

THORIUM-PLUTONIUM FUEL FOR LONG OPERATING CYCLES IN PWRs - PRELIMINARY CALCULATIONS

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Abstract

Preliminary calculations have been carried out to investigate the possibility of extending operating cycle length in PWRs by use of Thorium-Plutonium mixed oxide fuel (Th-MOX). The calculations have been carried out in two dimensions, using the fuel assembly burnup simulation program CASMO-5. The reload scheme and the operating parameters are modelled on the Swedish PWR Ringhals 3 and a normal UOX fuel assembly designed for this reactor has been used as a reference. Results show that an extension of the currently employed 12-month operating cycle length is possible, either with a burnable absorber or with a modified fuel assembly design, assuming the same 3-batch reload scheme as currently used in Ringhals 3.

The initial k_{∞} of the new Th-MOX fuel design was designed not to exceed that of the reference UOX fuel. The power peaking factor is initially significantly lower than the reference, but slightly higher later in the life of the fuel assembly. All reactivity coefficients are within acceptable range. The worth of control rods and soluble boron are lower than the reference, as expected for a plutonium-bearing fuel.

1 Introduction

The objective of the work described herein is to investigate the possibility of extending operating cycle length in PWRs by use of thorium-plutonium mixed oxide fuel (Th-MOX), as a part of the fuel development program at the Norwegian company Thor Energy. The broad goal of our company's research is to develop a safe, reliable and cost effective nuclear fuel for use in currently operating and future Light Water Reactors (LWR).

The general viability of Th-MOX fuel in LWRs (mainly PWRs) has been confirmed by several recent studies (Trellue et al., 2011; Fridman and Kliem, 2011; Tsige-Tamirat, 2011; Insulander Björk et al., 2011; Todosow and Raitses, 2010). In addition, the neutronic and physical properties of Th-MOX indicate that the fuel is not only viable, but may also improve the economy of a nuclear power plant by allowing for longer operating cycles and hence a higher availability of the reactor. Most importantly, the good material properties (Cozzo et al., 2011; Karam et al., 2008) of the thorium-plutonium mixed oxide ceramic indicates that Th-MOX fuel may be capable of sustaining higher burnups than UOX based fuel types. Secondly, the currently practiced uranium enrichment limit of 5% U-235 does not affect Th-MOX, which can be loaded with high amounts of plutonium. Finally, the slow depletion of Th-MOX makes it possible to achieve

high burnups without having an excessively high initial multiplication factor (Insulander Björk et al., 2011).

In this paper, we modify a standard PWR fuel assembly design in order to achieve longer cycles, and also investigate some of the consequences of these modifications. A PWR fuel assembly loaded with Th-MOX fuel is simulated under normal PWR conditions, using the fuel assembly burnup simulation program CASMO-5. This allows for an accurate assessment of the depletion behaviour and an estimation of the changes in neutronic safety parameters which may be expected.

The calculation tools and methods used in this study are described in Section 2 and the reactor system which has been simulated in Section 3. The results in terms of depletion behaviour and neutronic safety parameters are presented in Section 4 and conclusions are drawn in Section 5.

2 Method

2.1 Neutronic Simulation Software

The calculations are carried out in two dimensions, using the fuel assembly burnup simulation program CASMO-5 (Rhodes et al., 2007), together with the cross section library ENDF/B-VII.0 (Chadwick et al., 2006). These calculations do not take into account neutron leakage or spectral interactions between assemblies, except for that the neutron energy spectrum is corrected for the energy dependence of the leakage probability. Predictions of the depletion behaviour by this method are fairly accurate. Reactivity coefficients can also be estimated, but the results should mainly be regarded as indicative of the change which may be expected if UOX fuel is exchanged for Th-MOX fuel.

2.2 Estimation of reactivity coefficients

The reactivity coefficients were estimated by perturbation calculations, using Equation 1. Here, k_U is the infinite multiplication factor at normal operating conditions and k_P is the infinite multiplication factor at slightly perturbed operating conditions, where one of the operating parameters has been changed by a certain step Δx . The reactivity coefficient is denoted α_x .

$$\alpha_x = \frac{k_P - k_U}{\Delta x \cdot k_U}. \quad (1)$$

The control rod worth is estimated similarly, basically using Equation 1 but excluding Δx and letting k_P denote the multiplication factor with the control rod inserted.

2.3 Total energy release

The energy release demanded from a fuel assembly is determined by reactor operation parameters; the number of fuel assemblies in the core N , the reactor power P_{th} , the operating cycle length t_C and the number of fuel batches n that are simultaneously present in the core, which is equivalent to the number of cycles that a fuel assembly is used. For clarity, we wish to compare equivalent systems; the reference system with UOX fuel designed for 12-month cycles and the investigated system with Th-MOX fuel designed for longer cycles. For the systems to be equivalent, we keep N , P_{th} and n equal, and vary only t_C . As shown in Equation 2, the total energy

release is also the product of the fuel assembly mass M and the achievable discharge burnup D_B . An increase in t_C can thus be achieved by increasing M or D_B or both.

$$E_{tot} = M \cdot D_B = \frac{P_{th} t_C n}{N} \quad (2)$$

To determine the achievable discharge burnup from the CASMO-5 output, the reactivity of the core can be calculated as the average of the reactivities of each of the three fuel batches at end of cycle (Driscoll et al., 1990). This calculated value of the core reactivity must then be adjusted to take into account the neutron leakage out of the reactor, which is not accounted for in the infinite lattice simulations. Thus, for the core to be critical, the reactivity must comply with the condition set by Eq. 3.

$$\sum_{i=1}^n \rho(i \cdot B_C) = \Delta\rho \quad (3)$$

Here, $\rho = (k_\infty - 1)/k_\infty$ is the reactivity and $\Delta\rho$ and n are the neutron leakage and the number of batches, respectively. B_C is the cycle burnup. For determining the achievable discharge burnup, the largest B_C is sought for which ρ fulfills the condition, and the discharge burnup B_D is calculated as $n \cdot B_C$.

3 The modelled system

The modelled system is the PWR Ringhals 3 in Sweden. The operating parameters used in the simulation code input are listed in the table below.

Table 1: Reactor operating parameters at hot full power.

Parameter	Value
Power density [kW/dm ³]	105.50
Coolant temperature [K]	575
Cycle average boron concentration [ppm]	600

The reference fuel is a UOX fuel assembly designed for a standard 12-month cycle in the same reactor. It comprises 264 fuel pins, arranged in a standard 17-by-17 lattice, of which 12 contain Gd₂O₃ as a burnable absorber.

The three Th-MOX fuel designs were based on the reference design, with two important modifications.

Firstly, the uranium oxide fuel was exchanged for a mixture of thorium and plutonium oxides. The Pu isotope vector used in the Th-MOX fuel was: 2% ²³⁸Pu, 53% ²³⁹Pu, 25% ²⁴⁰Pu, 15% ²⁴¹Pu and 5% ²⁴²Pu. This corresponds to the Pu vector in spent LWR fuel burnt to approximately 42 MWd/kgHM, if reprocessed immediately (World Nuclear Association).

Secondly, the fuel rod thickness was increased in two of the designs, resulting in two parallel effects. As previously mentioned, one way to increase the total energy release, and hence the cycle length, is to increase the fuel mass, which can obviously be done by increasing the fuel rod radius. A more indirect effect of increasing the fuel rod radius is that the hydrogen-to-heavy-metal ratio is decreased when some of the water volume is replaced with fuel. This makes the neutron spectrum slightly harder, favouring neutron capture reactions at the cost of fission

reactions. The result is that the fuel depletes more slowly, meaning that the multiplication factor decreases more slowly. This makes it possible to maintain criticality for a longer time without a need for a very high multiplication factor for the fresh fuel assemblies. This favours an even power distribution within the reactor.

A third alternative was developed, using an Integral Fuel Burnable Absorber (IFBA), which is a thin layer of zirconium boride applied to the surface of the fuel pellets (Biancheria and Allio, 1969). This also makes it possible to avoid an initially very high multiplication factor but does not add the benefit of an increased fuel mass. The reason to choose IFBA over the more common Gd_2O_3 mixed into the fuel matrix is that it is very difficult to create a ternary mixture of ThO_2 , PuO_2 and Gd_2O_3 of sufficient homogeneity.

4 Results

4.1 Depletion behaviour

Three designs were created, two for 18-month cycles (Th-MOX-18 for the alternative with thicker fuel rods and Th-MOX-18-BA for the alternative with IFBA) and one for 24-month cycles (Th-MOX-24). The infinite multiplication factor k_∞ is shown in Figure 1, together with the k_∞ for the reference fuel assembly (UOX-ref). It is clear from the graph for UOX-ref that the k_∞ at the beginning of life is suppressed by the burnable absorber. This is done to avoid that the fresh fuel assemblies carry a too high power load compared to the other ones. The k_∞ of a fuel assembly with identical design and enrichment but without the burnable absorber is plotted for comparison (UOX-woBA). In order to maintain this power balance, the Th-MOX-18 and Th-MOX-24 assemblies are designed to have the same initial k_∞ as the reference assembly. From the graph, it is clear how the slower decrease of k_∞ in these assemblies results in a larger cycle length despite the fact that it starts from approximately the same point in all cases. The effect of the IFBA in the Th-MOX-18-BA assembly is also evident from the gradual increase of k_∞ at the beginning of life.

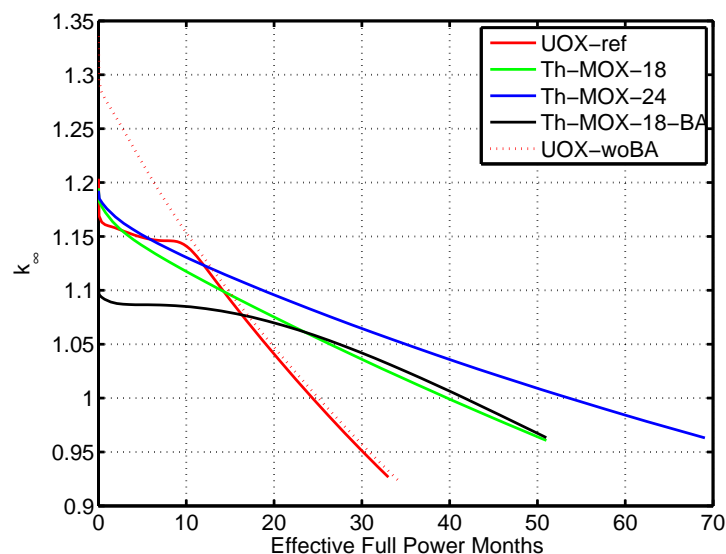


Figure 1: Infinite multiplication factor dependence on time. Note that the cycles include one month of down-time for revision, meaning that the fuel designed to operate for three 12-month cycles only needs to operate for $3 \cdot 11 = 33$ effective full power months, etc..

The changes made to the fuel assembly design are quantified by the properties listed in Table 2.

What we see in this table is that the discharge burnup has been increased in order to reach the desired cycle length, and to achieve this, the plutonium fraction has also had to be increased.

In addition, the fuel mass has been increased for Th-MOX-18 and Th-MOX 24, by increasing the fuel rod radius. An increase in the fuel rod radius results in a decrease of the flow area. This in turn causes an increased pressure drop over the fuel assembly and a smaller amount of water to carry away the same amount of energy, i.e. a higher enthalpy at the core exit. In order to avoid boiling at the core exit, the mass flow must be increased in correspondence with the flow area decrease, i.e. by 11% for Th-MOX-18 and by 33% for Th-MOX-24. The need for an increased mass flow and the increased flow resistance over the entire core makes it necessary to retrofit the thermo-hydraulic system to some extent, if the fuel is to be used in an existing reactor. The pumps would most probably need to be exchanged for more powerful ones, and all parts related to the flow would need to be checked in order to make sure they can operate at the increased flow rate. In addition, it would be necessary to verify that the safety injection systems are able to cool down the core also with the reduced flow area.

The higher pressure drop also means that these fuel assemblies cannot be used together with unmodified fuel assemblies in the same core, since the difference in flow resistance between the assembly types would cause cross-flow between the assemblies, resulting in vibrations and thermo-hydraulic effects which may be difficult to model.

The alternative to use IFBA instead of increasing the fuel rod radius avoids the problems related to the changed thermo-hydraulics. However, it does not, as mentioned, come with the added benefit of increased fuel mass. Thus, the fuel needs to reach a burnup of 73 MWd/kgHM in order to obtain a cycle length of 18 months, similar to what is demanded to reach a 24-month cycle length with the increased fuel rod radius. The lower fuel mass in the Th-MOX-18-BA case is due to the lower density of the (Th,Pu)O₂ ceramic compared to the UO₂.

It should be noted that the required discharge burnup in all Th-MOX cases is considerably higher than the currently employed limits on UOX fuel. There are several reasons to believe that these burnups will in fact be achievable. Firstly, although the burnup is high, the residence time within the reactor for the Th-MOX-24 design is only six years. Whereas this residence time is unusually long for PWR fuel, it is not an extreme residence time for BWR fuel. Secondly, it is expected that thorium based fuel will have a capacity to keep its integrity to high burnup, for several reasons. One is that the thermal conductivity of the thorium plutonium mixed oxide ceramic is higher than that of a corresponding uranium oxide ceramic, up to about 14% Pu (Cozzo et al., 2011). This means that the fuel temperature will remain lower in the pellet, causing less swelling, less PCMI (pellet-cladding mechanical interaction) and a larger margin to fuel melting. Another reason is that fission gases have a lower mobility in the thorium oxide matrix, which results in a lower fission gas release. The higher thermal conductivity also has a beneficial effect on the fission gas release, since the lower fuel temperature makes the fission gases diffuse more slowly. It should however be noted that the larger fuel pin radius may

Table 2: Mass flow and related data for the UOX reference and the Th-MOX assemblies.

Fuel name	UOX-ref	Th-MOX-18	Th-MOX-24	Th-MOX-18-BA
Cycle length [months]	12	18	24	18
B _D [MWd/kgHM]	44	63	72	73
Initial Pu fraction [%]	0.0	11.5	13.6	12.9
Fuel mass [kg]	461	498	592	431
Fuel rod radius [mm]	4.75	5.10	5.56	4.75
Flow area reduction [%]	-	10	25	-

outweigh the higher thermal conductivity, resulting in higher fuel temperatures in these designs. One possible remedy for this is to use annular fuel pellets.

4.2 Power Distribution

In addition to the initial k_{∞} , another factor of importance for avoiding putting an excessive power load on any individual fuel rod is the power distribution within the fuel assembly. The intra-assembly power peaking factor F_{int} is the ratio between the power of the rod having the highest power and the average rod power. This parameter is plotted in Figure 2, which shows that the F_{int} is initially very high for UOX-ref, due to the use of burnable absorber in only a few fuel rods, but then falls off very rapidly. For the Th-MOX-18 and Th-MOX-24 assemblies, however, the F_{int} is far more constant and fairly low throughout life. This is achieved by using two different levels of Pu content in the fuel rods. The need to do this arises from the fact that the fuel rods are made thicker. When the water gap between rods becomes narrower, the extra neutron moderation added by the water in the control rod guide tubes becomes more important, resulting in comparatively high power in the rods located close to the guide tubes. Two levels of Pu content are used also in the Th-MOX-18-BA case, although the moderation is similar to the reference assembly, which makes the F_{int} initially very low. When the IFBA has burnt out, however, the power distribution successively grows more inhomogeneous.

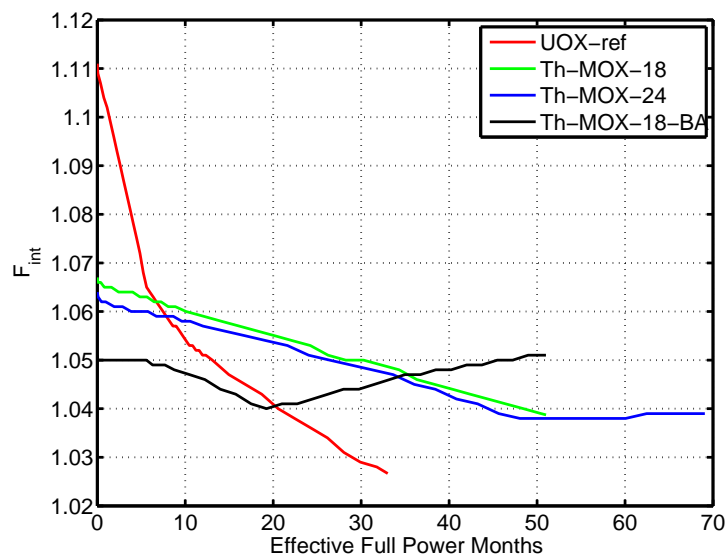


Figure 2: F_{int} dependence on time, where F_{int} is the ratio between the power of the rod having the highest power and the average rod power.

4.3 Safety parameters

The safety parameters generally change with burnup. However, since fuel assemblies having different burnup are simultaneously present in the core, the general core behaviour is reasonably well captured by the burnup averaged value, which is shown in Table 3.

The average Moderator Temperature Coefficient (MTC) is negative in all cases, meaning that an increase in temperature causes a decrease in reactivity, a negative feedback effect which counteracts the initial disturbance. Compared to the reference case, it is slightly larger in amplitude in the Th-MOX-18 case and slightly smaller in the Th-MOX-24 case. This somewhat unexpected

effect is due to two competing effects affecting the MTC. The lower hydrogen-to-heavy-metal ratio caused by the increased fuel rod thickness, which takes overhand in the Th-MOX-18 case, generally gives a larger negative MTC. A higher Pu-content, on the other hand, generally gives a less negative MTC, and it is this effect which takes overhand in the Th-MOX-24 case.

The void coefficient is also negative in all cases, which means that in the event that the coolant would start boiling, resulting in a small void fraction, this would cause a decrease in reactivity. The void coefficient shows a behaviour similar to that of the MTC. The coolant void reactivity listed below the void coefficient is the reactivity response to the entire core becoming voided at once, that is a Loss Of Coolant Accident (LOCA). This important safety parameter is also negative, as demanded, but smaller for the Th-MOX fuel types than for the UOX reference.

The Isothermal Temperature Coefficient (ITC) corresponds to the change in reactivity caused by simultaneous heating of all present materials. This parameter is thus important for the safety at startup. The ITC was calculated at isothermal zero power conditions at different temperatures with all control rods inserted, and was found to be negative at all temperatures for all fuel types. It is smaller for the Th-MOX fuel, but clearly on the safe (negative) side even at room temperature.

For both the MTC, void coefficient, coolant void reactivity and the ITC, the values for Th-MOX-18-BA are similar to those for Th-MOX-18, except that they are less negative at the beginning of life due to the presence of IFBA. This effect is due to the very strong thermal absorption being reduced by the loss of moderation.

Also the Doppler coefficient, or the fuel temperature coefficient, is negative for all fuel types. It is larger in amplitude for the Th-MOX assemblies, due to the specific resonance structure of ^{232}Th , and also due to the larger fuel mass in the Th-MOX-18 and Th-MOX-24 cases. This provides a very rapid feedback mechanism in the event of power or temperature transients.

The delayed neutron fraction is strongly affected by the fissioning isotope. The numbers in Table 3 mainly reflect the fact that the delayed neutron fraction is considerably lower (less than half) in ^{239}Pu and ^{233}U compared to ^{235}U . This indicates that the Th-MOX-fuelled reactor will respond more rapidly to changes in the operating conditions compared to the reference. However, this effect is very similar to what is encountered with normal U-MOX fuel (Insulander Björk and Fhager, 2009), and most modern reactor designs, which are capable of using 100% U-MOX fuel, should thus be capable of using Th-MOX fuel to the same extent. Furthermore, the large negative Doppler coefficient provides a very rapid negative reactivity feedback which mitigates the effect of the lower delayed neutron fraction.

Finally, Table 3 shows clearly that the worth of control rods and soluble boron are significantly reduced in the Th-MOX cases. This is expected, since the presence of large absorption reso-

Table 3: Safety parameters for the evaluated fuel assembly designs. All parameters are calculated for the operating conditions listed in Table 1, except for the isothermal temperature coefficient which is calculated for isothermal conditions at 293 K and 0% void fraction.

Fuel name	UOX-ref	Th-MOX-18	Th-MOX-24	Th-MOX-18-BA
Moderator temperature coefficient [pcm/K]	-33	-37	-27	-33
Void coefficient [pcm/% void]	-97	-103	-73	-91
Coolant void reactivity [pcm]	-33765	-16664	-10636	-13868
Isothermal temperature coefficient [pcm/K]	-11.0	-7.7	-8.9	-6.6
Doppler coefficient [pcm/K]	-2.9	-3.5	-3.8	-3.2
Delayed neutron fraction [pcm]	567	364	361	363
Control rod worth [pcm]	23355	16777	15281	16240
Boron worth [pcm/ppm]	7.2	2.1	1.1	2.5

nances in several of the Pu isotopes causes the flux to decrease at the thermal energies where the control materials have their highest absorption cross sections. This suggests that it might be difficult to obtain negative shutdown margins with Th-MOX fuel, given the control rods and the soluble boron available in Ringhals 3. However, there are several ways to mitigate this effect. One is to use boron enriched in ^{10}B in the moderator. Another is to use control rods of a different composition, using materials with higher absorption cross sections than the commonly used silver-indium-cadmium alloy. Finally, there is room for installing eight additional control rod clusters in Ringhals 3, indicating that in general, a larger number of control rods could be a way to compensate for their lower worth when used for controlling Pu-containing fuel.

5 Conclusions

The conclusion of these preliminary calculations is that it does indeed seem possible to extend operating cycle length by using Th-MOX fuel, but that retrofitting of the reactor system will be necessary if the fuel designs proposed here are to be used in an existing reactor. All reactivity coefficients are within acceptable limits and the challenges encountered with respect to the delayed neutron fraction and the worth of control materials are similar to those encountered by the introduction of conventional U-MOX fuel. The necessary retrofitting, as indicated by this assessment, amounts to upgrading the control rods and soluble boron systems and, if the fuel rod radius is to be increased, upgrading the flow system to handle higher mass flows and pressure drops over the core.

These preliminary calculations will be followed up by more advanced calculations, studying the effects of the changes in the fuel properties, including full core simulations, transient simulations and thermal-hydraulic simulations. The fuel assembly design could also be further refined; for example, a combination of the two approaches of increasing the fuel rod radius and using IFBA may be advantageous.

In addition, the claim that Th-MOX fuel can be driven to higher burnups than UOX fuel must be proven. The thermal-mechanical operating limit, which effectively limits the discharge burnup from a material point of view, depends on several properties such as thermal conductivity of the fuel material, fission gas release and pellet-cladding interaction. All of these will be assessed in an upcoming test irradiation planned by Thor Energy (Kelly and Fhager, 2010), which will start in December this year.

Finally, it should be noted that Th-MOX fuel has a large design flexibility. The fuel assembly design can also be adapted for extracting as much energy as possible out of the inserted Pu, which calls for an increased rather than a decreased hydrogen-to-heavy metal ratio. Which design features are desired depends on several factors, most importantly availability of plutonium.

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References

- A. Biancheria and R. J. Allio. Nuclear fuel elements, February 1969. US patent 3.427.111.
- M.B. Chadwick, P. Obložinský, M. Herman, N.M. Greene, R.D. McKnight, D.L. Smith, P.G. Young, R.E. MacFarlane, G.M. Hale, S.C. Frankle, A.C. Kahler, T. Kawano, R.C. Little, D.G. Madland, P. Moller, R.D. Mosteller, P.R. Page, P. Talou, H. Trellue, M.C. White, W.B. Wilson, R. Arcilla, C.L. Dunford, S.F. Mughabghab, B. Pritychenko, D. Rochman, A.A. Sonzogni, C.R. Lubitz, T.H. Trumbull, J.P. Weinman, D.A. Brown, D.E. Cullen, D.P. Heinrichs, D.P. McNabb, H. Derrien, M.E. Dunn, N.M. Larson, L.C. Leal, A.D. Carlson, R.C. Block, J.B. Briggs, E.T. Cheng, H.C. Huria, M.L. Zerkle, K.S. Kozier, A. Courcelle, V. Pronyaev, and S.C. van der Marck. ENDF/B-VII.0: next generation evaluated nuclear data library for nuclear science and technology. *Nuclear Data Sheets*, 107(12):2931–3060, 2006.
- C. Cozzo, D. Staicu, J. Somers, A. Fernandez, and R.J.M. Konings. Thermal diffusivity and conductivity of thorium-plutonium mixed oxides. *Journal of Nuclear Materials*, 416(1-2): 135–141, September 2011.
- M. J. Driscoll, T. J. Downar, and E. E. Pilat. *The linear reactivity model for nuclear fuel management*. Americal Nuclear Society, 555 North Kensington Avenue, La Grange Park, Illinois 60525, 1990.
- E. Fridman and S. Kliem. Pu recycling in a full Th-MOX PWR core. part I: Steady state analysis. *Nuclear Engineering and Design*, 241(1):193–202, 2011.
- K. Insulander Björk and V. Fhager. Comparison of thorium-plutonium fuel and MOX fuel for PWRs. In *Proceedings of Global 2009*, 2009. Paper 9449.
- K. Insulander Björk, V. Fhager, and C. Demazière. Comparison of thorium-based fuels with different fissile components in existing boiling water reactors. *Progress in Nuclear Energy*, 53(6):618–625, August 2011.
- M. Karam, F.C. Dimayuga, and J. Montin. Fission gas release of (Th,Pu)O₂ CANDU fuel. Technical Report CW-124950-CONF-002, Atomic Energy of Canada Limited, 2251 Speakman Drive, Mississauga, Ontario, Canada L5K 1B2, September 2008.
- J. F. Kelly and V. Fhager. Designing a thorium fuel irradiation experiment. In *Advanced Fuel Pellet Materials and Fuel Rod Design for Water Cooled Reactors*, number 1654 in TECDOC, pages 165–171. International Atomic Energy Agency, Vienna, 2010.
- J. Rhodes, L. Deokjung, and K. Smith. *CASMO-5/CASMO-5M, A Fuel Assembly Burnup Program, User's Manual*. Studsvik Scandpower, Inc., 2007.
- M. Todosow and G. Raitses. Thorium based fuel cycle options for PWRs. In *International Congress on Advances in Nuclear Power Plants 2010, ICAPP 2010*, volume 3, pages 1888–1897, San Diego, CA, 2010.
- H.R. Trellue, C.G. Bathke, and P. Sadasivan. Neutronics and material attractiveness for PWR thorium systems using monte carlo techniques. *Progress in Nuclear Energy*, 53(6):698–707, 2011.
- H. Tsige-Tamirat. Neutronics assessment of the use of thorium fuels in current pressurized water reactors. *Progress in Nuclear Energy*, 53(6):717–721, 2011.
- World Nuclear Association. Plutonium, April 2009. URL www.world-nuclear.org/info/inf15.html. Date accessed: November 3, 2011.