LA-UR-12-22319
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Intended for: INMM 53rd Annual Meeting, 2012-06-15 (Orlando, Florida, United States)

THE CALCULATION OF BURNABLE POISON CORRECTION FACTORS FOR PWR FRESH FUEL ACTIVE COLLAR MEASUREMENTS

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ABSTRACT

Verification of commercial low enriched uranium light water reactor fuel takes place at the fuel fabrication facility as part of the overall international nuclear safeguards solution to the civilian use of nuclear technology. The fissile mass per unit length is determined nondestructively by active neutron coincidence counting using a neutron collar. A collar comprises four slabs of high density polyethylene that surround the assembly. Three of the slabs contain ^{3}He filled proportional counters to detect time correlated fission neutrons induced by an AmLi source placed in the fourth slab.

Historically, the response of a particular collar design to a particular fuel assembly type has been established by careful cross-calibration to experimental absolute calibrations. Traceability exists to sources and materials held at Los Alamos National Laboratory for over 35 years. This simple yet powerful approach has ensured consistency of application.

Since the 1980's there has been a steady improvement in fuel performance. The trend has been to higher burn up. This requires the use of both higher initial enrichment and greater concentrations of burnable poisons. The original analytical relationships to correct for varying fuel composition are consequently being challenged because the experimental basis for them made use of fuels of lower enrichment and lower poison content than is in use today and is envisioned for use in the near term. Thus a reassessment of the correction factors is needed. Experimental reassessment is expensive and time consuming given the great variation between fuel assemblies in circulation. Fortunately current modeling methods enable relative response functions to be calculated with high accuracy. Hence modeling provides a more convenient and cost effective means to derive correction factors which are fit for purpose with confidence.

In this work we use the Monte Carlo code MCNPX with neutron coincidence tallies to calculate the influence of Gd_2O_3 burnable poison on the measurement of fresh pressurized water reactor fuel. To empirically determine the response function over the range of historical and future use we have considered enrichments up to 5 wt% $^{235}U/^{tot}U$ and Gd weight fractions of up to 10 % Gd/UO_2. Parameterized correction factors are presented.

INTRODUCTION

Active neutron coincidence collar measurements are routinely applied in nuclear safeguards inspections for the nondestructive assay of the linear density of ^{235}U in fresh fuel assemblies. Neutrons from an AmLi source are used to initiate induced fissions in the assembly and the resulting neutrons are counted in coincidence to distinguish them from the AmLi background. A passive assay can also be used to measure the linear density of ^{238}U in the fuel assembly but here we shall discuss only the active procedure.

Historically response function corrections based analytical approximations derived from experimental results on mock-ups have been applied. In practice there exist a wide variety of fuel assembly types with characteristics that differ substantially from the historical mock-ups. For instance compared to the mock-ups modern fuel assemblies have different arrays, may employ axial zones of different enrichment, and may have vacancies or burnable poison rods according to various patterns. The number of arrangements is vast and too large to explore experimentally. In this work we apply Monte Carlo simulation methods to explore the behavior more conveniently over a broad conjectured operational space [1-3].

The practical difficulties arise in part because commercial Pressurized Water Reactors (PWRs) use several core management techniques applied in combination to deliver near optimal economic performance within safety and other constraints [4]. Fuel excess reactivity is built into the core design in order to achieve the desired design cycle length (burnup). Along with chemical shim (boric acid concentration) adjustments, burnable poisons are typically used to holddown the excess reactivity during their first cycle of irradiation. Burnable poisons are also used by the core designers to flatten the assembly and pin radial power distribution across the core as well as set a negative moderator reactivity temperature coefficient. To accommodate higher fuel burnups a combination of higher fuel enrichment and higher loadings of burnable poison is needed. This challenges existing fresh fuel measurement strategies which make us of active thermal neutron collars corrected using algebraic factors based on limited historical data acquired on substantially different load patterns. Even for a given reactor and fuel assembly type the fuel designer and core designer have many different options. To make head way we make some largely arbitrary assumptions in order that we may evaluate the response of a standard PWR active collar in realms of potential future interest.

PROBLEM DEFINITION

In this work we consider a Low Enriched Uranium (LEU) fuel assembly (FA) comprising a 16x16 array on a pitch of 1.43 cm with 20 vacant Zircalloy® Guide Tube (GT) positions. This defines a 16x16-20=236 array of fuel pin positions. The FA has reflective symmetry about the center lines perpendicular to the sides, so we need only describe the lower left quadrant. In Cartesian coordinates the five empty GT locations in this quadrant are at locations (3,3), (3.6), (5,7), (6,3) and (7,5).

We assume the FA is fabricated using LEU enriched from feed stock of natural enrichment. The ^{234}U/U enrichment in weight percent (wt%), f_{234}, is estimated in terms of the ^{235}U/U enrichment in wt %, f_{235}, according to the relation $f_{234} = 0.005408+0.00928x(f_{235}-0.710971)$. Since ^{234}U is of minor im
portance we consider this relation to be adequate for our purposes for LEU up to f_{235}=5 wt %.

We adopt gadolinia, Gd_2O_3, as the burnable poison. Gd has a strong non-1/v cross section dropping off rapidly at about 0.2 eV and so remains almost black throughout its way to depletion. It is assumed that Gd_2O_3 is blended with UO_2 and scintered into pellets. Pellets in the absorber zone of a Burnable Poison Rod (BPR) are assumed to have the same density as pure UO_2 fuel of the same enrichment independent of the abundance of gadolinia. BPRs with the highest gadolinia weight percent have the lowest fuel content. Due to lower thermal conductivity of the gadolinia fuel, the ^{235}U enrichment BPRs is lowered with respect to the fuel in the remainder in the FA to ensure that these rods do not violate any of the fuel performance design criteria e.g. centre line temperature. The ^{235}U enrichment of the 'carrier fuel' containing gadolinia is taken to be 2.6 wt % in all cases. The gadolinia content is expressed as a weight percent with respect to the total fuel mass, that is as [mass of Gd_2O_3]/[mass of UO_2 + mass of Gd_2O_3] expressed in wt %. As indicated perfect chemical stoichiometry was assumed. The fuel density was taken to be 0.955 time Theoretical Density (TD) adjusted for the dependence of the molar mass of U on enrichment with the reference value being TD=10.97 g/cc for natural uranium at 20°C.

We consider FAs with 0, 4, 8, 12, 16, 20 and 24 BPRs. The locations of the BPRs are at the discretion of the core designer and depend on factors beyond the scope of this paper. We illustrate the different configurations we selected in Fig.1. The arrangements are all symmetrical about the diagonal. In general we tried to place BPRs close to a GT (to take advantage of the more highly moderated spectrum on these regions), not next to each other (to avoid self-shielding), and not too close to the edge of the FA. The degrees of freedom are limited, especially as the number of BPRs is increased and in practice some of our choices are unlikely to be used in reality. But for the purposes of studying instrument response and as an exercise in simulating deviations for better understanding of the robustness of the safeguards information the instrument can provide we feel this is a reasonable set.

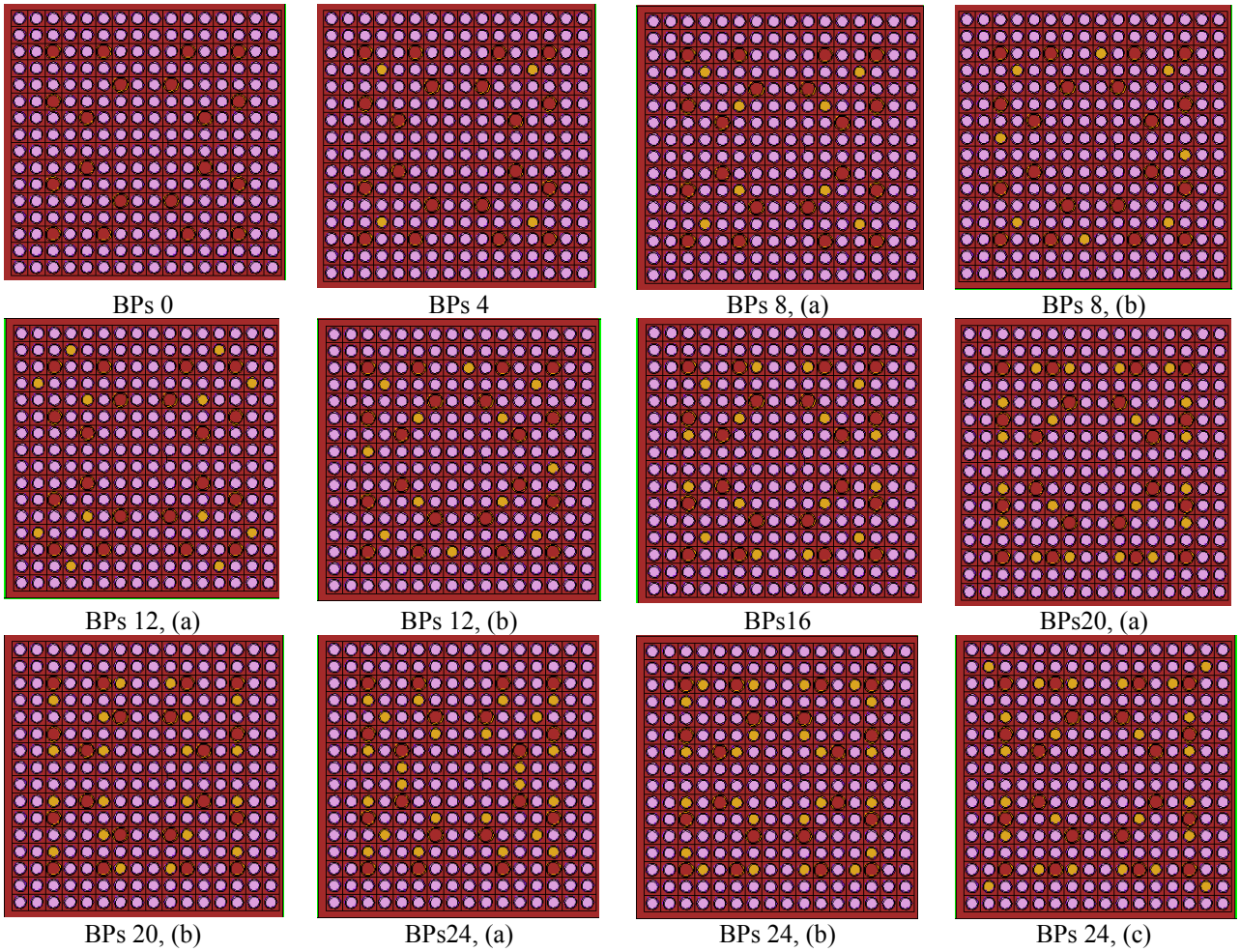

Fig. 1 Summary of the various fuel assembly configurations modeled. BPs indicates the number of BPRs presented in the scenario.

The active length of the fuel pins at 20°C is taken to be 390 cm. The bottom 30 cm and top 40 cm of the BPRs is taken to share the enrichment of the 'normal' fuel in the rest of the assembly. We apply the constraint that the average ^{235}U enrichment across the entire FA is the same for all FAs regardless of the number of BPRs present. We take the maximum permissible fuel ^{235}U enrichment to be up to 5 wt %. Then, for example, if we consider the extreme configuration to be 24 BPRs of 6 wt % gadolinia we can come in below this constraint by setting an average FA enrichment of 4.8 wt %. This is illustrated in TABLE 1 along with two other cases, namely gadolinia contents of 8 and 10 wt % also. Thus, we have two ways of changing the overall absorber content of the FAs containing BPRs. We can change the number of rods or change the gadolinia weight percent within each of the BPRs. To reiterate we have kept the FA average enrichment fixed at 4.8wt %.

No. of BPRs per FA	Normal fuel ^{235}U enrichment in wt %		
	6 wt % gadolinia	8 wt % gadolinia	10 wt % gadolinia
0	4.8	4.8	4.8
4	4.829	4.828	4.827
8	4.857	4.856	4.855
12	4.886	4.884	4.882
16	4.914	4.911	4.909
20	4.942	4.939	4.936
24	4.970	4.966	4.962

Table 1. The variation of 'normal' fuel enrichment to maintain a FA average ^{235}U enrichment of 4.8 wt% as the number of BPRs and gadolinia content in the poisoned pellets changes.

We have computed the response for the collar positioned at the mid-height of the FAs, that is in the absorber zone of the BPRs. The absorber zone (320 cm) is effectively infinite long from the perspective of the measurement and so details of the axial loading pattern need not be modeled. This would not be the case if a complete axial scan were to be performed.

EXISTING DATA TREATMENT

The LANL software, INCC [7], used by the IAEA to perform LWR fuel collar assays implements the active assay method described in [8]. Example data and software test results for which may be found in [9]. The calibration curve assumes the following Padé relationship:

$$KD = \frac{am}{1 + bm}$$

or, upon rearranging

$$m = \frac{1}{a/(KD) - b} = \frac{(KD)/a}{1 - b\,(KD)/a}$$

where

D = is the deadtime, $\frac{R+A}{A} - 1$ bias, and passive background corrected Doubles rate. In practice because the rates are low and similar for calibration and assay measurements not dead time correction is applied. Also, because a predelay is used in the shift-register circuit to eliminate electronic transients and because the rates are low no bias correction is needed;

$m = \frac{m_{235}}{L_{fuel}}$ = is the linear density of ^{235}U in the fuel assembly expressed as g.cm^{-1}, m_{235} being the mass of ^{235}U in the assembly and L_{fuel} the active length of the fuel present;

$a\ and\ b$ = are calibration constants; and,

$K = k_0k_1k_2k_3k_4k_5$ = is the overall correction factor and comprises the product of the six individual correction factors listed below.

k_0 = AmLi source yield factor and accounts for the relative strength between the source installed in the collar and the strength of the source (which must be of nominally identical in construction and placement) used during calibration. When the same source is being used the correction reduces to a simple decay correction, $k_0 = 2^{t/t_{1/2}}$, where t is the time elapsed between the reference date of the calibration and the time of the assay, and $t_{1/2}$ is the radioactive half-life of ^{241}Am, the α-emitter in the source. When a different source is being used then k_0 incorporates an additional factor $r_Y = Y/Y_{cal}$ the ratio of the neutron emission rate of the AmLi source installed and the source used for calibration determined at an arbitrary time.

k_1 = the efficiency or electronic drift factor can account for (slight) changes in the high voltage settings and temperature effects, for example. The value may be estimated from $k_1 = \left(\frac{S_{expected}}{S_{measured}}\right)^2$ where $S_{expected}$ is the expected dead time, background and decay corrected Singles rate with the AmLi only loaded in the collar and $S_{measured}$ is the corresponding measured rate at the time of the assay. Unless there has been a change to the operating conditions k_1 is normally set to unity unless there is compelling evidence to the contrary.

k_2 = corrects for relative detector responses under the detector cross calibration program. The correction factor k_2 is unity if the calibration coefficients entered into the software already include the detector cross calibration factor. Otherwise $k_2 = \frac{D_{ref\_collar}}{D_{applied\_collar}}$, where D_{ref_collar} is the corrected Doubles rate for the reference collar on the date of cross calibration with the reference fuel assembly and AmLi source, and $D_{applied\_collar}$ is the same thing but for the collar being used.

k_3 = poison correction factor
Fuel assemblies with pins (rods) containing burnable poisons will give a lower Doubles rate than an unpoisoned assembly with the same ^{235}U linear density. The form of the multiplicative poison correction factor, k_3, which notionally contains four calibration constants is:

$$k_3 = 1 + \frac{n_{poison} N_{ref}}{N_{assay}} A_c [1 - \exp(-\lambda_a G)](B_c - C_c E_{enr})$$

where, for a given fuel assembly design measured in a particular uranium neutron collar design:
n_{poison} = the number of poison rods in the fuel assembly being assayed;
N_{ref} = the total number of rods present in the reference fuel assembly used for calibration;
N_{assay} = the total number of rods in the fuel assembly being assayed;
E_{enr} = the ^{235}U enrichment of the fuel assembly being assayed in wt%;
G = the concentration of poison in the poison rods, for example wt% of gadolinium or wt% of boron;
λ_a = the poison absorption factor which depends on the type of poison rod and for a given poison material is determined by calibration measurements or simulation;
$A_c, B_c \; and \; C_c$ = are additional calibration constants determined along with λ_a by measurement or simulation.

The expression for k_3 may be simplified if the ratio N_{ref}/N_{assay} is by design and control unity, and we note that B_c can be set to unity without loss of generality reducing the number of free calibration parameters to three.

k_4 = uranium mass or heavy metal loading correction factor
As the heavy metal linear density of the assembly increases the observed Doubles rate per unit of fissile linear density increases as a result of additional neutron reflection. A correction factor is applied to Doubles rate so that the reference case calibration can be applied. The uranium mass correction factor k_4 has the form:

$$k_4 = 1 + D_c \left(E_c - \frac{m_U}{L_{fuel}} \right) = 1 + D_c (E_c - u)$$

where
m_U = total mass of uranium (heavy metal) in the fuel assembly expressed in grams and which is normally approximated as the sum of ^{235}U and ^{238}U masses;
L_{fuel} = the active length of the fuel, cm;
$\frac{m_U}{L_{fuel}} = u$ = the linear density of uranium in the assembly expressed as g.cm^{-1};
$D_c \; and \; E_c$ = are calibration constants specific to the fuel assembly design and measurement set-up.

k_5 = miscellaneous effects correction factor, itself the product of terms each describing a perturbing sample effect such as the presence of protective plastic bagging and cardboard around the fuel assembly.

Correction factors k_3, k_4, and k_5 are amenable to evaluation by simulation. In this work the factor of greatest influence is k_3. The essential point is that the current analytical correction factor makes use of a linear dependence on the number of BPRs present with coefficients that are design specific. There is no explicit allowance for how the BPRs are positioned within the FA. To simplify the discussion we note that in our simulations we have fixed the average enrichment averaged over the entire FA based on a fixed carrier enrichment and a three axial zone loading pattern, but E_{enr} is essentially fixed. For the a given gadolinia concentration the factor $[1 - \exp(-\lambda_a G)]$ is constant and in the limiting case will saturate out at unity. Our reference case and assay case have the same number of pin. Thus to first order the analytical correction factor model has the form $1/k_3 = 1/(1 + a \cdot n_{poison})$, where a

is a constant for a given poison concentration, and we give the reciprocal to describe the roll off in the response rather than the correction.

MCNPX SIMULATIONS

We used the same basic approach to collar modeling as recently described elsewhere for the case of BWR fuel [1,2]. We again used the transport code MCNPX 2.7 with coincidence tallies [5], ENDF-B-VII cross section library and the S(α, β) treatment for the High Density Polyethylene Density (HDPE). The collar geometry is different however being of the UNCC-II design for PWR FAs. A cross sectional view is provided in Fig.2. The key aspects of the MCNPX description are: 0.95 g/cm^3 HDPE of the body of the collar detector, 20 ^{3}He-filled proportional counters (Reuter-Stokes RS-P4-0813-101 type), with nominal active length of (330.30±0.76) mm, filled to a partial pressure of 4-atm in ^{3}He (see also [5] for a detail description). It worth pointing out that we included in the MCNPX input deck the description of the room to allow in the simulations neutron scattering.

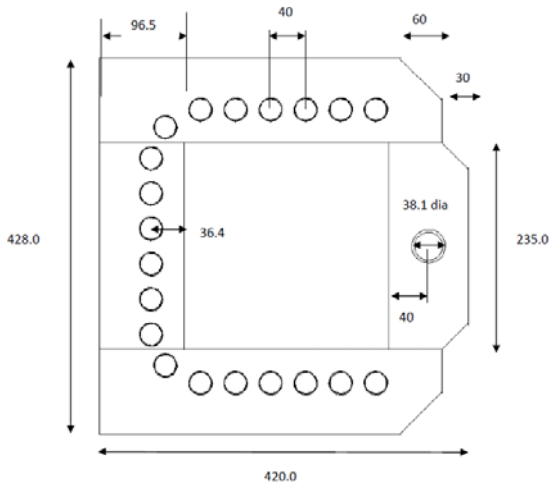

Fig. 2 Schematic of the LANL PWR Collar moderator assembly. All dimensions are in mm.

The description of the AmLi source is based on our best current understanding of source MRC-AmLi-117 manufactured by Monsanto Research Corp., Dayton, Ohio, USA and shipped to LANL in March 1983. It is a modified description to that provided previously [1]. The nominal ^{241}Am activity when shipped was 0.93 Ci corresponding to 0.271 grams of ^{241}Am or 0.307 grams of ^{241}AmO2 powder. The mass of Li target is given as 2.05 grams corresponding to 2.35 gram of LiH. The LiH to AmO_2 mass ratio in the source is therefore about 7.65.
The active source material is contained in a source capsule of model type 2724-BT which sits inside a tungsten shield to reduce the external gamma-ray dose-rate. The capsule is double walled 304 stainless steel. The external dimensions are 2.54 cm diameter by ~3.49 cm long including the weld seam. The combined base thickness is ~0.89 cm (including what appears to be a 0.13 cm thick disc which appears to be pressed into the inner container to hold the source material below the cap that is welded in place) and the combined top thickness ~1.14 cm while the combined wall thickness is 0.232 cm. The source sits 'upside down' (threaded lid down) in the tungsten shield and this defines the position of the pellet. The tungsten shield is ~5.73 cm long by 3.17 cm in diameter with an internal cavity 2.68 cm in dia. by 4.31 cm long. Expressed differently the base is 0.80 cm thick, the lid 1.28 cm thick and wall 0.248 cm thick. The cavity is lined with thin tape for protection. When the lid of the tungsten shield is screwed into place the neutron source capsule is held in place by compressing a piece of what looks like folded Carborundum paper. The internal capsule dimensions, taken from the engineering drawing provided, are 1.42 cm (0.5605") high by 1.03 cm (0.406") radius making the pellet volume 4.76 cc and the pellet density therefore estimated to be ≈0.558 g/cc.
The emergent neutron output of source MRC-AmLi-117 is known relative to that of MRC-AmLi-95, being a factor of 1.2197 times greater with an estimated 1-σ uncertainty of 0.13%. Our current best estimate of the output of MRC-AmLi-95 based on series of carefully conducted measurements using different detector and analyses

3.359×10^4 n/s referred to 22 April 1999 with a fractional uncertainty of about 1.1%. Assuming the source is stable apart from predictable radioactive decay of the ^{241}Am content (half-life 432.2±0.7 yr) the decay corrected rate of MRC-AmLi-117, our model source, is 4.011×10^4 n/s on 15 July 2012 with an estimated relative uncertainty of 1.1% at the 68% confidence level. In the present work however we report only relative behavior. We launched AmLi neutrons from inside the source material according to the spectrum recently unfolded and corrected for encapsulation by Hamid Tagziria [6].

MCNPX RESULTS

From our results shown in Fig. 3 it would appear that there is only a modest dependence on the gadolinia concentration over the range investigated. To put this another way, for a given number and configuration of BPRs increasing the weight fraction from 6, to 8 to 10 wt% decreases the response only marginally, which implies at these concentrations the BPRs are, at least to first order, almost equally black to the spectrum of interrogating neutrons for this thermal neutron collar design. The dependence on the number of BPRs is strong and somewhat linear. However, superimposed on this gross behavior there is a residual, relatively strong, dependence on the exact configuration of BPRs within the FA. For a given number of BPRs the reduction response seems to be greater for configurations in which the BPRs are closer to the edge suggestive of greater depression of the interrogating neutron flux.

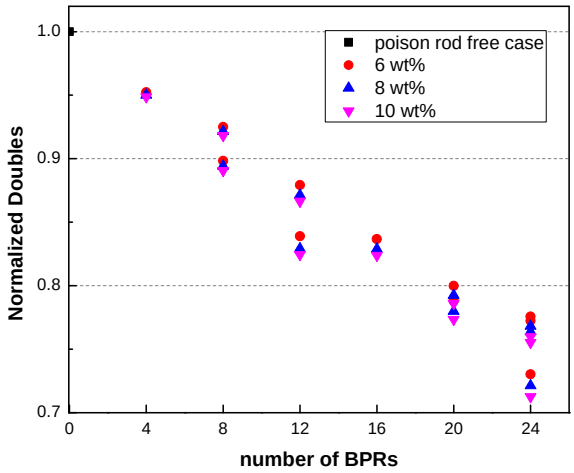

Fig. 3. A plot showing the net active Doubles count rate normalized to the poison rod free case as a function of the number of BPRs present in the FA.

CONCLUDING THOUGHTS

Reactor fuel performance has improved steadily since the 1970s as operating experience has grown and materials understanding and corrosion management have been refined. Substantially higher burnups are now achieved in PWRs using LEU fuel with higher initial enrichment and greater burnable poison concentration. Active collar correction factors developed in the 1980's do not span the modern range. This work reports on results from a bilateral project, undertaken with between the US and Brazil, for the purposes of building computational capability specifically in nuclear safeguards support of the Brazilian Angra reactor fleet. This project aims at strengthening and improving international safeguards by the IAEA and local state agencies by understanding and being able to correct collar measurements over a wide parameter range.

Fuel designers and core designers have a variety of options available. Also the state of the practice is dynamic. It seems to us that the practice of estimating active collar correction factors using empirical analytical relationships

established some 30 years ago, although simple and fast to implement at the point of use, is no longer necessary nor justified given the observed configuration dependence in this work. We have demonstrated to our satisfaction that a transport code such as MCNPX, once a model has been created, provides power fuel assembly specific simulation tool capable of represented the declared specification in detail. Thus, variations in normal fuel enrichment, as well as in the number, location, and type of burnable poison rods can be taken into account in detail on a case by case basis. This is especially important as a reactor is brought on-line with an initial charge of fuel and the characteristics of the reload fuel is adjusted over the early cycles to achieve equilibrium cycle conditions, or when some other change is made, for example to extend burnup as and when higher rated fuel becomes available and is approved for us. Additionally we have seen that general enrichment and the specification of burnable poison rods often go hand in hand and a simulation code approach can accommodate this explicitly. It is also apparent that in order to correctly interpret an axial scan a simulation will be needed to capture the response variation across the axial loading zones as well as the end effects. Experimentally this would be time consuming and expensive to generate for all the cases of interest (especially for an inspection agency with a broad portfolio). There are several possible approaches. Specific responses might be pre-calculated based on information provided prior to an inspection. A wide range of cases could be generated and stored in a data base for selection by the inspector depending on which fuel assemblies are selected for measurement. A reach back capability could be employed so that the inspectors could get real time help from experts back at home base. Or, a simple way to enter the fuel assembly information at the time of the measurement could be scripted allowing MCNPX or a similar simulation code to be launched as needed. This would allow protective packaging and other factors (correction factor k_5) to be included too.

ACKNOWLEDGEMENTS

We are grateful to Prof. Andrew Worrall, UK NNL, Springfields, for sharing his experience and wisdom on real life fuel and core design. For the purposes of studying the performance of the collar over an exaggerated parameter our crude configuration selection criteria, to what is a highly developed scientific art, retains the important nondestructive assay physics.
This work described in this paper was funded by the U.S. DOE under Action Sheet 14 between The National Nuclear Energy Commission of Brazil (CNEN) and the United States Department of Energy (DOE). We take his opportunity to thank Dr. Michael C. Browne of LANL for encouragement and advice throughout on this project.

REFERENCES

[1] S Croft, A Favalli, MT Swinhoe and Carlos D Rael, State of the art Monte Carlo modeling of active collar measurements and comparison with experiment, Proc. 52nd Annual INMM Meeting, July 2011 Palm Desert, CA, USA.

[2] A Favalli, S Croft and MT Swinhoe, Perturbation and burnable poison rod corrections for BWR uranium neutron collar, Proc. 33rd ESARDA ANNUAL MEETING, Helia Conference Hotel, Budapest, Hungary, 16-20 May, 2011.

[3] M Looman, R Jaime, P Peerani, P Shchillebeeckx, S Jung, P Schwalbach, MT Swinhoe, PJ Chare and WF Kloeckner, The effect of gadolinium poison rods on the active neutron measurement of LEU fuel assemblies, ESARDA P_2001_Bruges_17_12.pdf pp568-573
.

[4] Y Ronen (Ed), CRC Handbook of Nuclear Reactors Calculations. Vol. III. CRC Press, Inc., Boca Raton, Florida, USA (1986). ISBN 0-8493-2927-2(v.3).

[5] DB Pelowitz et al., MCNPX User's Manual, Version 2.7.B Extensions, Los Alamos National Laboratory Report, LA-UR-09-04 150 (2009); http://mcnpx.lanl.gov/ (last access June.6, 2012)

[6] H Tagziria, The ideal neutron energy spectrum of ^{241}AmLi(α,n)^{10}B sources, Applied Radiation and Isotopes. In Press.

[7] Bill Harker and Merlyn Krick, INCC Software Users Manual, Los Alamos National Laboratory report LA-UR-99-1291. July 21, 1998.

[8] HO Menlove, JE Stewart, SZ Qiao, TR Wenz and GPD Verrecchia, Neutron Collar Calibration and Evaluation for Assay of LWR Fuel Assemblies Containing Burnable Neutron Absorbers, Los Alamos National Laboratory report LA-11965-MS (1990). Issued Nov. 1990. Also known as ISPO-323.

[9] MS Krick, HO Menlove and JE Stewart, AWCC and UNCL algorithms for the IAEA, Los Alamos National Laboratory report LA-UR-91-2061 (June 10, 1991). Also known as US Program for Technical Assistance to IAEA Safeguards POTAS report ISPO-326.