclides, more than 100 actinides and more than 1000 fission products, was also updated[2] with JEF2.2 data. The present paper reports the results of the burnup calculations using the PROMETHEUS code system[3] which includes neutronics coupled with thermal hydraulics, offering a detailed spatial burnup treatment.

B. General Description.

The reactor concept INCOGEN[4] is a fission reactor in which HTR-type fuel pebbles are continuously loaded into the core. A high-velocity helium flow is used as a coolant. Due to the resistance of graphite to high temperatures the helium cooling gas can

be heated up to about 900°C. Incentive for investigating such a concept is the possibility to develop a system that is inherently safe with on-line addition of fuel, which is also advantageous from the fuel cycle economy point of view.

INCOGEN is a modular concept with power of a few tens of megawatts. The reactor consists of a cylindrical core volume of 4.5 meters height and 1.5 meter radius with a graphite reflector. With an initial core height of 1.05 m and 10% enriched uranium this reactor can be put into operation and then continuous addition of fuel is required. Without such fuel addition the reactor will shut down itself. An essentially new aspect of the neutronic study of this concept is the continuously changing core configuration and contents which creates additional problems to be resolved before conventional neutronic calculational tools can be applied. Other unconventional aspects are the strongly undermoderated condition of the core, the presence of a large voided cavity above the core and a number of small cavities. The undermoderated core causes strong flux gradients close to the boundary of the core with the reflector. Hence, the reflector plays a role as moderator as well. All these aspects imply that careful and detailed diffusion neutronic calculations are necessary in order to obtain reliable estimates of reactor behaviour.

II. REVIEW OF PROMETHEUS CODE SYSTEM OPTIONS

The PROMETHEUS code system, intended for analysis of nuclear reactors, was chosen as the basic tool for the study of the INCOGEN reactor concept. PROMETHEUS can solve a wide number of nuclear reactor physics problems (Figure 1) including steady-state forward and adjoint problems, prompt and delayed alpha eigenvalue problems, criticality searches, fast and slow

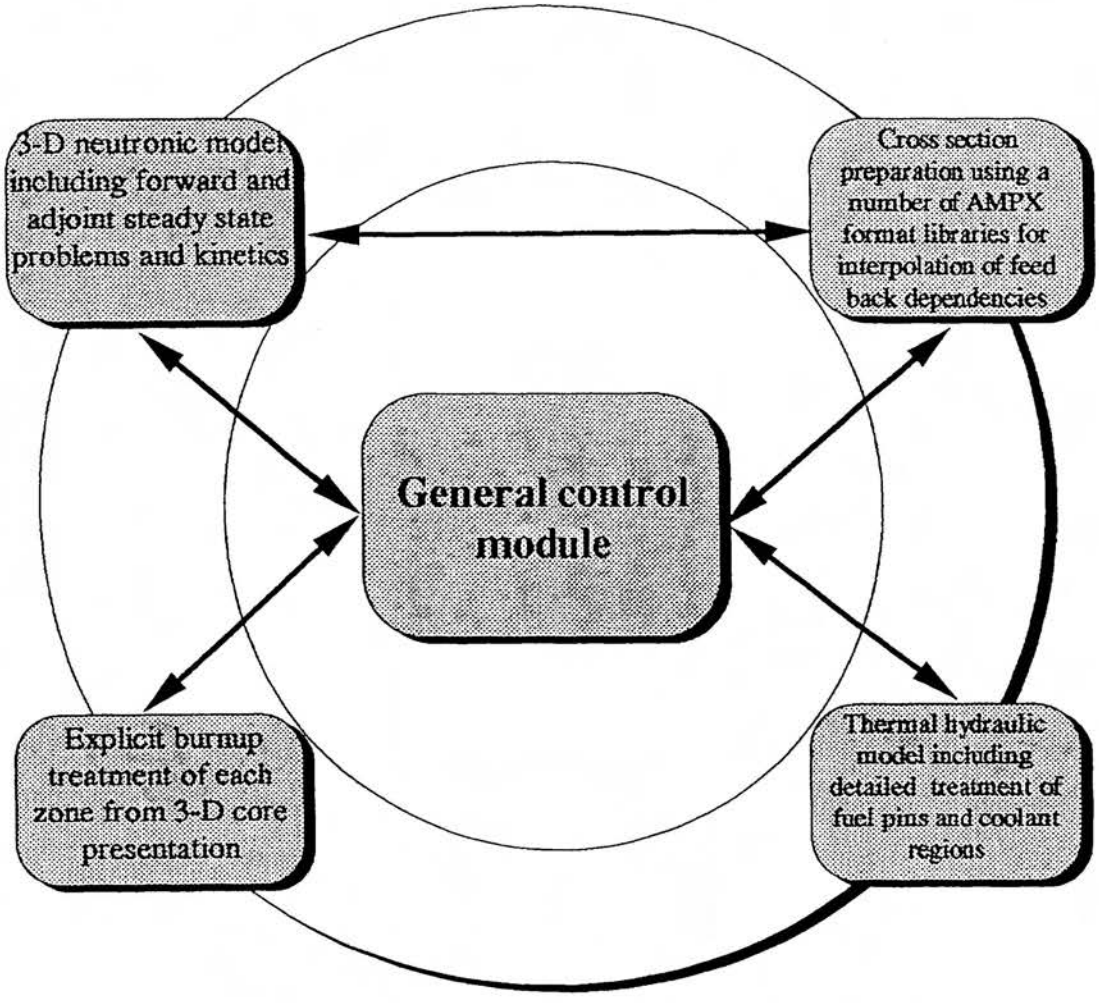

Figure 1: Layout of PROMETHEUS calculation system.

kinetics. It utilises a fine-mesh finite-difference scheme to solve the multi-group diffusion theory equations and has proven to be capable of replacing finite-difference codes of previous generations due to higher efficiency and the presence of some additional features such as treatment of non-diffusion cavities.

Three numerical techniques are available to solve the neutronic set of equations in PROMETHEUS: a simple iteration method, a point-symmetrical successive over-relaxation iterative method and a modified three-layer Chebyshev iteration scheme. The first scheme is the slowest one, but it has the widest range of stable convergence as compared to the other two. The last scheme is the fastest one. However, care should be taken to stay within the range of the scheme applicability. Selection of the scheme is done automatically in PROMETHEUS for each particular computation taking into account convergence properties of the problem, which are estimated in the initial stage of the computation.

To treat time dependency there are two solution techniques employed: the fully implicit scheme, and the prompt jump approximation, with the modified source iteration method for summation of the Neumann series being used at each time step in both techniques.[5]

The thermal hydraulic module utilises two physical models to solve transient heat-conduction problems: an adiabatic model and a detailed model solving the heat-conduction equation in a fuel pin, cladding, and non-boiling coolant for each reactor zone in 2-D or 3-D geometry of the core. In PROMETHEUS feedback effects due to changes of fuel temperature, coolant temperature, coolant density and soluble absorber concentration are basically taken into account using linear relations, while more complicated (polynomial) temperature dependencies are also included as an option.

III. BURNUP CHARACTERISATION AND COMPARISONS FOR ACCURACY AND COMPUTATIONAL EFFICIENCY

Study of 3-D burnup fields in the INCOGEN reactor was the primary purpose of the current research. A new burnup option of PROMETHEUS was used which incorporates a number of unique features composed from the general purpose PROMETHEUS code system , the SAS6 module as added to the SCALE code system, and the updated ORIGEN burnup library based on JEF2.2. These unique features determine the higher accuracy and efficiency of burnup modelling, the main aspects of which are presented in the following sections.

A. General 3-Dimensional Geometry Simulation Capability.

Burnup simulation was performed for each zone of a 2-D or 3-D reactor model. This simulation can be performed with an arbitrary number of energy groups and number of delayed neutron groups. A zone may be any geometrical region of the core with homogeneous or homogenised material composition. Arbitrary design of control rods can be treated, i.e. control rods may consist of an arbitrary number of axial zones and each of these may have a different set of cross sections while moving from one part of the core to another (for example, from core to reflector).

B. Explicit 3-Dimensional Burnup Calculations.

PROMETHEUS is capable of treating burnup problems spatially distributed throughout the reactor core using the full isotopic transition matrix rather than a limited number of transmutation chains. This was achieved by applying the approach used in creating the SAS6 depletion sequence. Burnup calculations are actually performed with the ORIGEN-S code which requires that the power, the thermal, resonance and fast threshold cross sections and relative fluxes for the thermal, resonance and fast ranges are to be pre-calculated.

Determination of the relative fluxes for each calculation zone and specified time is performed on the basis of broad group neutronic calculations done by a 3-D neutronic module. From these calculations updated three-group cross sections are obtained for all nuclides explicitly treated in the neutronic calculations. The code COUPLE is used for updating these values and the relative fluxes in the ORIGEN-S library.

Some general options like fuel reshuffling and some specific options to meet INCOGEN project requirements like core dimension change and fresh fuel addition were included and are input controllable. Thus, the calculation of depletion process can be continued after almost arbitrary changes of core geometry and composition.

The depletion runs are restartable (continuation of the depletion process), in general the range of the cross section feed-back parameters is expandable and it is possible to include new values of parameters at any later time in order to improve the accuracy of cross section interpolation, for example.

Simulation of the burnup process of the INCOGEN reactor requires all features mentioned above to be switched on. The zone-wise 3-D burnup distribution as shown in Figure 2 demonstrates the high degree of non-uniformity of burnup as a result of

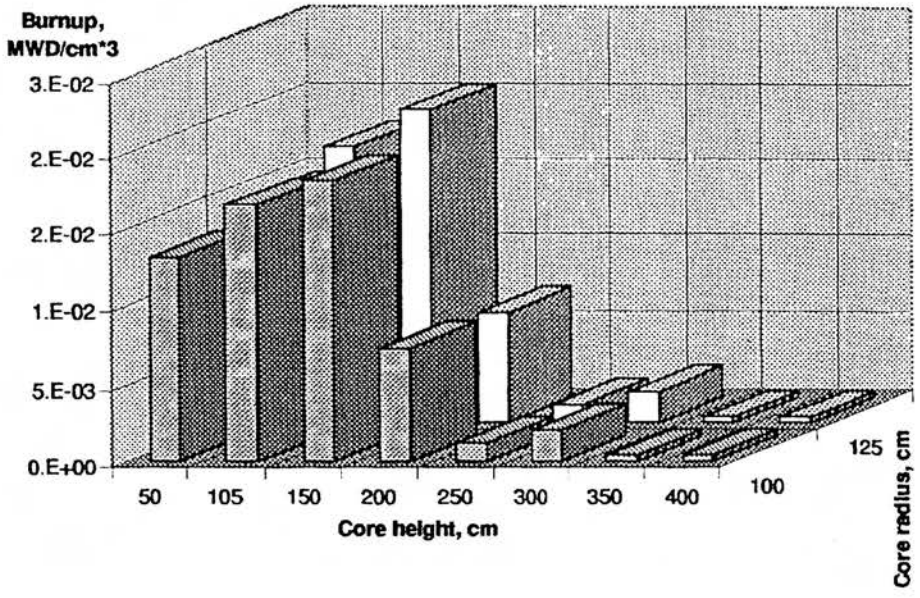

Figure 2: Burnup of INCOGEN core after 4668 exposure days.

fuel addition that has been performed step by step for maintaining criticality. The initial core height was 105 cm. The analysis of ^{239}Pu build up in two zones having similar initial contents but being irradiated by a different spectrum confirms expectations that a strong spectral variation in the core is present, ranging from an intermediate-reactor spectrum in the centre to thermal reactor conditions at the core boundaries. With 3-D burnup distributions being available, it becomes apparent that a single cell burnup calculation using the SAS2H or the SAS6 modules in the SCALE cannot provide a reliable estimate of nuclide concentrations in this reactor type, although such calculational tools include the COUPLE and the ORIGEN-S codes.

C. Higher Accuracy due to Ability to Account for All Nuclides.

Since neutronics plays a very important role in the assessment of the transmutation potential of nuclear reactors, spatially distributed burnup calculations are a vital part of such assessments. A widely used approach[6,7] is the application of a neutronic code systems (e.g. CITATION, BOLD-VENTURE) where the burnup equations are solved by an analytical solution method. Only a limited number of nuclides can be taken into account in these codes. The error associated with accounting for only a limited number of nuclides can be unacceptably high in some cases. Our estimations of this kind of error have been done for a

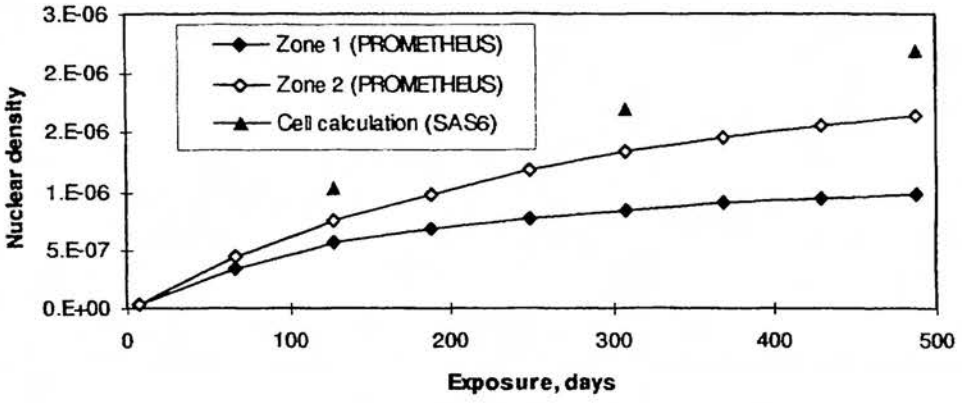

Figure 3: Comparison of zone-wise PROMETHEUS results with single cell calculations.

10 year depletion process of the INCOGEN reactor core (Figure 3,Figure 4). The neutronics have been simulated using explicit treatment of about 120 nuclides (uranium isotopes, plutonium isotopes, samarium isotopes, xenon, possible impurities, etc.). The concentrations of pseudo nuclides were calculated in every burnup step. Their contribution to the corresponding reaction rate gave an indication of the error that would arise if the pseudo nuclide technique had not been used.

The burnup calculations in PROMETHEUS include all possible actinides and fission products in every zone of a 3-D reactor model. A number of these nuclides will contribute significantly to the reaction rates. However, for the neutronic (flux) calculation only a limited number of nuclides can be taken into account to determine the cross sections. Therefore, we introduced dynamically defined pseudo fission products and pseudo actinides[8] assigned to each zone and renewed after each burnup step. Three pseudo products are defined to account for the thermal, epithermal and fast threshold neutron capture, respectively, for all nuclides not treated explicitly in the neutronic calculations. Three pseudo fissionable actinides

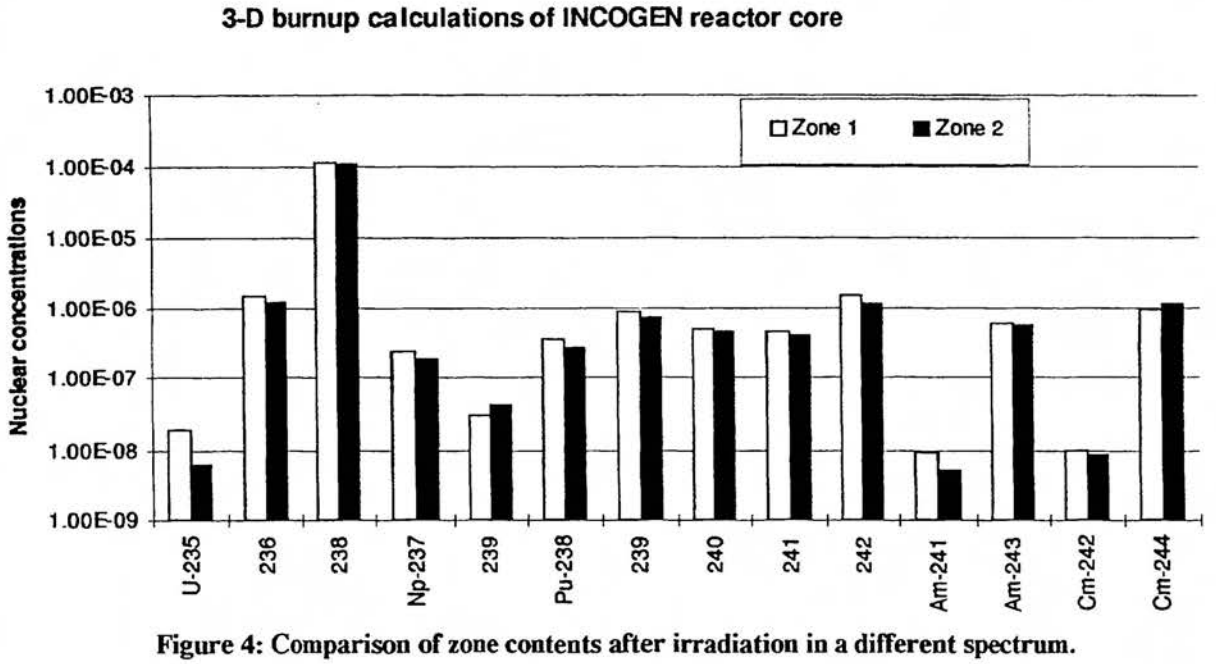

Figure 4: Comparison of zone contents after irradiation in a different spectrum.

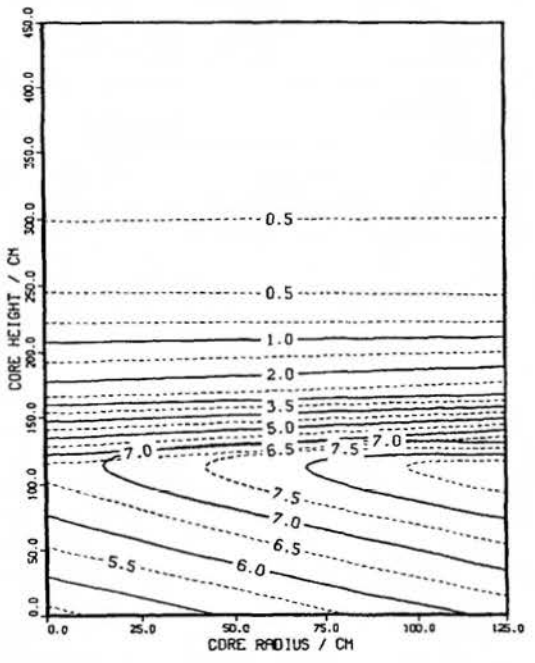

Figure 5: Errors (%) in capture properties of the INCOGEN core after 10 years of burnup if a limited number of nuclides is used for modelling.

are introduced to account for the thermal, epithermal and fast threshold fission reactions. In order to estimate the errors, we compared the results obtained by using pseudo nuclides and without using this technique (Figure 5, Figure 6). Burnup simulation of a 10%-enriched uranium core clearly indicates that neglecting the contribution of the vast majority of nuclides not treated explicitly in the flux calculation may lead to 9% error in capture properties of some particular zone of the core and up to 5% error in its fission properties (Figure 6). Furthermore, spatial distribution of the errors is very complicated (Figure 5). Its effect on the neutronic field cannot be easily predicted and eliminated, unless the pseudo nuclides technique is employed.

Besides improvement of accuracy, application of the pseudo nuclides technique allows us to make use of another advantage. Namely, the full list of nuclides (~2000) and concentrations is also available for each zone at every time step. Figure 7 shows the contents of two zones of the INCOGEN reactor core after 4668 days of burnup. Thus, detailed 3-D analysis of core burnup can be done.

D. Computational Efficiency Based on Comprehensive and Regularly Up-Dated Libraries.

For the burnup calculations, the data library used for all nuclides is based on JEF2.2. As was mentioned in the introduction, the current library containing three-group cross sections, radioactive decay data and fission product yields for about 750 light nuclides, more than 100 actinides and more than 1000 fission products, was recently updated in co-operation with the Netherlands Energy Research Foundation (ECN) replacing the original SCALE/ORIGEN library.

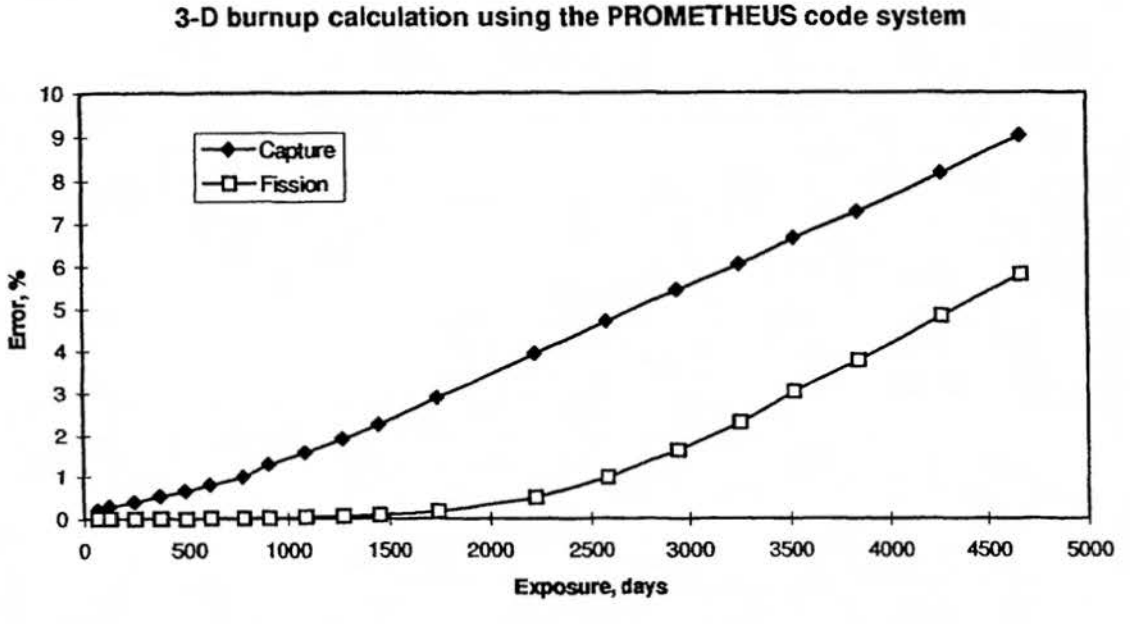

Figure 6: Estimation of errors in fission and capture properties of the INCOGEN reactor if a limited number of nuclides is used.

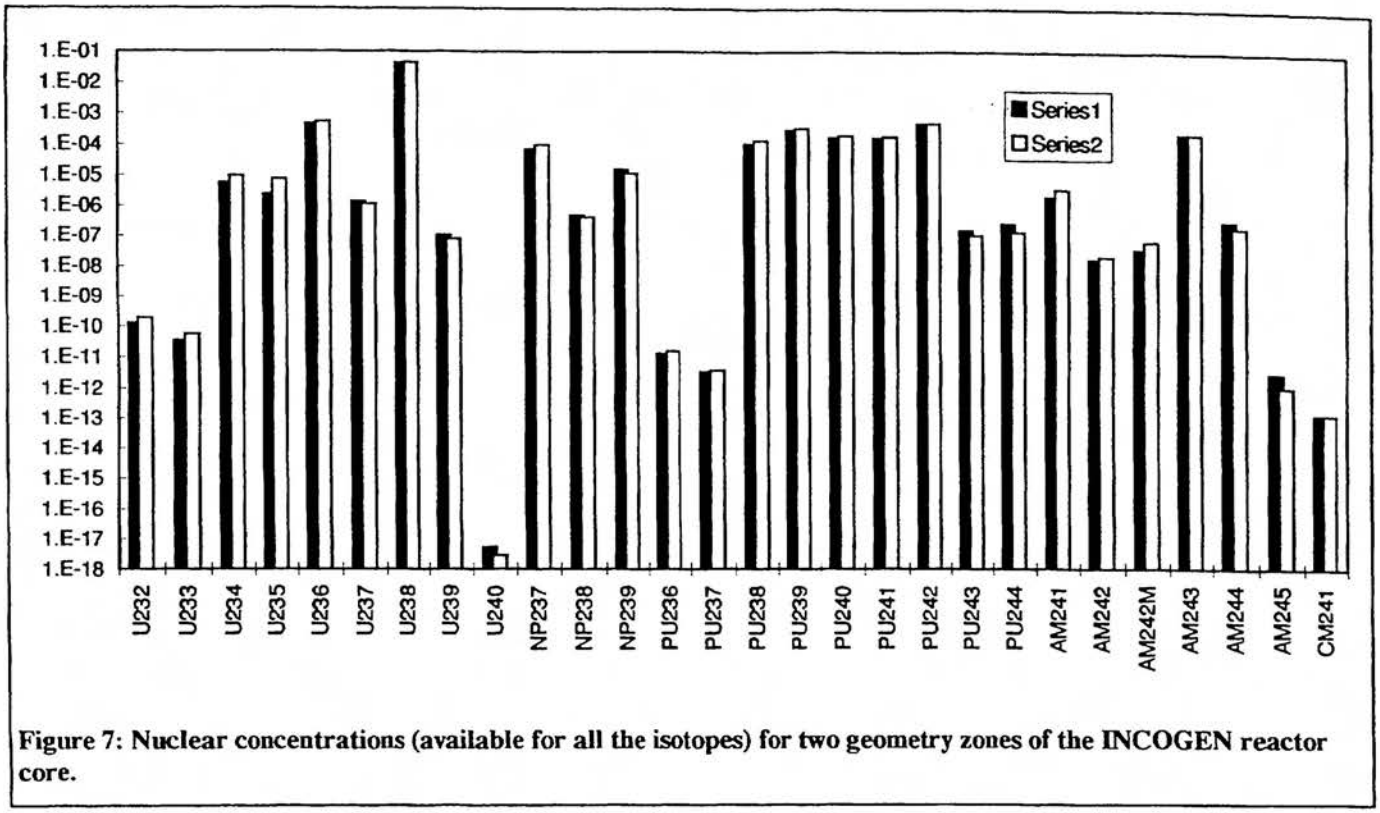

Figure 7: Nuclear concentrations (available for all the isotopes) for two geometry zones of the INCOGEN reactor core.

E. Flexibility in Using Broad-Group Problem Dependent Libraries.

In order to ensure high accuracy of results, a system to handle parameterized sets of composition files and libraries was designed and implemented in the PROMETHEUS code. The parameters are burnup and temperature, but there is some room left for other parameters. The problem of naming nuclides and libraries was solved in such a way that the nuclide original identity (ZA number, e.g.) is preserved and cross sections can be found easily by parameter pointers. The naming method is compatible with existing restrictions in the XSDRNM code, the CCCC formats, BOLD-VENTURE, etc. So that producing burnup and temperature dependent macro cross sections is a built-in routine.

The use of parameterised (burnup and temperature dependent) cross section libraries in the neutronic module is very well facilitated. Burnup and temperature field variables (such as group flux) may be defined in every mesh point as well as in every homogeneous or homogenised zone. Point-wise or zone-wise burnup and temperature are used in a selection-interpolation procedure to obtain the best cross section set. The number of libraries that can be used in a single run is not limited.

F. Treating the Multiple Occurrence of a Nuclide in a Zone.

In the homogenisation procedure for constituting a zone, a nuclide may be present in more then one geometrical region, e.g. oxygen in the UO_2 of the fuel pin and in the H_2O of the coolant. Because of the differences in the neutron flux spectrum in these regions the broad-group cross sections are different. These differences can be taken into account in PROMETHEUS. A more significant case occurs in a high-temperature pebble bed reactor where ^{10}B can appear as an impurity in the coating layer of a micro fuel particle and in the graphite matrix as well as in the unfueled graphite shell of a fuel pebble and a moderator pebble. Table 1 gives an impression about the differences between the absorption cross sections of the ^{10}B isotope contained in the

Table 1: Differences in absorption cross sections of the ^{10}B isotope contained in the coating layer and in the matrix of a typical high-temperature reactor core layout.

Group number	1	2	3	4	5	6	7
^{10}B in coating	0.91751	8.61408	65.257	264.40	613.32	1573.4	3975.5
^{10}B in matrix	0.91430	8.61733	65.517	265.86	625.16	1631.1	4180.9
Difference, %	0.3	-0.1	-0.5	-0.8	-1.9	-3.7	-4.8

coating layer and in the matrix if a 7-group condensed cross section library is considered. Differences are certainly not negligible when analysing the space-dependent neutronic distribution and reactivity effects. On the other hand, the burnup module cannot make a distinction between these two isotopes as it operates with ZA numbers, which are identical for both of them. To resolve this controversy, a composition-decomposition approach has been employed that allows all repeating isotope mixtures to be treated differently in the neutronic and burnup parts of the code system.

IV. ON-SITE SPENT FUEL MANAGEMENT BASED ON ELECTROREFINING

Computations incorporating ~2000 nuclides in burnup analyses have also been undertaken to investigate a unique strategy for managing spent nuclear fuel. Presently, the analyses have only been applied to PWR-type operations but will be extended to other reactor types -- including novel concepts like INCOGEN -- shortly. Several concepts to close the nuclear fuel cycle through burial of waste in deep, stable geological formations have been examined, but no facility has yet been provided which is acceptable to all those involved. We herein examine another approach, one in which electrorefining is used to remove neutron-absorbing fission products from the spent fuel, all other species being recycled back into the reactor core for subsequent burnup or transmutation to more stable isotopes. Because this type of reprocessing requires that only the extracted fission products be disposed of, the volume of waste, its radioactivity and its lifetime are significantly reduced, potentially allowing for on-site storage as a sufficient means of closing the fuel cycle. Also significant is the fact that the process does not involve the isolation of plutonium and thus avoids the associated risk of nuclear proliferation.

A. On-site Strategy.

Central to the spent fuel management strategy considered here is the electrorefining technique which removes some or all of the neutron-absorbing fission products, but retains the rest of the fuel -- notably the remaining fissile and transuranic materials -- for use in the manufacture of new fuel pellets. Since the main long-term (≥500 - 1000 years) radiological hazards in spent nuclear fuel are the minor actinides and other transuranic elements due to their long half-lives and large decay heats, their recycling back into the core and thus removal from the waste stream means that any isolation barriers involved in a disposal concept are no longer required to possess insolubility and immobility for ~10000 - 100000 years, or longer. Instead, only the relatively short-lived (~500 years) fission products require disposal, reducing the time scale for the integrity of the isolation barriers to remain intact, and thus easing the requirements on any type of disposal concept.

The electrorefining process has been developed over a number of years[9, 10]. It uses high temperature (500° C) molten-salt and molten-metal solvents to electrochemically remove over 99.9% of the U, Pu, and other transuranic elements from the spent fuel. All these isotopes are removed collectively as one medium, and thus there is no isolation of Pu with its attendant proliferation risks. The operation takes place in a heavily shielded hot-cell facility, and begins by chopping up the spent fuel assemblies into small pieces (~6-7 mm) which are placed in a steel basket that acts as the anode in the electrochemical cell. A voltage of ~1 V is applied between this anode and one of several cathodes; a steel cathode collects essentially pure uranium, while uranium, neptunium, plutonium, americium, curium and some rare-earth fission products are collected at a liquid cadmium cathode. The majority of the fission products -- alkali metals, alkali-earth metals and rare-earths -- are left behind in the electrolyte salt, while structural materials and a few other fission products (i.e. noble metals) remain in the molten cadmium or the anode basket [11, 12]. Fission product gases like xenon and krypton, and other gases such as tritium are recovered from the argon over-gas following the electrolysis and are stored for decay.

The same process is possible for non-metal fuelled reactors by adding some preliminary steps to the electrorefining operation. In the case of light water reactor fuel, the zircalloy cladding is first removed, followed by reduction of the oxide fuel to metallic form by a lithium reactant. The remainder of the fission product extraction process is as outlined above, and finally the lithium reactant is recovered by electrolysis prior to the manufacture of new fuel elements[13].

B. Assessment.

To evaluate the merits of such a spent fuel management strategy, a comparison of the out-of-core radioactivity -- as a function of time -- derived from such an operation is made to that of a simpler once-through fuel cycle (without reprocessing). Essentially the build-up of activity for the materials extracted during the electrorefining operation is calculated for a PWR, subject to typical operation and fuel burnup scenarios.

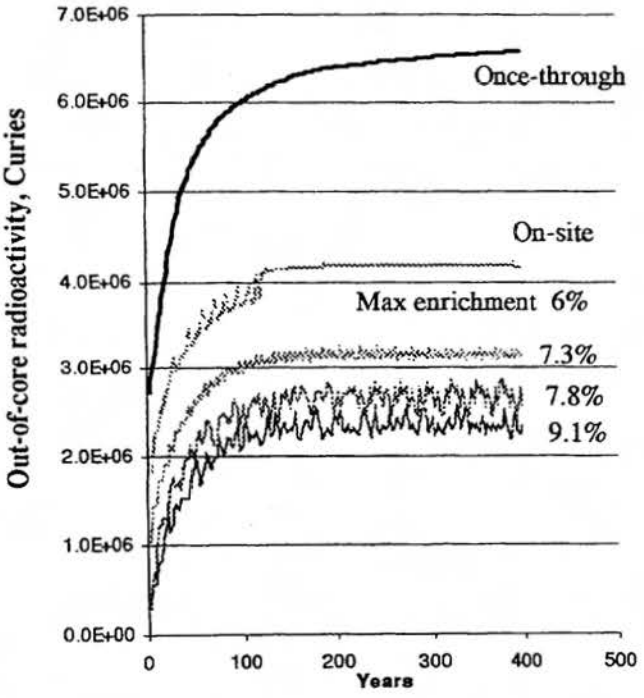

Figure 8: Comparison of the out-of-core waste accumulation in the once-through cycle with that of the on-site strategy for several values final fuel enrichment.

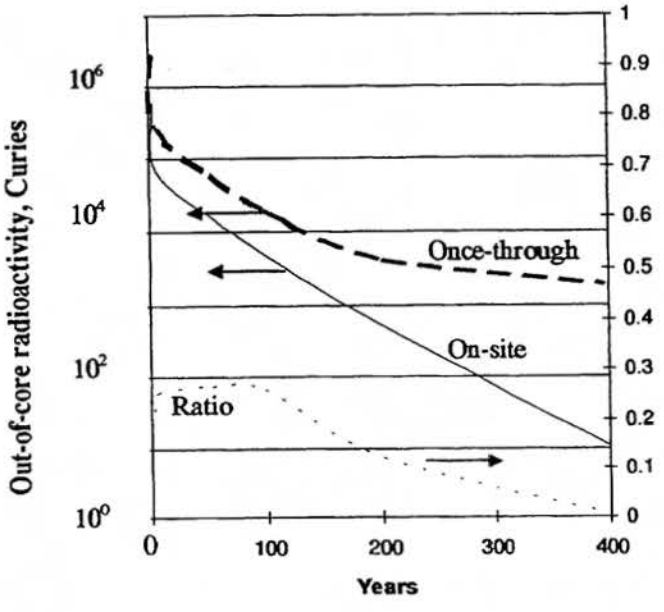

Figure 9: Activity of waste associated with one fuel assembly.

We consider a unit volume from within the reactor core initially with a fresh fuel loading. Following operation for ~3 years, the majority of the fission products are removed and replaced with new fissile fuel. Any remaining fission products and all the actinides created during the burnup period remain in the volume considered. The burnup cycle is repeated, and again removal of the most neutronically poisonous isotopes and top-up with fresh fuel occurs. The procedure is repeated, and the accumulation of activity from the waste removed between each burnup cycle is calculated. Comparison of this activity accumulation with that of removing the entire volume of fuel following each cycle provides a measure for the reduced amount of radioactive waste generated by employing this spent fuel strategy.

Burnup assessments are made with the SCALE 4.3 calculational sequences. Initially, neutronics calculations are used to generate cross-section libraries, first for individual fuel elements and subsequently for entire fuel assemblies, considering ~200 distinct species. These libraries are then used along with the initial material concentrations to assess the burnup of ~2000 isotopes over the duration of the burn period. During each burnup period an updated neutron spectrum is calculated from intermediate isotopic concentrations at several stages so that each cycle is divided into at least 3 subsections, each with its own spectrum and libraries to provide for a more accurate assessment of the material burnup.

As a typical unit volume, a fuel assembly and its surrounding structure, coolant, moderator and control mediums are chosen for use in these calculations. The power density is held constant at values representative of the average power density in a PWR, accomplished by adjusting a poison concentration in the volume itself.

Following each burnup period, all gases -- fission product or otherwise -- are removed as they are released in the electrorefining procedure. The fission products' absorption is characterized by flux-averaged macroscopic cross-sections, the least radiologically hazardous elements removed until sufficient volume and negative reactivity has been extracted to allow fresh uranium dioxide to make up for the reactivity deficit. The total volume of the assembly considered must obviously remain the same at the beginning of each burn cycle.

Calculational modelling showed that the out-of-core activity accumulation per unit power produced, and the lifetime thereof, compared to a conventional system is significantly reduced, Figure 8. For a continuously operating system with storage of the wastes on site, the activity ratio at beginning of each batch's removal was found to be the determining factor in the extent of waste activity reduction, Figure 9. This is because the relatively short-lived fission product species are the only ones of relevance in the waste stream, and thus each addition to

the waste collection becomes the major component thereof. The long-term effects are instead, primarily determined by the activity ratio after an extended length of time. The effect of the nuclear composition of the newly added fuel, i.e. low or high enrichment uranium, mixed uranium and plutonium, or plutonium alone on the degree of waste activity reduction will also be investigated.

V. CONCLUSIONS

The PROMETHEUS calculational system extended with a comprehensive burnup option has been used for study of the INCOGEN advanced reactor concept. It has been demonstrated that the burnup option integrated with general purpose neutronics and thermal hydraulics can produce qualitatively new results as compared to other commonly used related tools. The explicit burnup treatment takes into account up to 2000 actinides, fission products, and light nuclides available in the updated burnup library.

An on-site spent fuel management strategy, in which electrorefining is used to remove fission products from the spent fuel and all other species are recycled back into the reactor for subsequent burnup or transmutation to more stable nuclides is also analysed using similar calculational sequences, and appears to hold significant promise. Calculation modelling demonstrated the volume of out-of-core waste, its radioactivity and its lifetime are significantly reduced, potentially allowing for on-site storage as a sufficient means of closing the fuel cycle.

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