Strål säkerhets myndigheten
Swedish Radiation Safety Authority

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Research

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Research within technical safeguard at Chalmers University of Technology during 2016-2017

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SSM perspective

Bakgrund

Kompetens inom området nukleär icke-spridning är en förutsättning för att ett land ska kunna leva upp till de internationella krav som följer av fredlig användning av kärnenergi. Det är därför viktigt att det finns långsiktiga forskningsprogram på flera universitet för att upprätthålla kompetensnivån i landet. Eftersom det finns begränsade satsningar från andra aktörer, såsom Vetenskapsrådet, bidrar SSM till finansieringen av forskning vid bl.a. Chalmers tekniska högskola inom kärnämneskontroll. SSM bidrar både i form av en bredare satsning på en forskningsgrupp, men även med mer riktade satsningar.

Kärnämneskontrollen är till sin natur ett internationellt område och SSM sätter stor vikt vid att de forskningsgrupper vi stöder aktivt deltar för att bidra till utvecklingen av internationella krav och riktlinjer, likväl som i det internationella vetenskapssamhället. Gruppen vid Chalmers tekniska högskola har lång tradition av att arbeta i nära samarbete med andra forskningsorganisationer och utländska universitet.

Resultat

Gruppen vid Chalmers tekniska högskola för subatomär fysik och plasmafysik har forskat på metoder för att verifiera och karakterisera använt kärnbränsle och strålkällor. De två huvudprojekten handlar om att ta fram en ny metod för korrelation- och koincidensmätningar från signaler av fissionskammare och det andra om en metod för att identifiera saknade bränslestavar i en bränslepatron med hjälp av ett kluster av fiberdetektorer. Forskningen har resulterat i ett flertal artiklar publicerade i vetenskapliga tidskrifter. Gruppen vid Chalmers har även deltagit i flera internationella konferenser och möten och presenterat sina forskningsresultat.

Relevans

För SSM finns dubbla syften med satsningen på forskning inom området för nukleär icke-spridning vid Chalmers tekniska högskola. Forskningen i sig bygger upp långsiktig nationell kompetens både vid universitetet och hos SSM:s egen personal. Satsningen bidrar också till utveckling av metoder för verifieringen och karaktärisering av kärnbränsle, avfall och strålkällor vilket är väsentligt för att uppfylla våra internationella åtaganden.

Behov av vidare forskning

Inom projektet för korrelation- och koincidensmätningar ser Chalmers att det finns behov av ytterligare simulering och teoretisk analys, och sedan laboratoriemätningar. Inom forskningen kring möjligheterna att identifiera saknade eller utbytta bränslestavar behövs bland annat utveckling av den teoretiska beskrivningen samt utveckling av mjukvaran och hårdvaran. Likande forskning pågår även på andra universitet så möjlighet finns för framtida samarbeten.

Projektinformation
Kontaktperson SSM: Lars Hildingsson
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Sammanfattning

Avdelningen för Subatomär fysik och plasmafysik i Chalmers, och dess föregångare Nukleär teknik, har under längre tid bedrivit forskning inom nukleär icke-spridning (safeguards). Målet med forskningen är att utarbeta beröringsfria kärntekniska metoder för upptäckt, identifiering och kvantifiering av speciella radioaktiva material, både strålkällor och utbränt bränsle. Efter att föregående projekt avslutades 30 april 2016, har Chalmers erhållit medel från SSM för att utföra forskning under tiden 2016-06-30 - 2017-12-31. Föreliggande rapport redovisar verksamheten som genomförts under denna period.

Forskningen bedrevs inom två huvudområden, men innehöll även andra bidrag. En kort sammanfattning av dessa följer nedan.

1) Utveckling av en ny metod för korrelation- och koincidensmätningar från signaler av fissionskammare i Campbell-mod

Bestämning av aktiviteten av ett okänt prov görs traditionellt med användning av så kallad multiplicitetsräkning. Metoden är baserad på utnyttjande av informationen från de tre första statistiska momenten av detektering av diskreta detektorpulser, dvs pulsraten av detektering av enkla, dubbla och tredubbla koincindenser (S, D och T rates) av neutroner. Dessa används för att bestämma tre okända parameterar av provet, främst den mest intressanta, fissionsraten, som är proportionell med massan. Vår forskning inom projektet handlar om en helt ny, alternativ metod, som går ut på att bestämma samma parametrar genom att använda de första tre momenten av den kontinuerliga strömmen av en eller flera fissionskammare. Metoden är fri från det så kallade dödtidsproblemet som den traditionella pulsräkningsmetoden är belastad med, och använder inte He-3 detektorer, som är en bristvara.

Vi har först härlett teorin med antagandet att alla neutroner från en spontan fission detekteras samtidigt i en eller flera detektorer. Under föreliggande period har vi utvidgat teorin för att kunna ta hänsyn till att neutroner från samma fission detekteras i olika tidpunkter p.g.a. deras olika ursprungliga hastigheter (snabbneutrondetektering) eller p.g.a. fluktuationer i tiden för deras nedbromsning (detektering av termiska neutroner). Vi har utarbetat konkreta formler för detektering av snabba neutroner, där fluktuationer av detekteringstiderna är jämförbara med bredden av detektorpulsen. Detta genom att bestämma tidsfördelning av detekteringarna, utgående från hastighetsfördelningen av fissionsneutronerna med ett givet energispektrum. Nedbromsning till termisk energi, å andra sidan, innebär avsevärt större skillnader i detekteringstiderna än pulsbredden, varför dessa bör hanteras med korrelationsmetoder, vilket kommer att ske i projektets fortsättning.

Formlerna för detektering av snabba neutroner, och därmed bestämning av den fissila massan med mätningar från fissionskammare, bekräftades genom numeriska simuleringar. Resultaten presenterades vid konferenserna Mathematics and Computation (april 2017, Jeju, Korea, och INMM-58 i Indian Wells, juli 2017; se kap. 3.). Metoden presenterades också vid ett NDA User Group möte vid INMM-58, och väckte stort intresse. Gruppens ordförande, Susan Smith, bestämde att försöka göra pilotmätningar vid Oak Ridge National Laboratory. En tidskriftspublikation är också på gång.

Detta projekt har bedrivits av en dubbeldegree doktorand från Tekniska högskolan i Budapest (BME), Lajos Nagy, med undertecknad som handledare i Chalmers och Máté Szie-

berth vid hemmainstitutet BME. Prof. L. Pál har också bidragit till arbetet. Lajos tillbringade 6 månader på Chalmers mellan 13 juni – 12 december 2016, och har börjat sin andra 6-månaders vistelse på Chalmers den 16 juni 2017. Han blev antagen som forskarstuderande vid hemmainstitutet BME 2016-09-01, och hans antagning vid Chalmers, vilket är på gång, kommer också att räknas från detta datum. Lajos kommer att disputera september 2020, därför planerar vi att fortsätta med projektet mellan 2018 – 2020. Förutom de två konferens-proceedings som nämnts ovan, har två tidskriftsartiklar publicerats inom projektet (publikationerna 1 och 2 i kap. 2), och en tredje artikel är på gång (publikation 4 i kap. 2).

2) Pilotundersökning av en metod för att identifiera saknade bränslepinnar i en bränslepatron med ett kluster av fiberdetektorer

Vi har tidigare använt olika typer av fiberdetektorer för neutrondetektering i trånga utrymmen. Det långsiktiga målet med detta projekt är att kunna genomföra mätningar på använt kärnbränslen i syfte att hitta saknade bränslepinnar. Principen är att från de deklarerade data kan man beräkna fördelning av flödestätheten i patronen med antagandet att patronen är hel. Genom att göra mätningar i flera radiella positioner och jämföra de med den förväntade fördelningen kan man i princip upptäcka saknade/ersatta bränslepinnar och bestämma deras position. Själva principen inte är ny, sådana mätningar har gjorts med traditionella detektorer i PWR-bränsle, där det finns ledrör för styrstavspinnar med tillräckligt utrymme.

Vårt projekt innebär en betydande vidareutveckling av metoden. Dels, den är baserad på användning av fiberdetektorer vilka också skulle kunna användas i BWR- bränsle. Själva diagnosen, dvs upptäckt och bestämning av position av saknade pinnar kan starkt förbättras genom användning av neurala nätverk-metoden samt genom mätning av neutronström (istället för den skalära neutronflödestätheten). Experimentell verifiering kan ske vid CLAB i Oskarshamn.

På grund av de begränsade resurserna gjordes bara en pilotstudie för att se möjligheten och fördelarna med att använda neutronströmmen. Detta bedrevs som ett examensarbete vid BME av masterstudenten Ádám Aranyosi, med Máté Szieberth som handledare. Monte-Carlo beräkningar gjordes av både det skalära flödet och neutronströmmen, med antagandet av både en intakt bränslepatron, och för det hypotetiska fallet när en bränslepinne var borttagen. En jämförelse mellan dessa två beräkningar visade att både det skalära flödet och neutronströmmen är potentiellt lämpliga för att detektera och identifiera saknade pinnar, men också att neutronströmmen är en mer känslig indikator av en saknad pinne.

För att kunna vidareutveckla metoden i sin helhet, som bl.a. innefattar utveckling av fiberbaserade detektorer för mätning av neutronström, krävs större resurser och en längre tid, t.ex. i form av ett doktorandprojekt. Förberedelser görs för att starta ett gemensamt finansierat doktorandprojekt i samarbete med SCK•CEN, som har erfarenhet inom området. Ett mer omfattande projektförslag kommer att skickas till SSM senare i september detta år.

3) Hantering av cross-talk i mätningar vid scintillationsdetektorer

Vi har också deltagit i ett projekt för att ta hänsyn till ”cross-talk”, dvs dubbla eller flerfaldig detektering av en och samma neutron, i utvärderingen av mätningar av snabba neutroner med en array av organiska scintillationsdetektorer. Samma master-ekvation baserad formalism kan användas som vid vanlig multiplicitetsräkning, men med förekommande modifieringar. Arbetet genomfördes vid Universitetet i Michigan av en doktorand, Tony

Shin, och undertecknad tjänstgjorde som handledare vid arbetets teoretiska del. En tidskriftsartikel har redan publicerats i Nuclear Science and Engineering (publikation 3 i kapitel 2).

4) Besök av gästforskare

Yasunori Kitamura från Kyoto University Research Reactor Institute (KURRI) har vistats hos oss mellan början av december 2016 till slutet av maj 2017. Dr. Kitamura har varit gästforskare hos oss två gånger tidigare, och har bl.a. hjälpt till att starta upp vår verksamhet inom safeguards. Merparten av hans nuvarande vistelse har finansierats av hemmainstitutet KURRI. Dr. Kitamura har genomfört ett mycket omfattande arbete angående statistik av strömmen i fissionskammare för att kunna genomföra Feynman-alfa och Rossi-alfa mätningar med dessa. Hans arbete med tidskorrelationer (Rossi-alfa metoden) kommer att kunna utnyttjas också i fortsättningen i Lajos Nagy:s doktorandprojekt, d.v.s. multiplicitetsmätningar med fissionskammare för termiska neutroner.

Vår dubbeldegree doktorand Lajos Nagy har bidragit till arbetet genom Monte-Carlo simulering i syfte att verifiera de analytiska resultaten. Dr. Kitamura har skrivit rapporter som utkast till tre tidskriftsartiklar. Han kommer att besöka oss för en andra 6-månaders period, från 1 december 2017.

I denna rapport ges också en redovisning av publikationer som härrör från projektet (kap. 2), samt konferensdeltaganden samt de proceedings som publicerades (kap. 3). Dessa finns längre fram i rapporten. Vi planerar också att fortsätta med projekten 1) och 2) som nämnts ovan, och ett förslag finns vid slutet av rapporten.

Summary

The Division of Subatomic and Plasma Physics in Chalmers, formerly the Division of Nuclear Engineering, has performed research in nuclear safeguards and non-proliferation since 2004, with support from SSM and its predecessor, SKI. Due to various organizational and personnel changes in Chalmers, this research was pursued with limited resources and personnel during 2016-06-30 – 2017-12-31 with a research grant, Dnr SSM2016-662 from SSM, with undersigned (I.P.) as the project leader. The research was concentrated on two main subjects:

1) Developing a method for multiplicity counting from the continuous current signals of fission chambers;
2) Feasibility study of a method of detecting the absence of one or more missing or replaced fuel pins in a spent fuel assembly, from neutron measurements with the help of a cluster of fibre detectors.

The research in both subjects was performed in collaboration with the Institute of Nuclear Techniques, Budapest University of Technology and Economics (BME), Hungary. In addition, some work was done on the treatment of “cross-talk” in multiplicity measurements with scintillation detectors for fast neutrons. This latter was performed in cooperation with the DNNG group of the University of Michigan, where the majority of the work was made. Undersigned project leader also attended an evaluation meeting of a NNSA-supported safeguards consortium in Ann Arbor on 16 February 2017.

Between 5 December 2016 and 31 May 2017 we hosted a visitor from Japan, Yasunori Kitamura, with whom we have also collaborated in the past through his visits. During his recent stay he was working on the statistics of fission chamber currents for both reactor diagnostics and safeguards problems. This research has strong connections with item 1) above, and will come to a good use in the continuation.

This report gives an account of

- The work performed in the above subjects
- List of publications
- Conference and workshop participations, session organisations
- National and international collaborations, visitors

Finally, plans for the continuation of the research are given.

1. Research topics

1.1. Developing a method for multiplicity counting from the continuous current signals of fission chambers

Important parameters of an unknown sample have traditionally been determined by multiplicity counting methods. These consist of detecting single, double and triple pulses within certain detection gates, and using the so-called singles (S), doubles (D) and triples (T) rates. These are given in the Böhnel model of superfission (which assumes that all neutrons born in a spontaneous fission leave the sample simultaneously, even if they underwent internal multiplication) as:

$$S = F\varepsilon\mathbf{M}\nu_{sf,1}(1+\alpha) \tag{1}$$

$$D = \frac{F\varepsilon^2 f_d}{2}\mathbf{M^2}\left[\nu_{sf,2} + \left(\frac{\mathbf{M}-1}{\nu_{i1}-1}\right)\nu_{sf,1}(1+\alpha)\nu_{i2}\right] \tag{2}$$

and

$$\begin{aligned} T = \frac{F\varepsilon^3 f_t}{6}\mathbf{M^3}\Bigg[\nu_{sf,3} &+ \left(\frac{\mathbf{M}-1}{\nu_{i1}-1}\right)^2\left[3\nu_{sf,2}\nu_{i2} + \nu_{sf,1}(1+\alpha)\nu_{i3}\right] \\ &+ 3\left(\frac{\mathbf{M}-1}{\nu_{i1}-1}\right)^2 \nu_{sf,1}(1+\alpha)\nu_{i2}^2\Bigg] \end{aligned} \tag{3}$$

where the three unknown parameters are the sample fission rate F, the leakage multiplication $\mathbf{M}$, and the so-called α rate (the unknown fraction of neutrons born in (α, n) reaction, out of all source neutrons). The detector efficiency ε is assumed to be known. The so-called doubles and triples gate factors f_d and f_t, both smaller than unity, express the fact that neutrons from the same fission event are not detected simultaneously, rather they are detected within a certain detection gate, hence only a fraction of the true double and triple coincidences can be registered. The other symbols stand for the (known) factorial moments of the number of neutrons generated in spontaneous and induced fissions, respectively. The above formulae do not account for the fact that due to dead time effects, some of the counts may be lost at high count rates. There exist no simple general formulae which would express how the higher order expressions (2) and (3) need to be modified to account for the dead time effects.

Our proposal was to see whether the information in the higher order moments of the continuous current signals of fission chambers, which consist of the aggregate of the finite width pulses, can be used for extracting the same information as the traditional pulse counting methods. A fission chamber can detect not only overlapping pulses from two detections close to each other in time, but even two or more neutrons detected exactly simultaneously. This way it does not suffer from the dead time effect. Use of fission chamber could also alleviate the shortage of He-3 detectors. (It is for the same reason that multiplicity counting systems with organic scintillators, detecting fast neutrons, are increasingly being used). An illustration of the structure of the signal of a fission chamber with simple Poisson detection events is shown in Fig. 1.

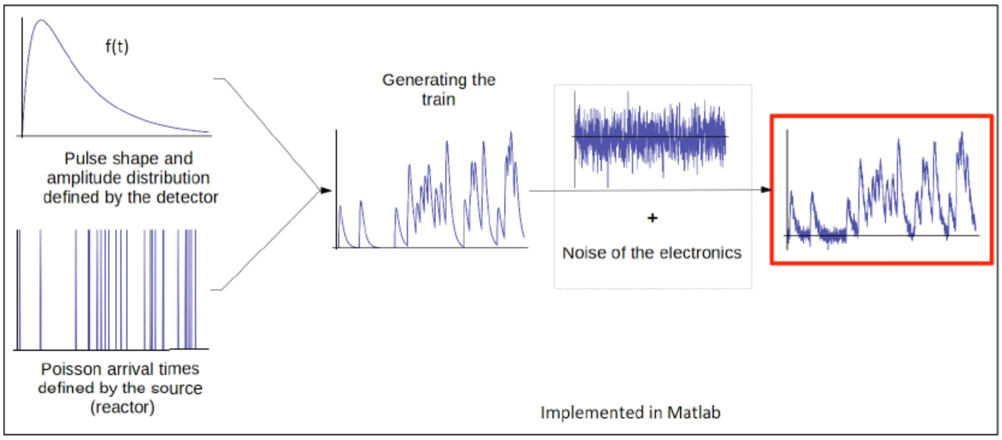

Figure 1. A sample of the fission chamber signal pulse shape $f(t)$ and an illustration of the formation of the detector current as an overlapping pulse train, for simple Poisson detection events.

A powerful formalism for the stochastic theory of fission chamber signals was developed by us recently in the frame of another, recent PhD project (Ref. [1]). In that project, only the efficient determination of the mean neutron flux was aimed at, hence the detection process was assumed to have simple Poisson statistics, each count inducing a detector pulse with a constant shape $f(t)$ and with a random amplitude **a**. In order to extract the multiplicity rates from the moments of the fission chamber current, the methodology had to be extended to account for detection processes with compound Poisson statistics, where the number distribution of the compound Poisson distribution is given by the multiplicities of the Böhnel model. This latter assumes that all neutrons born in the same fission leave the sample simultaneously, i.e. the internal multiplication takes place instantaneously, and also that the detection takes place immediately when the neutrons leave the sample. These assumptions correspond to those of the traditional pulse counting method, but assuming the doubles and triples gate factors in Eqs (2) - (3) being equal to unity.

Generalisation of the method to this case was made during 2016. It was also realised that together with the further theoretical work, which was necessary for describing the more realistic case of not simultaneous detection of neutrons from the same source event, and the verifications and sensitivity analysis of the method in numerical (Monte-Carlo) simulations, the work to be performed is suitable for a whole PhD project. A double degree PhD student, Lajos Nagy was assigned to the project. Lajos acquired an M.Sc. in nuclear engineering at the Institute of Nuclear Techniques of BME, Budapest. He was enrolled as a PhD student at BME on 1 September 2016. Enrolment in Chalmers requires a complicated paperwork and agreement between BME and Chalmers, which is expected to be concluded soon. His PhD time in Chalmers will also count from 1 September 2016. He will take a PhD both in Chalmers and BME, in late summer 2020. He will spend about half of the time at BME and about half in Chalmers. He already spent 6 months from June to December last year, and took all the obligatory PhD courses. With this, and with the number of published and submitted papers, he is soon eligible for a licentiate exam. He is currently spending his second half year in Chalmers.

Lajos Nagy joined in at the end of the first stage of the work, which concerned the accounting for the source multiplicity, but simultaneous detection of all source neutrons from one source event. He contributed to the verification of the results with Monte Carlo simulations. The extension was successful, and despite the complicated formalism, it led to surprisingly simple expressions for the first three central moments of the current of one detector, or the cross-moments of two and three different detectors, respectively. It turned out that these can all be related to the *S*, *T* and *D* rates through factors that only depend on the (known) characteristics of the detector (its pulse shape $f(t)$ and the moments $\langle \alpha^n \rangle, n = 1,2,3$, of its random amplitude). The following simple relationships were found between the first three central moments (mean, variance and skewness) of the current *y* of one detector and the *S*, *T* and *D* rates:

$$\langle y \rangle = \mathbf{S} \langle a \rangle \cdot I_1 \tag{4}$$

$$\sigma_y^2 = (\mathbf{S} \langle a^2 \rangle + 2\mathbf{D} \langle a \rangle^2) \cdot I_2 \tag{5}$$

and

$$sk\{y\} = (\mathbf{S} \langle a^3 \rangle + 6\mathbf{D} \langle a \rangle^2 \langle a \rangle + 6\mathbf{T} \langle a^3 \rangle) \cdot I_3 \tag{6}$$

where the I_n are defined as

$$I_n \equiv \int_0^\infty f^n(t) dt \tag{7}$$

These latter are known from calibration. In (4) - (6), the *S*, *T* and *D* rates are the same as in (1) - (3), but with $f_d = f_t = 1$. Similar relationships can be derived for the covariance and third order cross-moments between two and three separate detectors. These are not given here for brevity, but are found in the journal publication No.2 in Section 2.
This means that in this hypothetical case (exact simultaneous detection of all neutrons from one fission), and in knowledge of the detector efficiency ε the same information can be extracted from the fission chamber currents as from the traditional multiplicity counting. These results were presented at the INMM conference (Proc. 1, in Section 3 below) and as a journal article in NIM A (Publication No. 2 in Section 2).

However, it is clear that even if one detects fast neutrons, i.e. they do not need to be slowed down as in an Active Well Coincidence Counter (AWCC), rather detected directly from the source/sample, the neutrons from a single source event will not be detected simultaneously. This is because of the different neutron velocities, corresponding to the non-monoenergetic spectrum of neutrons, will lead to different flight times for the different neutrons. Even if the time difference due to different neutron velocities is rather small, even this small difference in the detection times will make a large difference. If the detections are made with a time difference comparable with the pulse width of the detector (the time duration of the pulse shape $f(t)$, see Fig. 1.), the formulae above will not be valid. The detector pulse shape width is in the range of a few tens of nanoseconds, i.e. rather narrow. Since neutrons in spontaneous fission are born with a relatively wide energy spectrum, the travelling time for the neutrons with different energies will deviate in a comparable order of magnitude.

The next step was thus to account for the fact that the detection of neutrons from the same fission does not take place simultaneously, but with a random probability density $u(\tau)$ of time delay τ. It turned out that such a case can easily be accommodated into the formalism, and still closed form formulae can be derived for the relationships between the moments of the detector currents, and the *S*, *T* and *D* rates, with the difference to the previous case that new factors, corresponding to the doubles and triples singles rates, appear in the formulae. These can be written in the form

$$\langle y \rangle = \mathbf{S}\, \langle a \rangle \cdot I_1 \tag{8}$$

$$\sigma_y^2 = (\mathbf{S}\, \langle a^2 \rangle + \xi_1 2\mathbf{D}\, \langle a \rangle^2) \cdot I_2 \tag{9}$$

and

$$sk\{y\} = (\mathbf{S}\, \langle a^3 \rangle + \xi_2 6\mathbf{D}\, \langle a \rangle^2 \langle a \rangle + 6\xi_3 \mathbf{T}\, \langle a^3 \rangle) \cdot I_3 \tag{10}$$

where the ξ_i are defined as

$$\xi_1 = \frac{\int_0^\infty \left[\int_0^\infty f(t-\tau)u(\tau)\mathrm{d}\tau\right]^2 \mathrm{d}t}{\int_0^\infty f^2(t)\mathrm{d}t} \tag{11}$$

$$\xi_2 = \frac{\int_0^\infty \left[\int_0^\infty f(t-\tau)u(\tau)\mathrm{d}\tau \ \int_0^\infty f^2(t-\tau)u(\tau)\mathrm{d}\tau\right] \mathrm{d}t}{\int_0^\infty f^3(t)\mathrm{d}t} \tag{12}$$

and

$$\xi_3 = \frac{\int_0^\infty \left[\int_0^\infty f(t-\tau)u(\tau)\mathrm{d}\tau\right]^3 \mathrm{d}t}{\int_0^\infty f^3(t)\mathrm{d}t} \tag{13}$$

These have the same significance and role as the gate factors in traditional multiplicity counting. With simple considerations, it can for instance to be shown that, to a good approximation, one has

$$\xi_1 \approx \frac{\Delta_f}{\Delta_u} < 1 \tag{14}$$

and

$$\xi_2 \approx \xi_3 \approx \xi_1^2 \ll 1 \tag{15}$$

where Δ_f is the width of the detector pulse $f(t)$, and Δ_u is the width of the time delay distribution function.

From the above it follows that for fast neutron detection, in practical circumstances (a few tens of cms distance between the sample and the detector, and neutron velocity differences corresponding to the width of the source energy spectra), these factors are smaller than unity, but not negligible. Actually we have calculated these by deriving the time delay distribution $u(t)$ for such cases, assuming a Watt energy spectrum of the spontaneous fission source neutrons, and evaluated them quantitatively. These results were presented at the Mathematics and Computation, as well as at the INMM-58 conferences, (Proc. 2 and 3 in Section 3 below) and were submitted for publication to NIM A (publication 4 in Section 2).

In the case of detecting thermalized neutrons, the width of the time delay distribution will be several orders of magnitude larger than the detector pulse shape. As is seen from equations (8) - (10) and (14) - (15), for such a case, the factors ξ_i will become negligible, and only the singles rates remain in expressions (8) - (10). This shows that from the one-point (simultaneous) distributions, one cannot determine the sample parameters. Two- and three-time distributions (temporal correlations between the detector signals) have to be calculated. This is planned in future work, together with the experimental verification. It is at this point where the work of our visiting scientist, Yasunori Kitamura, will come to good use (see Subsection 1.4).

1.2. Feasibility study of a method of detecting the absence of one or more missing or replaced fuel pins

A typical task of nuclear safeguards measurements is to localize missing fuel pins in fresh or irradiated fuel assemblies which were replaced by "dummy" fuel pins with the intention to illegally use the diverted fissile material. Such measurements should be performed in the spent fuel pool. There exist non-intrusive methods for this purpose, such as measurement of the Cherenkov radiation, but there is both room and need for alternative methods.

We proposed a method based on the measurement of the neutron flux at several radial positions inside a fuel assembly, and from the comparison with the neutron flux which can be calculated from the declared (intact) assembly, draw conclusions on the existence of missing/replaced pin or pins. The principle is not new, and it was already used in pilot measurements (Refs. [2] and [3]). However, there is substantial room for further developments, which is the basis for the proposed project. In the above measurements, miniature fission chambers were used, which only fit into the control rod guide tubes of PWR assemblies. Moreover, (as is standard in any reactor and even safeguards applications), only the scalar flux (the angularly integrated flux) was measured. Finally, although calculations of the neutron flux were made by intact and defect fuel assemblies (i.e. those with missing pins) to see the sensitivity of the neutron field to missing/replaced pins), no algorithmic method was proposed to declare the existence of missing fuel pin(s), and to identify their position(s) from a comparison of the measurement with the calculated fluxes.

Our proposal is to use thin fibre detectors, developed among others by Japanese researchers ([4] - [6]), which are composed of a scintillation (ZnS(Ag)) tip mixed with neutron converter material (typically LiOH or LiF) on the top of a light guiding fibre. Such detectors are small enough to be inserted in between the fuel pins. Such detectors are relatively cheap and easy to produce, and one can use a cluster of them to explore the neutron field produced inside the fuel assembly either due to some external source in case of fresh assemblies or due to the spontaneous fission in the irradiated assemblies. Based on the comparison of the calculated or calibrated effect of a replaced pin on the neutron field and the measured values, a "dummy" rod can be localized.

A primary question is the sensitivity of the method for the missing/replaced fuel pin. We have already suggested in previous work (Refs [7], [8]) that the neutron current vector, or even the flux gradient, which is also a vector quantity, is more sensitive for localised perturbations, due to the directional information contained in them. Hence, in addition to measuring the scalar (angularly integrated) neutron flux, we propose also the use of the first angular moment of the flux, the neutron current vector (roughly equivalent with the spatial gradient of the neutron flux). Scintillation detectors with light guiding fibre are suitable for such measurements. We have some previous experience, both theoretical and experimental, in using the neutron current in localisation problems ([7], [8]). In the later stages of the project, detector development for this purpose is planned.

Preliminary investigations have been started in collaboration with BME via Monte Carlo neutron transport simulations to assess the sensitivity of the neutron flux and current for a missing pin. These investigations were performed as the master thesis work of a student at BME (Ádám Aranyosi), and the project was jointly defined by Chalmers and BME. The SERPENT Monte Carlo transport code [9] was used for the investigations, in which a WWER-1200 fuel assembly has been modelled [10]. This type of fuel assembly has a hexagonal lattice. The base enrichment is 3.6% and 12 pins contain 4.2% enriched uranium. The main parameters are given in Table 1, while the cross-section of the assembly is shown in Fig. 2. The spontaneous fission neutron source was assumed to be evenly distributed in the fuel pins[1]. Detailed flux and current distributions were calculated for the reference geometry and for cases where fuel pellets in one fuel pin were replaced with steel ("dummy" fuel pin).

Fig. 3. shows the flux, while Fig. 4 the current distribution in the reference geometry. As it is expected, the current is much lower in the middle of the assembly and points outwards as approaching the boundaries. In Fig. 5 the relative change in the flux distribution can be seen after the replacement of an inner 3.6% enriched fuel pin with a dummy pin. As it can be observed, a small flux depression is shaped in the vicinity of the missing pin. The maximum change is ~2% which is significant compared to the statistical error. In Fig. 6. The relative change of the scalar current compared to the maximum current value in the geometry[2] can be seen. It reaches about 4% as a maximum, and appears to effect somewhat larger vicinity of the missing pin. Furthermore, the change in the direction of the current clearly directs towards the missing pin as it can be seen in Fig. 7. This is in line with the expectations as a fissile material is replaced with an absorber.

[1] The specification of more accurate source term definition based on detailed burnup calculations is in progress.
[2] Since the original current is close to zero in the middle the relative change in the given position was not practical for representation.

Table 1: Geometrical parameters of the WWER-1200 fuel assembly [10]

Assembly parameters	
Number of pins	312
Active length	375
Assembly pitch	23.6 cm
Pin pitch	1.275 cm
Fuel pin parameters	
Central pin parameters	0.12 cm
Pellet diameter	0.76 cm
Cladding inner diameter	0.773 cm
Cladding outer diameter	0.91 cm

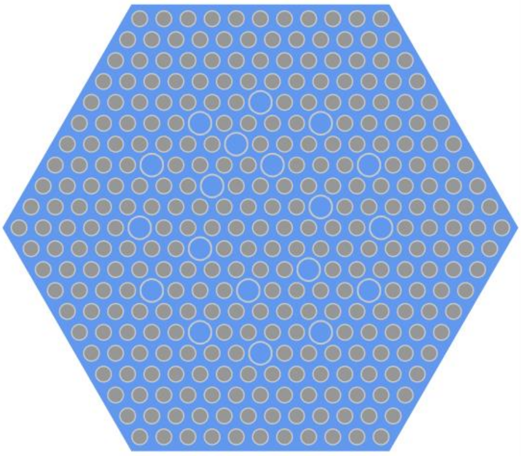

Figure 2: Cross-section of the WWER-1200 fuel assembly with the fuel pins and the 18 empty control rod positions.

As a conclusion, the preliminary calculations show that the missing pin produces a detectable perturbation in the flux and current field in its close vicinity, which may be measurable by fibre detectors inserted between the pins. The change in the current field appears to be more significant and if the change in the direction is determined by a comparison with measurements, it clearly identifies a missing pin around the measurement position. The preliminary results are encouraging for the continuation of the research, but the feasibility of the method relies on the measurement technology.

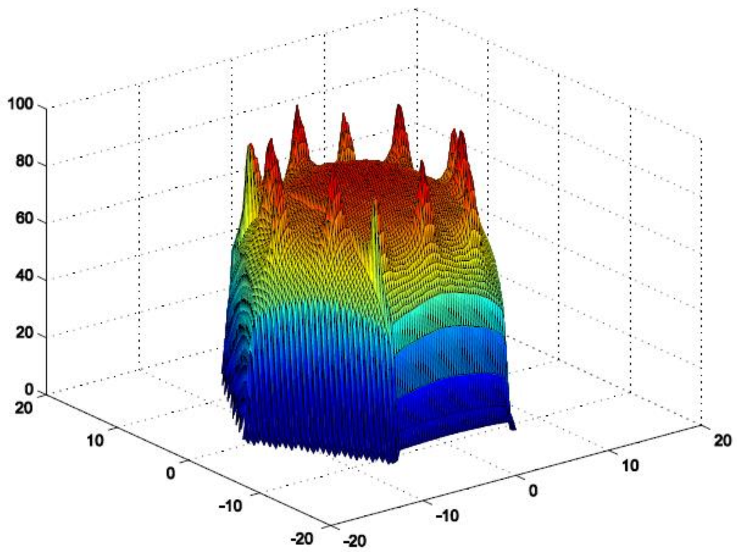

Figure 3. Flux distribution in the assembly. 4.2 % enriched pins produce local maxima.

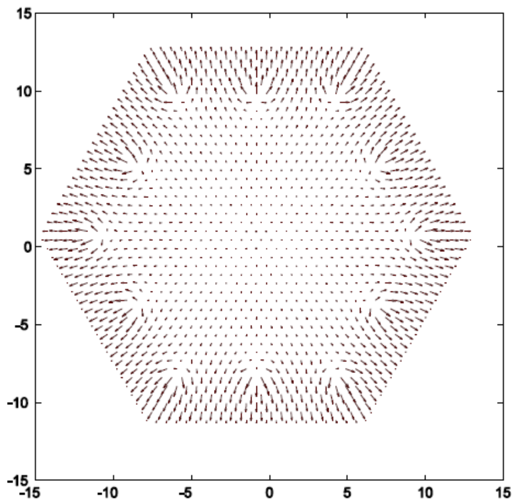

Figure 4. Current field in the assembly

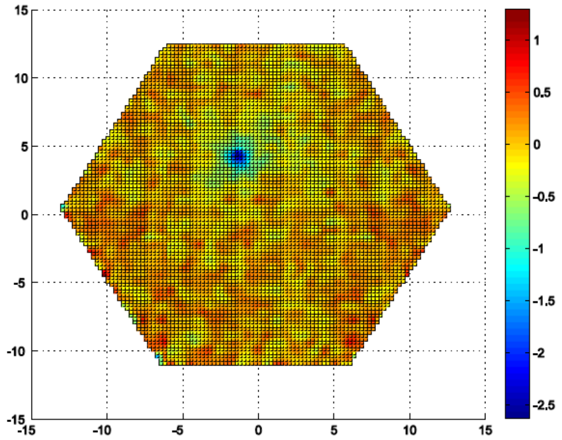

Figure 5. Relative change in the flux (%)

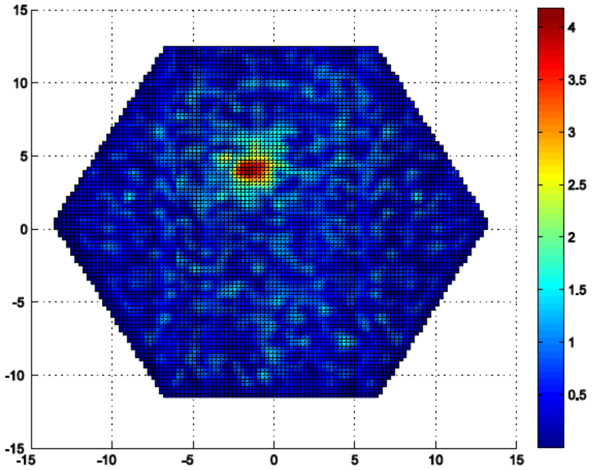

Figure 6. Change in the scalar current relative to the maximum (%)

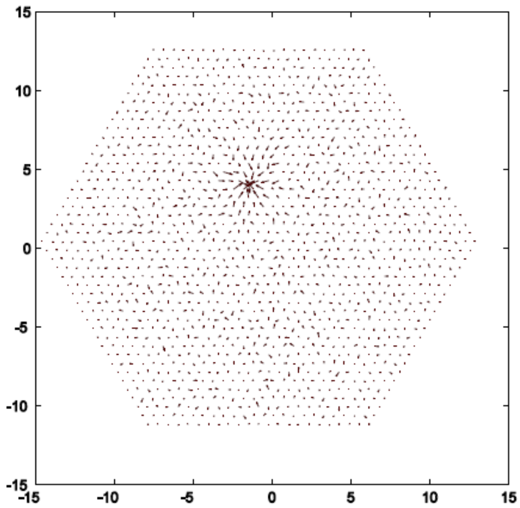

Figure 7. Change in the current vector field

1.3. Calculation of the cross-talk between scintillation detectors

The background of this work is the increased interest in using fast organic scintillators for multiplicity counting ([11] - [14]). The motivation for this latter is partly the attempt to replace the He-3 proportional counters with alternative detectors, due to the He-3 shortage, and partly the faster counting process and hence a shorter measurement time for the same accuracy.

Similarly to the AWCC systems using He-3 counters, such fast scintillation detector systems consist of a large array of detectors, to increase detector efficiency, and to decrease the dead time effect by connecting each detector to a separate counting electronics. However, in contrast to the "capture-gated" He-3 counters, these scintillation detectors are "scatter-gated", i.e. detection is made through the energy transferred to the detector in a scattering effect. Since the neutron is not absorbed in the detector, it can cause registration counts in more than one detector if it is scattered consecutively to one or more neighbouring detectors ("cross-talk"). Neutrons counted twice or more are not accounted for in the traditional formulae for the multiplicity rates, hence the application of the traditional formulae for the evaluation of such measurements will lead to an overestimation of the fissile mass of the item.

The cross-talk effect can be handled with a formalism similar to the Böhnel equations, by introducing the probability that a neutron, having been scattered in one detector, will lead to a count in one more detector. The possibility of multiple detections can be accounted for in the same recursive way as with the treatment of the internal multiplication in the Böhnel mode.

A master equation based formalism was applied to the treatment of the cross-talk effect, and formulae were derived for the multiplicities as functions of the cross-scattering probability. The results were verified in Monte-Carlo simulations and also with measurements, leading to a more accurate assessment of the fissile mass than with the traditional formulae which do not account for the cross-talk effect.
This work was performed as part of the project of a PhD student in Michigan, Tony Shin, with undersigned as advisor of the theoretical work. The results were published recently as a journal paper in Nuclear Science and Engineering (publication 3, Section 2).

1.4. Work performed by the guest researcher Yasunori Kitamura

Yasunori Kitamura, from Kyoto University Research Reactor Institute (KURRI) visited us between 5 December 2016 – 31 May 2017. His visit was mostly supported by his home institute (KURRI), by letting him using his salary while in Sweden. Dr. Kitamura has thus not received any salary or fellowship from us. However, we covered his air ticket and accommodation during his stay, from the grant we received from SSM.

Dr. Kitamura is an internationally acknowledged researcher in the field of neutron fluctuations in nuclear systems, with applications in both reactor diagnostics, including reactivity measurements with the Feynman- and Rossi-alpha methods, as well as in nuclear safeguards, including dead time effects. We have long had a collaboration in these subjects. He visited us earlier on two occasions as a senior research fellow, at the time when research in safeguards was started up at our Department in 2004. Among others, it was him who prepared Chalmers contribution to the international multiplicity benchmark, organised by Los Alamos National Laboratory, in which our results were among the best.

Independently from our work with fission chambers, Dr. Kitamura has recently become interested in utilising fission chamber currents in neutron fluctuation measurements, basically to mitigate the dead time problem [15]. The objective of this work is to alleviate the problems of certain pulse counting methods, in particular the Feynman- and Rossi alpha methods for the case of high count rates, when dead time effects become significant. Although, on the first sight, this area is different from our effort of using the fission chamber currents for multiplicity counting (the former concern the statistics of counts over a time period or at two different time points, whereas the latter concerns the one-point (in time) distributions), there is a substantial overlapping regarding the methodology, since in both cases one seeks methods of extracting the statistical moments of discrete random processes which lie behind the continuous detector signals. During his stay in Chalmers, Dr. Kitamura elaborated the methodology of extracting the same information from the fission chamber signals as from the Feynman- and Rossi-alpha measurements, first without delayed neutrons, then with the inclusion of the delayed neutrons. Finally, he also elaborated the theory of triple correlations between the signals of three fission chambers. The results are summarised in publications 5-7 in Section 2 below, in form of manuscripts which will be submitted for publication to an international journal.

Dr. Kitamura's work has several connections to our on-going research. One is that our double degree PhD student, Lajos Nagy, performed a substantial numerical work to verity the theoretical formulae. This simulation software was developed as a flexible tool for simulating a wide range of experimental situations, both multiplicity counting and correlation methods. Besides, our conclusions that for slowed down (thermalized) neutrons, the multiplicity counting with fission chamber signals cannot be performed by one-point (simultaneous) measurements, rather only by double or triple correlations, the above work is directly relevant for the continuation of the multiplicity counting with fission chamber signals. The PhD project of Lajos Nagy benefits therefore from the collaboration with Dr. Kitamura in a manifold way.

Dr. Kitamura will visit us for another 6-months period from 1 December 2017 to 31 May 2018. This time his stay will be fully financed by Japanese sources, in that in addition to his salary being paid by KURRI, he obtained a grant for the extra expenses such as his flight ticket and accommodation.

Our co-operation will also include another aspect. By the initiative of Dr. Chen Dubi of the Nuclear Research Centre of the Negev, a new edition of the multiplicity counting manual of Ensslin et al is being planned with C. Dubi, Stephen Croft of ORNL, Andrea Favalli of LANL, and undersigned as Editors. My responsibility will be the part on the theory of multiplicity counting methods, in which I plan to involve Dr. Kitamura as a co-author. A book proposal to Elsevier has been submitted, and in case the proposal is accepted, work will start around the next visit of Dr. Kitamura.

2. Publications in international journals

1. Pál L. and Pázsit I. Stochastic theory of the fission chamber current generated by non-Poissonian neutrons. *Nucl. Sci. Engng* **184**, 537-550 (2016) doi:10.13182/NSE16-18

2. Pázsit I., Pál L. and Nagy L., Determining sample parameters from fission chamber signals in the current mode. *Nucl. Instr. Meth.* **A 839**, 92 - 101 (2016) doi:10.1016/j.nima.2016.08.048

3. Shin T. H., Hua M. Y., Marcath M. J., Chichester D. L., Pázsit I., Di Fulvio A., Clarke S. D. and Pozzi S. A., Multiplicity Expressions for Fissile Mass Estimation in a Fast-Neutron Detection System. *Nucl. Sci. Engng* (2017) doi: 10.1080/00295639.2017.1354591

4. Nagy L., Pázsit I., and Pál L., Multiplicity counting from fission detector signals with time delay effects. *Nucl. Instr. Meth.* **A 884**, 119 – 127 (2018)

5. Kitamura Y., Pázsit I. et al., Time and frequency domain fluctuation analyses of neutron detection currents. To be submitted to *Annals of Nuclear Energy* (2017)

6. Kitamura Y., Pázsit I. et al., Delayed neutron effect in time and frequency domain fluctuation analyses of neutron detection currents. To be submitted to *Annals of Nuclear Energy* (2017)

7. Kitamura Y., Pázsit I. et al., Absolute measurement of subcriticality by third order fluctuation analyses of neutron detection currents. To be submitted to *Annals of Nuclear Energy* (2017)

3. Conference and project evaluation participations and proceedings papers

INMM 57th Annual Meeting, Marriott Marquis, Atlanta, GA, USA, 24 – 28 July 2016

Paper presented:
Proc 1: Pázsit I. and Pál L., *Multiplicity counting from fission detector signals*. Paper a499

International Conference on Mathematics & Computational Methods Applied to Nuclear Science & Engineering, Jeju, Korea, 16-20 April, 2017

Paper presented:
Proc 2: Nagy L., Pázsit I. and Pál L., *An Extended Theory of Multiplicity Counting from Fission Chamber Signals in the Current Mode.*

Special session organization:
At this conference, together with (and to the initiative of) Prof. John Mattingly of North Carolina State University (NCSU), undersigned organised a special session SS13A: Solving Inverse Problems for Nuclear Non-proliferation Applications. There were four sessions under the topics: Sensitivity Analyses for Safeguards, Multiplicity Counting for Nuclear Material Characterization, Solving Inverse Problems for Safeguards, and Analysis of Safeguards Measurements. The sessions were co-chaired by undersigned, John Mattingly, James Peltz of NNSA, and Chen Dubi of Nuclear Research Center Negev.

The special sessions covered recent developments in parameter estimation, model calibration, sensitivity analysis, and uncertainty quantification applied to problems relevant to monitoring states' compliance with non-proliferation obligations and detecting incipient proliferation activities. A total of sixteen talks and two posters were presented, out of which 5 invited papers. The technical content included mathematical analyses, deterministic and stochastic modelling, and statistical inference methods to estimate system inputs and parameters from experimentally measured system responses. The Chalmers paper above was also presented in this session.

More information on the special session and the papers presented can be found in the conference program:
http://www.mc2017.org/program/images/MC2017ProgramBook.PDF

Imre Pázsit (left), John Mattingly (center), James Peltz (invited speaker, right)
at the M&C 2017 (from the Newsletter of NCSU)

INMM 58th Annual Meeting, Renaissance Indian Wells, Indian Wells, California, USA, 16 – 20 July 2017

Paper presented:
Proc. 3: Nagy L., Pázsit I. and Pál L., *Multiplicity counting from fission detector signals with time delay effects.* Paper a295.

NDA User Group Meeting and presentation:
An NDA User Group Meeting was organised on Sunday, 16th July 2017, before the start of the conference. It was chaired by Susan Kane Smith of Oak Ridge National Laboratory, who took over the chair of this UG from the previous Chair, Stephen Croft. I proposed to give a contribution of method of the multiplicity counting with fission detector signals, to have an in-depth discussion on the need for an alternative method, and on its potential advantages and possible pitfalls. I gave a presentation with the title: *Can we use fission chambers to replace He-3 counters? How to extract multiplicity rates from a fission chamber signal.*

In the discussion that followed, the general conclusion was that the method had definite potentials for measurements on spent fuel, where the high count rate and the ensuing dead time problem can pose significant difficulties for the traditional pulse counting methods. The main question for the new method is the efficiency of the fission chambers; the high efficiency is one definite advantage of the He-3 detectors. The Chair, Susan Smith decided to try to perform pilot measurements for the proof-of-principle of the method. ORNL has access to spent fuel pins, and they are prepared to purchase off-the-shell fission chambers for the experiment.

Project evaluation meeting

Undersigned was invited by the U.S. Department of Energy's Office of Defense Nuclear Non-proliferation R&D, to be one of reviewers of the “Advanced Safeguards Tools for Accessible Facilities” Thrust Area within the Consortium for Verification Technology (CVT). This was the third time I attended such a project evaluation meeting. The CVT has a total budget of 25 MUSD, and is coordinated by the University of Michigan, with Prof. Sara Pozzi as the Principal Investigator. The review meeting took place on 16 February 2017 in Ann Arbor, to where I had to travel from Tokyo, since I spent the whole month of February as a Visiting Scholar at the University of Tokyo. The CVT consists of twelve leading universities and nine national laboratories and will deliver new instruments and methods for nuclear non-proliferation and safeguards, as well as educate next generation safeguards scientists. Several sub-projects of the CVT were presented and discussed, and the meeting included a visit to the laboratories and experimental facilities.

4. Continued work

Continuation is planned in both projects described under subsections 1.1 and 1.2 above, i.e. multiplicity counting with fission chambers (Project 1) and partial defect identification with fibre detectors (Project 2.). In Project 1, so far the principles of the fast neutron detection were elaborated, and some investigations were performed regarding the properties of the time delay forms. Both theoretical and simulation work is needed for a thorough feasibility, as well as sensitivity analysis of the method. In numerical simulations, not only the variations in the neutron flight times, but also the variations in the emission and detection positions can also be investigated, as well as the effect of the detector efficiency, which may be a crucial factor in the applicability of the method. These can only be treated by numerical methods. Proof-of-principle measurements are also planned, both at BME and in Oak Ridge.

The next step will be the extension of the methods, which so far only treated fast neutron detection, to the treatment of thermal neutron detection, after that the fast neutrons from the source were slowed down. This requires the extension of the one-point distributions, to two- or three-point (in time) distributions, i.e. double and triple correlations. Here a substantial theoretical effort is needed. Once the corresponding formulae are derived, similarly to the fast neutron detection part, models of the time delay function need to be established, and the suitability of the formulae for the unfolding of the sample parameters need to be investigated. A similar numerical simulation effort, as well as proof-of-principle experimental program is envisaged.

Regarding Project 2, both hardware (detectors and detection methods) and software (calculations and unfolding methods) is needed. Development of fibre based detectors for measurement of the neutron current is planned. Regarding the calculations, first an overview of the methods of calculating the flux and current distribution in the fuel assembly is to be done. These calculations should take into account the real surrounding of the fuel assembly by other assemblies in the fuel pool. Since calculation of a large number of scenarios with a large number of combinations of fuel assemblies is necessary, one should investigate the need and possibility of deterministic flux calculation methods and parallel computing methods. About the detection and location of the defects, the use of non-parametric inversion methods, such as artificial neural networks, is planned. These can be trained on calculated values of the flux and the current for both intact and various defect fuel assemblies, after which the trained network can take a measured flux distribution as input, and first supply a decision whether the assembly is intact or not, and in the latter case supply the position of the missing fuel pins.

These projects are being discussed with SSM, and more detailed proposals will be submitted on these during late summer-early autumn 2017.

5. Bibliography, references

[1] Pál L., Pázsit I. and Elter Zs. Comments on the stochastic characteristics of fission chamber signals. Nucl. Instr. Meth. A 763, 44 - 52 (2014)

[2] A. Borella, G. Marzari, R. Rossa1, K. van der Meer, Monte Carlo simulations of the Partial Defect Tester for the detection of fuel pins diversion. https://chalmersuniversity.box.com/s/i6q684za8xlj27h8rqrz5bw8vutz0ron

[3] Y. Ham, P. Kerr, S. Sitaraman, R. Swan, R. Rossa, and H. Liljenfeldt, Partial Defect Verification of Spent Fuel Assemblies by PDET: Principle and Field Testing in Interim Spent Fuel Storage Facility (CLAB) in Sweden. https://chalmersuniversity.box.com/s/7dyyn7d7o472cx4vmgynh7o0ystvdcft

[4] T. Yagi, T. Misawa, C.H. Pyeon, S. Shiroya: A small high sensitivity neutron detector using a wavelength shifting fiber, Applied Radiation and Isotopes, Volume 69, Issue 1, January 2011, Pages 176-179

[5] Yamane Y., Lindén P., Karlsson J. K-H. and Pázsit I. Measurement of 14.1 MeV neutrons with a Th-scintillator optical fibre detector. Nucl. Instr. Meth. A 416, 371 – 380 (1998)

[6] Yamane Y., Uritani A., Misawa T., Karlsson J. K-H. and Pázsit I. Measurement of the thermal and fast neutron flux in a research reactor with a Li and Th loaded optical fiber detector. Nucl. Instr. Meth. A 432, 403 - 409 (1999)

[7] Lindén P., Karlsson J. K-H., Dahl B., Pázsit I. and Por G. Localisation of a neutron source using measurements and calculation of the neutron flux and its gradient. *Nucl. Instr. Meth*. **A 438**, 345 - 355 (1999) http://www.nephy.chalmers.se/staff-pages/imre/publ/gradient.pdf

[8] Avdic S., Lindén P. and Pázsit I. Measurement of the neutron current and its use for the localisation of a neutron source. *Nucl. Instr. Meth.* **A 457**, 607 - 616 (2001) http://www.nephy.chalmers.se/staff-pages/imre/publ/senada.pdf

[9] J. LEPPÄNEN, M. PUSA, et al., "The Serpent Monte Carlo code: Status, development and applications in 2013," *Ann. Nucl. Energy*, **82,** 142-150 (2015).

[10] Vasilchenko, I., Dragunov, Y., Ryzhov, S., Kobelev, S., Vyalitsyn, V., & Troyanov, V. (2008). Choosing the governing solutions for FA of AES-2006. 7 International conference on WWER fuel performance, modelling and experimental support, Bulgaria.

[11] Dolan J. L. et al., "Plutonium Measurements with a Fast- Neutron Multiplicity Counter for Nuclear Safeguards Applications," *Nucl. Instr. Meth.* **A 763**, 565 (2014)

[12] Frame K. et al., "Development of a Liquid Scintillator Neutron Multiplicity Counter (LSMC)," *Nucl. Instr. Meth.* **A 579**, 192 (2007)

[13] Chichester D. L. et al., “Statistical Estimation of the Performance of a Fast-Neutron Multiplicity System for Nuclear Material Accountancy,” *Nucl. Instr. Meth.* **A 784**, 448 (2015)

[14] Di Fulvio A. et al., “Passive Assay of Plutonium Metal Plates Using a Fast-Neutron Multiplicity Counter,” *Nucl. Instr. Meth.* **A 855**, 92 (2017)

[15] Kitamura Y., Fukushima M. Count-loss effect in subcriticality measurement by pulsed neutron source method, (II) proposal for utilization of neutron detection system operated in current mode, *J. Nucl. Sci. Technol*. **52,** 801–810 (2015)

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