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Extending the Continuous Fuel Cycle Model with Additional Nuclides and Constraints

Stuart Christie, Danny Lathouwers, Jan Leen Kloosterman, Tim van der Hagen

Physics of Nuclear Reactors, Mekelweg 15, 2629 JB Delft, The Netherlands

Tel. +31-(0)-15-27-84182, Fax. +31-(0)-15-27-86422, E-mail: S.A.Christie@tudelft.nl

ABSTRACT: The continuous fuel cycle model [1] was created to predict the composition of reactor fuel after many cycles without the need for multiple lengthy burn-up calculations. Averaging the behaviour of the fuel composition over a cycle and modelling unloading and re-loading as continuous processes results in significant simplification of the equations governing fuel composition, and the information gained remains useful in studies of the long term issues of the nuclear fuel cycle. Given that the questions of sustainability and waste minimization are major issues for nuclear power, it is appropriate to return to the continuous model and examine how it may be usefully applied to contemporary problems.

We have experimented with adding nuclides to the continuous model in order to make it more applicable to modern fast reactor designs. The composition and system growth values obtained from this extended model are compared to those from more traditional fuel cycle modelling. The effects of replacing the constraint on the reactivity of each core with a power constraint are also investigated.

KEYWORDS: *Continuous fuel cycle model, multi-recycling, fast reactors, actinides*

I. INTRODUCTION

The behaviour of reactor fuel composition over multiple burn-up cycles is time consuming to model. Approximations such as homogenisation allow the time required to be reduced, in some cases by a very large amount. In a reactor that repeatedly recycles its fuel, the composition tends towards an equilibrium value. Changes over any given cycle are relatively small, making the use of a continuous approximation to the discrete burn-up cycles appear reasonable. A continuous model of this type was proposed by Ott and co-workers in a series of papers (e.g. [1], [2], [3], [4], [5], [6], [7]), of which the best summary is [1].

The original form of the continuous fuel cycle model is based around a fast breeder reactor operating on the uranium-plutonium fuel cycle. The reactor produces more fuel than it uses during operation and the excess generated is assumed to be loaded into new reactors of the same type. This process is modelled as continuous by allowing for the existence of fractional reactors, of a size appropriate to the amount of excess fuel at a given time. The model employs the observation that the majority of the plutonium content of a reactor is made up of the isotopes Pu-239 and Pu-240 and these reach their equilibrium behaviour after relatively few cycles of operation. The isotopes Pu-241 and Pu-242 are also included, although they typically take longer to reach equilibrium they make up a significant fraction of the fuel. The composition is used along with an appropriately formulated description of the production and loss rates to create a problem that can be solved to predict the equilibrium plutonium composition of the reactor and its associated fuel cycle operations.

The model is based on the construction of the production and loss matrix $\boldsymbol{C}$, which operates on the composition vector $\underline{\boldsymbol{N}}$. Given that the reactor in the model is producing its own fuel, the composition is driven towards an equilibrium state. The equilibrium composition $\underline{\boldsymbol{N}}^{\infty}$ is then the fundamental eigenvector of the production and loss matrix, and the fundamental eigenvalue is the asymptotic growth rate γ^{∞}.

$$\boldsymbol{C}\,\underline{\boldsymbol{N}}^{\infty}=\gamma^{\infty}\,\underline{\boldsymbol{N}}^{\infty} \tag{1}$$

Provided the matrix $\boldsymbol{C}$ does not change significantly over the life of the reactor, this allows the composition to be predicted without the need for lengthy burn-up modelling. The meaning of the asymptotic growth rate can be understood by considering a system of identical reactors. Any excess fuel that is produced is placed in a fractional reactor of the appropriate size for that amount of fuel. Such a system of breeder reactors would grow as time passed, and once the reactors reach the asymptotic fuel composition, the rate of growth would be γ^{∞}. Obviously this is not an accurate representation of reality, but it does provide some valuable insight. The value of γ^{∞} can be determined in a manner unaffected by changes in the fuel composition (see section II.2.) and, as such, provides a fuel independent means of comparing the breeding capabilities of different reactor designs. For a more detailed discussion of this method of describing reactor growth see [5].

The original model studies the uranium-plutonium fuel cycle and uses a composition vector comprised of four plutonium isotopes: Pu-239; Pu-240; Pu-241; Pu-242. These are the main constituents of the plutonium in the reactor fuel and the material bred in the blankets. There is no explicit requirement given to restrict the model to these four isotopes however and, with contemporary reactor fuel cycles considering the recycling of minor actinides, it is reasonable to investigate the addition of more nuclides. The continuous model could then be used to predict the asymptotic composition of such fuel cycles, while still maintaining a decrease in computational expense over fully-detailed calculations.

II. THEORY

This section will provide a summary of the theory involved in the continuous fuel cycle model and its extensions. A full explanation would require more space than is available and it is suggested the interested reader review the references given for further details.

The production and loss matrix is constructed from the microscopic reaction rates of absorption and capture reactions, along with radioactive decay constants. While there are other processes occurring that can produce or remove isotopes, these are comparatively small and can be neglected without producing serious inaccuracies. The main source of production of Pu-239 is neutron capture in U-238, followed by two β decays. Assuming all captures in U-238 lead immediately to the production of Pu-239, the balance equations for the isotopes in asymptotic operation can be written as:

$$\begin{gathered}-(\lambda_a^8+\lambda_d^8)N_8^\infty+\xi_8^\infty=0\\ -(\lambda_a^i+\lambda_d^i)N_i^\infty+\lambda_c^{i-1}N_{i-1}^\infty\\ +\xi_i^\infty-\kappa_i^\infty-\gamma^\infty N_i^\infty=0\\ i=9,0,1,2\end{gathered}\tag{2}$$

The indices 8, 9, 0, 1, 2 refer to U-238, Pu-239, Pu-240, Pu-241, Pu-242 respectively. ξ_i is the rate of feed of isotope i and κ_i the rate of removal. λ_a and λ_c represent microscopic reaction rates for absorption and capture respectively, while λ_d is the decay constant. This results in a set of five equations, expressed in seven unknowns $\boldsymbol{N}$, ξ and γ. These equations are inhomogeneous and cannot be formulated as an eigenvalue problem, as γ^∞ does not appear in the U-238 equation.

1. Constraints

Both of these problems can be resolved by the use of a pair of constraints on the asymptotic core, one on the number of atoms, equation (3), and the other on the reactivity, equation (5).

$$\sum_{i=8}^{2} N_i^\infty = K_N \tag{3}$$

Although in a reactor atoms of these isotopes are being lost due to e.g. fission, the continuous model allows their number to be held constant due to its treatment of unloading and refuelling as continuous processes. All atoms lost by whatever means are replaced and so the total number can be considered constant, labelled K_N.

Similarly, the reactivity contribution of the isotopes modelled can be considered to have a constant value, K_ρ. The contribution of each isotope to the reactivity is measured using the reactivity weight, defined as:

$$w_i^\rho = \nu\sigma_{f_i} - \sigma_{a_i} \tag{4}$$

This approximation to the reactivity is treated further in [8]. The constraint can then be expressed as:

$$\sum_{i=8}^{2} w_i^\rho N_i^\infty = K_\rho \tag{5}$$

The two constraints can be combined to remove U-238 from the set of equations being studied, while still describing the production of Pu-239.

$$\frac{1}{K_N}\sum_{8}^{2} N_i^\infty = 1 \tag{6}$$

$$\sum_{8}^{2} w_i^\rho N_i^\infty = \frac{K_\rho}{K_N}\sum_{8}^{2} N_i^\infty \tag{7}$$

$$\sum_{8}^{2}\left(w_i^\rho - \frac{K_\rho}{K_N}\right)N_i^\infty = \sum_{8}^{2} b_i N_i^\infty = 0 \tag{8}$$

Re-arranging as above to obtain a sum over the isotopes that equals zero allows the value of N_8^∞ to be expressed in terms of the plutonium composition.

$$N_8^\infty = \sum_{9}^{2} -\frac{b_i}{b_8} N_i^\infty = \sum_{9}^{2} \beta_i N_i^\infty \tag{9}$$

With a little further re-arrangement, an equation that takes the form of a weighted sum over the plutonium isotopes and is equal to unity can be achieved. This can then be used to make the inhomogeneous terms in the balance equations homogeneous.

$$\sum_{9}^{2} \beta_i N_i^{\infty} + \sum_{9}^{2} N_i^{\infty} = K_N \qquad (10)$$

$$\sum_{9}^{2} \left(\frac{1+\beta_i}{K_N} \right) N_i^{\infty} = \sum_{9}^{2} d_i N_i^{\infty} = 1 \qquad (11)$$

The production of Pu-239 from U-238 can now be described in terms of β_i and a sum over the four plutonium isotopes. The balance equations for the plutonium isotopes can be written:

$$\begin{gathered} -\left(\lambda_a^9 + \lambda_d^9\right) N_9^{\infty} + \lambda_c^8 \sum_{9}^{2} \beta_i N_i^{\infty} \\ +\left(\xi_9 - \kappa_9\right) \sum_{9}^{2} d_i N_i^{\infty} = \gamma^{\infty} N_9^{\infty} \\ -\left(\lambda_a^i + \lambda_d^i\right) N_i^{\infty} + \lambda_c^{i-1} N_{i-1}^{\infty} \\ +\left(\xi_i - \kappa_i\right) \sum_{9}^{2} d_i N_i^{\infty} = \gamma^{\infty} N_i^{\infty} \end{gathered} \qquad (12)$$

This formulation is slightly different to that used in [1], but it is felt to be clearer as the feed and removal terms have been left separate and explicit.

2. Isotopic breeding worths

The adjoint fuel cycle problem is simple to obtain from equation (1).

$$\boldsymbol{C}^{*} \boldsymbol{w}^{*} = \gamma^{\infty} \boldsymbol{w}^{*} \qquad (13)$$

The fundamental eigenvector of this equation is composed of the isotopic breeding worths. These can be understood as representing the fuel importance for breeding, meaning the contribution of an isotope to the asymptotic critical mass [6].

The breeding worths can also be used in calculating the asymptotic growth rate of the system.

$$\gamma^{\infty} = \frac{\left(\boldsymbol{w}^{*}, \boldsymbol{C}\, \underline{\boldsymbol{N}}\right)}{\left(\boldsymbol{w}^{*}, \underline{\boldsymbol{N}}\right)} \qquad (14)$$

Using these values as weight factors in this equation allows the asymptotic growth rate to be determined without knowing the asymptotic composition. Using this expression produces a value for the asymptotic growth rate which is stationary to perturbations to the composition [5].

3. Additional Nuclides

It is straightforward to extend the composition vector to include plutonium isotopes above Pu-242. The form of the balance equations remains the same. Once elements other than plutonium are considered however, a few extra changes are necessary. Here the effects of including Am-241 will be considered, as it is close to the plutonium isotopes already included in both atomic and mass numbers, is typically the most populous isotope of americium, and provides a good illustration of the changes necessary to include other isotopes in the system.

The main issue in the inclusion of Am-241 is the production path, as it is not a result of neutron capture in lower isotopes, but rather of β-decay of Pu-241. The existing set of equations allows for decay as a loss method, but not for production (beyond the special case of capture in U-238 producing Pu-239). The balance equation must be adjusted to reflect this. The index 51 will be used to refer to Am-241 in the equations.

$$\begin{gathered} -\left(\lambda_a^{51} + \lambda_d^{51}\right) N_{51}^{\infty} + \lambda_d^{1} N_1^{\infty} \\ +\left(\xi_{51}^{\infty} - \kappa_{51}^{\infty}\right) \sum_{i} d_i N_i^{\infty} = \gamma^{\infty} N_{51}^{\infty} \end{gathered} \qquad (15)$$

It is assumed here that americium is being recycled alongside plutonium, leading to asymptotic growth of the americium content of the fuel.

A more generic form of the balance equation for the various nuclides can be written, which covers all possible production paths, rather than simply capture.

$$\begin{gathered} -\left(\lambda_a^i + \lambda_d^i\right) N_i^{\infty} + \sum_{j \neq i} \lambda_{j \rightarrow i}^{j} N_j^{\infty} \\ +\left(\xi_i^{\infty} - \kappa_i^{\infty}\right) \sum_{k} d_k N_k^{\infty} = \gamma^{\infty} N_i^{\infty} \end{gathered} \qquad (16)$$

i is the index of the nuclide in question, *j* represents any nuclide except *i*, and *k* all nuclides in the composition vector.

4. Constant Power Constraint

It can be assumed that the power produced by a given reactor core will be held constant. This should be true not only in asymptotic operation, but throughout the life of the reactor. This is true for both the physical reality and the continuous approximation, unlike the two constraints in the original model, which can only really be considered accurate in the asymptotic phase of the continuous description. It can be expressed relatively easily in terms of the microscopic fission reaction rate λ_f^i and the power produced per fission q_i.

$$\sum_{i} q_i \lambda_f^i N_i = K_Q \qquad (17)$$

Other constraints can also be envisaged, for example neglecting the power per fission term to produce a fission rate constraint, or looking at the mass of actinides in the core to give an actinide mass constraint. In each case, the most useful expression gives the value of the constraint in terms of a sum over all the nuclides in the model. Exchanging one of the existing constraints with a new one leads to different forms of d_i and β_i. For two generic constraints, labelled *A* and *B*, the derivation would be:

$$\sum_i w_i^A N_i^\infty = K_A \tag{18}$$

$$\sum_i w_i^B N_i^\infty = K_B \tag{19}$$

$$\frac{1}{K_A} \sum_i w_i^A N_i^\infty = 1 \tag{20}$$

$$\sum_i w_i^B N_i^\infty - \frac{K_B}{K_A} \sum_i w_i^A N_i^\infty \tag{21}$$

$$\sum_i \left(w_i^B - \frac{K_B}{K_A} \right) N_i^\infty = \sum_i b_i N_i^\infty = 0 \tag{22}$$

$$N_8^\infty = \sum_{i'} -\frac{b_{i'}}{b_8} N_{i'}^\infty = \sum_{i'} \beta_{i'} N_{i'}^\infty \tag{23}$$

$$\sum_{i'} \left(\frac{1+\beta_i}{K_N} \right) N_{i'}^\infty = \sum_{i'} d_{i'} N_{i'}^\infty = 1 \tag{24}$$

i' is used here to represent the full set of nuclides except for U-238.

III. REACTOR MODEL

It is necessary to compare results from the continuous model to those of full burn-up calculations in order to determine their accuracy. The values used in the continuous model must also be obtained from a reactor model. This section describes the model used for these calculations.

A simplified model of a sodium cooled fast reactor was used for the burn-up calculations in this work. The model was based on that used by Borg [7], and is a 1000 MWe oxide fuelled breeder reactor. The reactor is divided into 6 regions, two representing the inner and outer core, two the axial blanket and two the radial blanket. These are surrounded by a steel reflector. Slices through the reactor model are shown in Figure 1. The central core was initially loaded with 13.7% Pu-239. The outer core was initially loaded with 15.5% Pu-239. The blankets were started with natural uranium. Burn-up was modelled using the TRITON module of the SCALE code system [9].

In order to determine the asymptotic behaviour of the reactor, a script was written to produce a new TRITON input file based on the composition given at the end of the previous cycle. The compositions of the new core materials were determined by mixing the remaining fuel in the old materials with that produced in the blankets. The appropriate reloading was determined using the same constraints as the continuous model and assuming a feed of solely U-238. The blankets were simply reloaded with the same composition with which they began the previous cycle. The model was run through thirty burn-up cycles, by which time the fuel composition had largely settled into the asymptotic composition. The growth rate was estimated at each cycle by calculating the number of cores that could be fuelled with the material available and comparing it to the same number from the previous cycle.

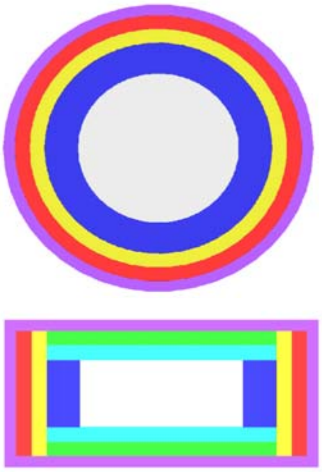

Figure 1: Horizontal and vertical slices through the SFR model

IV. RESULTS

1. Composition Evolution

The predictions from the continuous model were compared to the results of the TRITON calculations. The evolution of the fuel composition over the burn-up calculations is shown in Figure 2. The dashed lines give the average values over each cycle. These results show that, as expected, the fuel composition did tend towards an equilibrium value, and that this state was reached within a realistic number of cycles.

It is evident that as the composition changes, the cross-sections and fluxes in the reactor will change, affecting the production and loss matrix. The question of how many cycles are necessary to achieve a reasonable prediction of the equilibrium composition should be investigated. Figure 3 shows how the prediction of the equilibrium composition from the continuous model changes over thirty cycles, with the values given in relative terms to those of the thirtieth cycle.

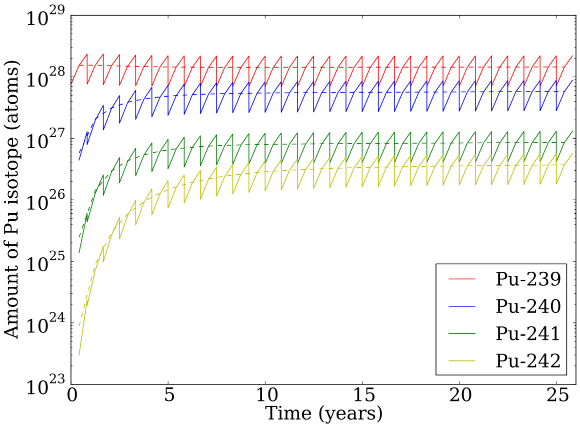

Figure 2: Evolution of plutonium composition over 30 cycles comparing values throughout the cycle (solid lines) with cycle averages (dashed lines)

It can be seen that the predictions changed over the cycles, starting off with some significant differences to the final value, and tending towards it. The accuracy of the prediction also appears to be related to the amount of the isotope in question, with the values for Pu-239 being quite accurate throughout and those for Pu-242 starting off considerably different to the final value. The predictions do not move smoothly towards their final value, this is caused by statistical variations in the TRITON results, due to the use of Monte Carlo methods to determine the flux in the core.

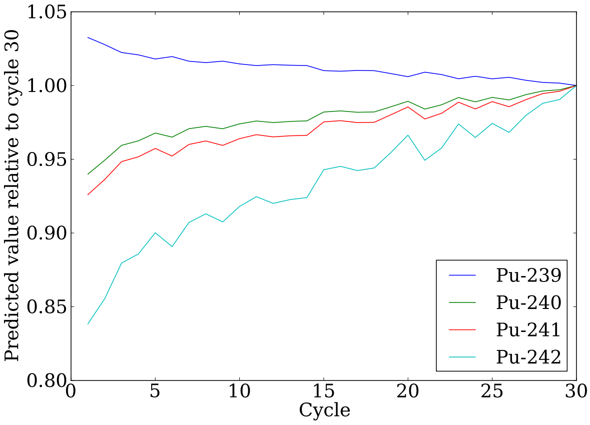

Figure 3: Continuous model predictions of equilibrium composition relative to the value at cycle 30

Comparing the predicted values to those seen during the evolution of the fuel shows that the predictions are quite accurate. The actual fuel composition tended towards that predicted and appeared to stabilise around those values. These results are shown in Figure 4. The results have been normalised to the value at the end of the thirtieth cycle of TRITON calculations to allow for a good comparison.

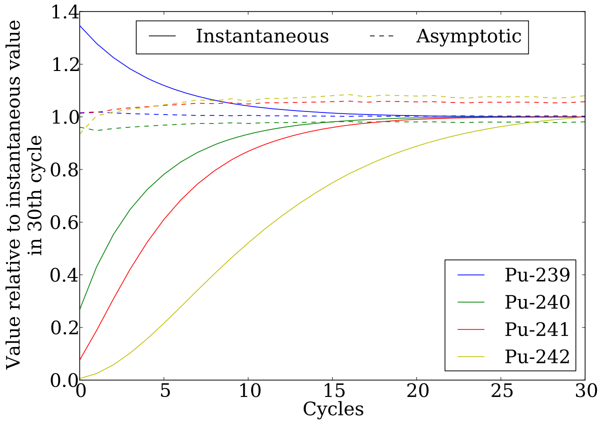

Figure 4: Comparison of composition and predicted values for asymptotic composition relative to the value at the 30th cycle

2. System growth

The growth of the system as a whole can also be predicted from the continuous model and compared to that seen in the TRITON calculations. The burn-up model showed the system moving towards an asymptotic composition, eventually increasing the amount of each nuclide at the same rate, as shown in Figure 5. The dashed lines in Figure 5 show the asymptotic growth of each isotope and how they tend towards it with time.

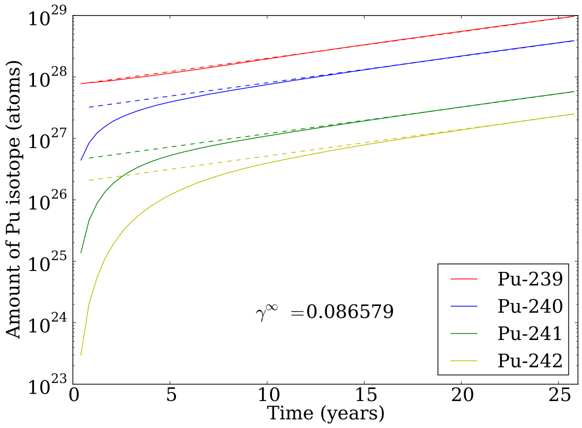

Figure 5: Evolution of plutonium inventory in a system of breeder reactors

The growth rate of the system can be found from the eigenvalue problem. In combination with the isotopic breeding worths from the adjoint problem, it is possible to predict the asymptotic growth rate using a non-asymptotic fuel composition. A comparison of the predicted asymptotic growth rates from each cycle and the growth rate seen in that cycle is shown in Figure 6.

It can be seen that after a number of cycles the asymptotic growth rate prediction stays relatively stable. The value found is close to that seen in the instantaneous case for the thirtieth cycle.

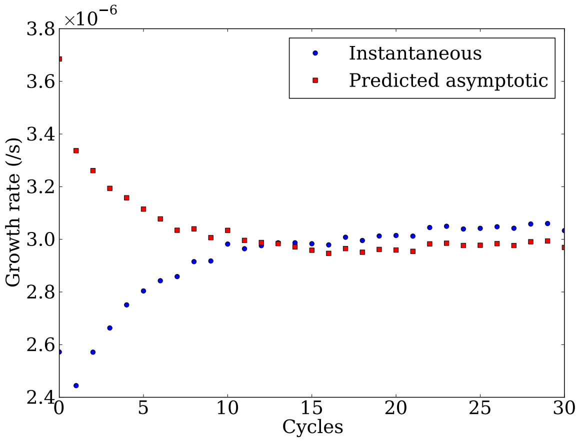

Figure 6: Comparison of predicted asymptotic growth and instantaneous growth rates

3. Additional Nuclides

The isotope Am-241 was added to the system described by the continuous model. A set of TRITON calculations in which plutonium and americium were recycled was also performed. The values from these calculations were compared to the predictions from the continuous model. The composition predictions over thirty cycles are shown in Figure 7.

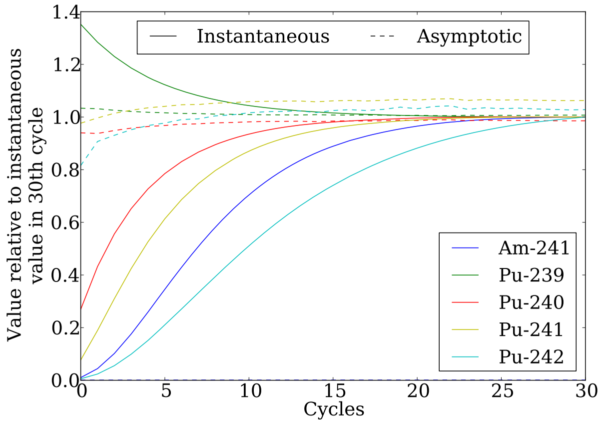

Figure 7: Evolution of composition and predictions of asymptotic composition including Am-241

The predictions of the asymptotic composition remain fairly constant throughout the thirty cycles. The results for plutonium isotopes were quite good, differing from the values of the thirtieth cycle by a few percent at most. The results for americium were significantly worse, with the prediction from the thirtieth cycle being 3.7×10^{-6}% compared to the actual figure of 3.9×10^{-3}%. This may be due to the model not including some pathways for the production of Am-241, although the main route of production via the decay of Pu-241 was present. Without usable values for Am-241, there was little point in including higher isotopes, as the incorrect value would have had a knock-on effect.

4. Constant Power Constraint

The two constraint equations play an important role in the set-up of the continuous model. The power produced by a core will be held constant during operation, so this constraint applies even during the transition phase of the reactor's life, while the actinide number and reactivity constraints only apply once in asymptotic operation. The constant power constraint was substituted into the continuous model in place of the reactivity constraint. This new model was used to predict the asymptotic composition of the fuel from thirty cycles of calculations of a plutonium recycling system with a pure U-238 feed. The TRITON code uses a constant power approximation in its calculations, so no changes were required to model this. The results produced were compared to those from the model using the reactivity constraint in Figure 8.

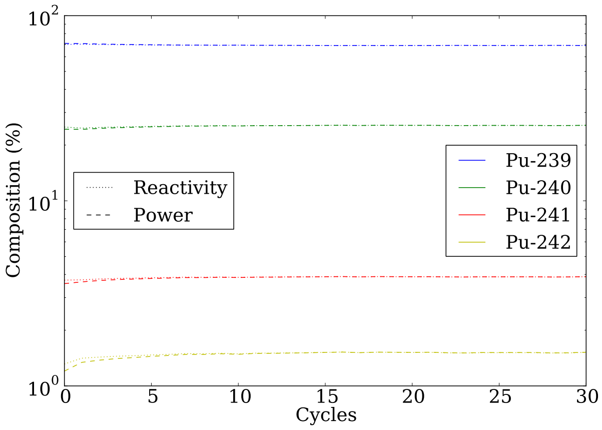

Figure 8: Comparison of asymptotic composition predictions from models using constant power and reactivity constraints over 30 cycles

The two sets of results were very similar, differing only slightly for the first few cycles and being in close agreement thereafter. While this means the constant power constraint may be used in the model without negatively affecting the results, there is also no improvement in their quality. It is possible that if the reloading of the TRITON model was not determined using the reactivity constraint, the constant power constraint would have a more pronounced effect.

V. CONCLUSIONS

The continuous model can be used in the study of fuel cycles of breeder fast reactors. The approximations made allow values such as the asymptotic composition to be estimated without the use of a full burn-up study. The values found for the asymptotic composition are a reasonable estimate when compared to those from full burn-up calculations. They are not completely accurate however, and cannot replace detailed analysis when precise values are required. The growth rate values are of similar accuracy as the composition predictions, but are subject to the same caveats.

Extending the model by including Am-241 was not successful. The accuracy of the predictions of the plutonium composition remained unchanged. The values found for the americium were much worse and cannot be considered useful. It is possible some of the production and loss paths for this isotope were missing from the model, although this seems unlikely to explain the magnitude of the differences seen.

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The inclusion of the constant power constraint in place of the reactivity constraint had little effect on the quality of the results. While this means there is no issue preventing the use of the constant power constraint, there is also little to be gained. Possibly a system with larger shifts in fuel reactivity from cycle to cycle would produce more interesting results.

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