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METHODOLOGY TO ASCERTAIN CORE RECRITICALITY FOLLOWING A SEVERE FISSION REACTOR ACCIDENT

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Abstract
Recriticality concerns have been expressed following severe power reactor accidents, involving fuel melting and possible core relocation out of the reactor vessel. Analyses of reactor coolant liquid samples must carefully evaluate fission isotope activities when assessing the possibility of a recriticality. Assessments should consider the criticality duration, sampling time, sampling location, and core operating history to provide an accurate determination of a recriticality. By considering these factors, the $^{131}I/^{137}Cs$ activity ratio can lead to a determination if a recriticality occurred.

Keywords
recriticality analysis, reactor coolant samples, TMI-2 accident; Fukushima Daiichi Accident

1.0 Introduction

Following the Three Mile Island Unit-2 (TMI-2) [1, 2] and Fukushima Daiichi [2, 3] accidents, concerns were raised regarding recriticality following the loss of well-defined core geometry. The United States Nuclear Regulatory Commission (NRC) also reviewed the possibility of a recriticality following a core damage event [4]. These references acknowledge the possibility of a recriticality, but do not evaluate sampling methods to verify its occurrence.

The loss of core geometry was severe at TMI-2, but the reactor vessel remained intact and recriticality was prevented using high boric acid concentrations in the reactor coolant system to maintain adequate shutdown margin ($k_{eff} \leq 0.98$) [1]. The boron concentration was maintained by requiring double valve or equivalent isolation to ensure no lower concentration boric acid or demineralized water reached the damaged core [2]. Since TMI-2 defueling activities were dynamic, these isolation barriers changed as defueling progressed and various flow paths were required to support the various defueling methods. The defueling methods changed as the various layers of core damage (e.g., loose rubble, fused fuel assemblies, melted fuel, melted fuel/steel eutectics, and melted fuel adhering to reactor vessel structures) were excavated and removed from the reactor vessel.

At the Fukushima Daiichi Nuclear Power Station (FDNPS) Units 1, 2, and 3, core geometry was not only lost, but reactor pressure vessel (RPV) and containment vessel (CV) integrity may have been compromised [5]. Fuel apparently melted through the bottom of the RPV and CV. It is believed that quantities of fuel reside on the bottom of the RPV, bottom of the CV, and possibly on the reactor building floor [5].

The loss of core geometry and loss of fission product barrier integrity enhances the possibility of core recriticality, and complicates it detection. This condition is exacerbated because the status of the control rods is uncertain, the core cooling flow paths are dynamic, fuel may have been relocated out of the reactor vessel, and changing plant conditions permit the potential for deboration of the water cooling the fuel. Each of these events could result in a subsequent recriticality [2].

A criticality would normally be detected by the neutron count rate, but the loss of well-defined core geometry with fuel relocation in a severe accident may limit the effectiveness of nuclear instrumentation [1]. This instrumentation may have been damaged during the accident or become inoperable as defueling proceeds [1]. The

unavailability of nuclear instrumentation would require an alternative method to determine if a recriticality occurred. This paper outlines an alternative approach to determine if a recriticality occurred and its effectiveness in assessing the nature of the event

2.0 General approach

A recriticality could be recognized from the production of short-lived fission isotopes (e.g., ^{131}I) if the event occurred during recovery operations after the core's fission product inventory of short-lived nuclides had decayed. In the case of TMI-2, in core instrumentation was damaged when the fuel melted [1]. As such, recriticality detection depended upon other measurements such as reactor coolant samples and temperature measurements.

Gas samples could also detect a recriticality, but these samples might not be definitive since fission gases will be trapped in undamaged fuel as well as melted fuel. Noble gases were liberated during TMI-2 defueling operations, particularly during plasma arc cutting operations [1]. In view of the potential issues with nuclear instrumentation and possibility of noble gas emissions during defueling operations, this paper examines the analysis of liquid samples and their potential to determine if a recriticality occurred.

Matsui [3] performed an analysis of Fukushima Daiichi liquid samples to ascertain the possibility of a recriticality following the accident. However, the analysis only considered radioactive decay as a fission product removal mechanism and did not examine details of the core operating history. Based on experience at TMI-2, other removal mechanisms and the specific sampling location must be also addressed to determine if a recriticality occurred.

3.0 Formalism

A criticality results in a nuclear reaction that produces neutrons and fission products. For the purpose of this paper, neutron detection is assumed to be unavailable or unreliable. This is a reasonable assumption, because the neutron count rate determination is affected by core alterations that occur during defueling operations and original nuclear instrumentation and associated systems may have been damaged during the accident or recovery activities [1]. In addition, fuel may have been relocated out of the reactor vessel, which limits the usefulness of original nuclear instrumentation.

The quantity of fission products produced in a criticality depends on a number of factors including the number of fission events, nuclide that fissions, the duration of the criticality, time the liquid sample is drawn following termination of the criticality, and the sampling location. The number of atoms of isotopes i following the fission of nuclide k created from time t_q to t_f is given at a subsequent sampling time t by the relationship:

$$N_{ik}(t) = \int_{t_q}^{t_f} f_{ik} N_o e^{-(\xi_i(t-t'))} dt' - N_i^b = f_{ik} N_o e^{-(\xi_i(t-t_f))} \frac{1 - e^{-(\xi_i(t_f - t_q))}}{\xi_i} - N_i^b \quad (1)$$

where N^b_i is the number of atoms of isotope i present before the criticality, N_o is the number of fissions per unit time during the recriticality, f_{ik} is the fraction of isotope I produced per fission of nuclide k, and ξ_i is the total removal rate of isotope i, which can be written as:

$$\xi_i = \lambda_i + \alpha_i + \beta_i + \ldots \quad (2)$$

where λ_i is the removal rate of isotope i from radioactive decay, and α_i, β_i, …represent other removal rate processes that have an exponential behavior [6, 7]. These processes are not utilized in this paper. B N^b_i is obtained from previous sample analyses performed before the suspected criticality.

The activity of isotope i is given by:

$$A_{ik}(t) = \lambda_i N_{ik}(t) \quad (3)$$

In addition to radioactive decay, other processes reduce the activity in a fluid stream. These removal mechanisms depend on the isotope produced and its chemical properties within the liquid being sampled. During accident conditions, these removal terms are time dependent (e.g., depend on the chemical composition of the liquid and associated pH). Other reduction terms from processes such as filtration and demineralization must also be considered [7, 8].

The sampling location reduction terms are analogous to the chemical efficiency factor applied to a radiopharmaceutical isotope generator. For example, ^{99m}Tc is extracted from a ^{99}Mo generator by elution and the efficiency of this chemical removal of about 95% [7]. Similar reduction terms have been observed in previous reactor

accidents, and will be illustrated by data derived from the TMI-2 accident [8].

The important consideration is that only considering radioactive decay is not always sufficient to determine the activity of isotope i following a fission event. Table 1 illustrates the removal of fission isotopes as TMI-2 reactor coolant was transferred to other plant locations [8].

Table 1 illustrates the removal of TMI-2 fission products from the reactor coolant during the 1979 accident. At TMI-2 fission products were transferred from the fuel pellet/clad in the following sequence: 1) from the pellet/clad into the reactor coolant; 2) from the reactor coolant into the reactor containment basement sump; 3) from the containment sump to the auxiliary building sump; and 4) from the auxiliary building sump to the environment [7]. At TMI-2, minimal activity of cesium and strontium were released to the environment in spite of the loss of the fuel, reactor coolant system, and containment fission product barriers. The reader should note that the containment building remained intact, but reactor coolant was transferred to the auxiliary building sump where radioactive material, primarily noble gases, were released through the waste gas system to the environment [7]. For example, only a fraction (3 /40) of the soluble Cs reached the auxiliary building from reactor building. Much of the Cs removal was due to chemical reactions, which removed the fission product from the liquid transported to the auxiliary building. Similar reactions occurred for other fission products. These results support the author's contention that reduction mechanisms other that radioactive decay affect the sample results.

The values summarized in Table 1 also illustrate the conservatism that was inherent in the reactor safety analysis utilized at the time of the TMI-2 accident [9]. In particular, the actual TMI-2 iodine release fraction was significantly overestimated by a factor much greater than 10 in the WASH-1400 analysis [9]. One of the lessons of the TMI-2 Accident is the significant overestimate of the release source term, and the need to account for inherent physical and chemical removal mechanisms. This lesson is reflected in current NRC regulations that permit a facility specific source term instead of a conservative default accident source term. These considerations must also be included in analyses of sampling data.

Table 1. Location specific reduction factors in reactor coolant samples from the TM-2 Accident

Fission product and associated form	Percentage of fuel isotope activity at the specified location			
	Reactor vessel	Reactor building	Auxiliary building	Environment
Cesium				
- liquid	50	40	3	-
- gaseous	-	<< 1	-	-
Strontium				
- liquid	2	1	-	-
Noble gas	70	70	5[a)]	5[a)]
Iodine				
- liquid	30	20	3[b)]	-[b)]
- gaseous	-	0.6	0.0001	0.00001

[a)] WASH-1400 [9] predicted a 100% value;
[b)] WASH-1400 [9] predicted a 50% value;
Derived from Knief [8].

The experience from the TMI-2 accident reflected in Table 1 suggests that the activity equation Eq. 3 must be modified to account for the sampling location (j) within the plant:

$$A_{ikj}(t) = g_{ij}\,\lambda_i\,N_{ik}(t) \qquad (4)$$

where gij represents the fraction of the theoretical activity of isotope i present at the j^{th} sampling location. This is effectively an isotope specific sampling location reduction factor (SLRF). At a given sampling location, the ratio of the activities of two isotopes (I and m) is given by:

$$\left[\frac{A_{ikj}(t)}{A_{mkj}(t)}\right] = \frac{g_{ij}\,f_{ik}\,\lambda_i\,e^{-(\xi_{ij}(t-t_f))}\,\frac{1-e^{-(\xi_{ij}(t_f-\tau_q))}}{\xi_{ij}} - N_i^b}{g_{mj}\,f_{mk}\,\lambda_m\,e^{-(\xi_{mj}(t-t_f))}\,\frac{1-e^{-(\xi_{mj}(t_f-\tau_q))}}{\xi_{mj}} - N_m^b} \qquad (5)$$

In the case of the TMI-2 accident, the g_{ij} values are provided in Table 1. The Table 1 values have a significant impact on the sampled activity since they alter the usually assumed activity ratio based on Eqs. 1 and 2 and require the utilization of the more general relationship of Eq. 5.

The problem of isotopic composition is not yet complete because Eq. 5 assumes that only a single nuclide fissions. In a commercial light water reactor, fission dominantly occurs in three separate processes. These are the thermal fission of ^{235}U and ^{239}Pu and fast fission of ^{238}U. As noted in Ref. 10-12, the yields of the various fission products depend on the specific nuclides that fission. Accordingly, Eq. 5 must be modified to account for the individual nuclides that undergo fission:

$$\left[\frac{A_{ij}(t)}{A_{mj}(t)}\right] = \frac{\sum_{k=1}^{3} a_k \, A_{ikj}(t)}{\sum_{k=1}^{3} a_k \, A_{mkj}(t)} = \frac{\sum_{k=1}^{3} a_k \, g_{ij} \, f_{ik} \, \lambda_i \, e^{-(\xi_{ij}(t-t_f))} \frac{1-e^{-(\xi_{ij}(t_f-t_q))}}{\xi_{ij}} - N^b_i}{\sum_{k=1}^{3} a_k \, g_{mj} \, f_{mk} \, \lambda_m \, e^{-(\xi_{mj}(t-t_f))} \frac{1-e^{-(\xi_{mj}(t_f-t_q))}}{\xi_{mj}} - N^b_m} \quad (6)$$

where k labels the nuclide undergoing fission, a_k is the fraction of total fissions resulting from the fission of nuclide k (k = 1 for ^{235}U, k = 2 for ^{238}U, and k = 3 for ^{239}Pu), and f_{ik} and f_{mk} are the fission yields of isotope i and m from the fission of nuclide k. In principle, the removal rates ξ_{ij} and ξ_{mj} are dependent of the sampling location j.

However, in this paper, that flexibility is not utilized and these removal rates are just the physical decay constant for isotopes i and m. The ak values for ^{235}U, ^{238}U, and ^{239}Pu depend on the reactor operating history, fuel enrichment, burnup, and initial isotopic compositions.

To complete the model specification, the ak fission fraction values are defined in terms of readily available nuclear data [10-14]:

$$a_k = \frac{N_k \, \sigma_k^{fission} \, \phi_k}{\sum_{r=1}^{3} N_r \, \sigma_r^{fission} \, \phi_r} \quad (7)$$

where N_k is the number of atoms of nuclide k per unit fuel volume that fissions [13], fission k s is the microscopic fission cross section for nuclide k [12, 13], and k f is the core thermal neutron fluence rate (flux) for ^{235}U and ^{239}Pu and the fast neutron flux rate for ^{238}U [14]. In Eq. 7, r labels the same three fissioning nuclides as defined in Eq. 6

In applying Eq. 6, specific operating conditions must be assumed. The results depend on the core operating history, but this paper can be sufficiently illustrated by assuming a specific set of core conditions. For specificity, the TMI-2 vintage core parameters [12-14] are used to derive the calculational assumptions. This paper assumes:

- ^{235}U and ^{239}Pu fissions occur predominantly by thermal neutrons and ^{238}U fissions are induced by fast neutrons [12, 13];
- Core average thermal and fast neutron fluence rates are 3×10^{13} and 5×10^{13} n/cm^2-s, respectively [14];
- In the beginning of life (BOL) core case, the fuel has the following composition: ^{235}U 3 wt% and ^{238}U 97 wt% [11];
- In the middle of life (MOL) core case, the fuel has the following composition: ^{235}U 1.9 wt%, 236U 0.2 wt%, fissile plutonium 0.33 wt%, nonfissile plutonium 0.13 wt%, fission products 1.5 wt%, and ^{238}U 96 wt% [11]; and
- In the end of life (EOL) core case, the fuel has the following composition: ^{235}U 0.8 wt%, ^{236}U 0.4 wt%, fissile plutonium 0.65 wt%, nonfissile plutonium 0.25 wt%, fission products 2.9 wt%, and ^{238}U 95 wt% [11].

4.0 Results and discussion

The model of Eq. 6 and 7 was verified by reproducing the results of Matsui [3] for the simplified case of ^{235}U thermal fission, which was independent of the sampling location and only included radioactive decay in deriving the removal term. This is demonstrated by comparing Eq. 6 to the equivalent equation of Ref. 3. These results are equivalent in the limit $a_k \rightarrow 1$, $g_{ij} \rightarrow 1$, $g_{mj} \rightarrow 1$, $N^b i \rightarrow 0$, and $N^b_m \rightarrow 0$. Since Ref. 3 only presents a semi-log graph and not specific numerical values, the calculations of this paper were compared to Ref. 3 by scaling values from the Ref. 3 graph. The resulting calculations reproduced the values and trends of the calculations published in Ref. 3. The comparison was sufficient to verify that the model was performing as desired.

In order to assess a recriticality via liquid sampling, this paper focuses on two radionuclides with abundant fission yields. ^{131}I and ^{137}Cs meet this criterion and have half-lives of 8.023 d and 30.07 y, respectively. Given these half-lives, their activity ratio decreases on a timescale on the order of days after termination of a fission reaction. Comparing the calculated ^{131}I/^{137}Cs activity ratio to sample results permits determination of when the criticality terminated. In addition, the magnitude of the ratio indicates the duration of the criticality. These are useful factors in assessing and mitigating a recriticality in a damaged light water reactor core, and their time variation is illustrated in subsequent discussion.

Eq. 6 was used to calculate the ratio of ^{131}I/^{137}Cs activities for a variety of conditions. These conditions include the criticality duration, sampling location, SLRF, sampling time relative to the termination of the criticality, and core operating characteristics. These parameters complicate the use of sampling data to ascertain a criticality event. To illustrate their importance, the TMI-2 specific sampling locations and SLRF values are utilized in subsequent discussion.

The sensitivity of the $^{131}I/^{137}Cs$ activity ratio to the criticality duration is provided in Fig. 1 for BOL conditions and sampling of the reactor vessel. Criticality durations of 1 d (top curve), 7 d, 1 mo, 3 mo, 6 mo, and 12 mo (bottom curve) were utilized. In the calculations of Fig. 1, sampling times of 1 to 100 days after the criticality duration are illustrated. These assumptions are arbitrary, but illustrate the inherent difficulties in investigating a possible criticality event based solely on liquid sample analysis.

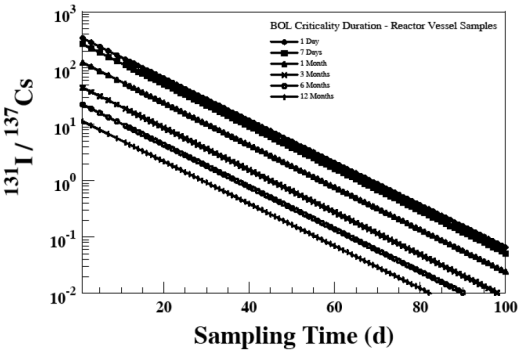

Figure 1. Calculated $^{131}I/^{137}Cs$ activity ratio for an assumed BOL criticality based on reactor vessel samples. Criticality durations of 1 d, 7 d, 1 mo, 3 mo, 6 mo, and 12 mo are assumed. The criticality durations of the calculated events from the top to the bottom curve are 1 d, 7 d, 1 mo, 3 mo, 6 mo, and 12 mo, respectively.

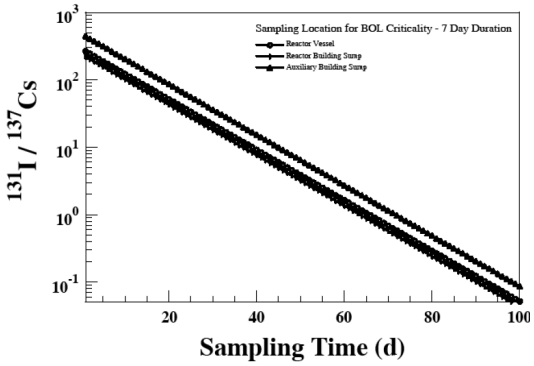

Figure 2. Calculated $^{131}I/^{137}Cs$ activity ratio for an assumed BOL criticality based on three sample locations with SLRF values derived from Table 1. A criticality duration of 7 d is assumed. The upper, middle, and lower curves are based on sampling the auxiliary building sump, reactor vessel, and reactor building sump, respectively.

The results illustrate that the criticality duration has a significant impact on the $^{131}I/^{137}Cs$ activity ratio. This ratio increases as the criticality duration decreases. A measurement of the magnitude of the $^{131}I/^{137}Cs$ ratio provides an indication of the criticality duration. The results of Fig. 1 provide a natural calibration curve that would determine the criticality duration, but specific details of the criticality would be difficult to infer from the sampling data.

Figure 2 examines the influence of the sampling location for a 7 d BOL criticality using the TMI-2 SLRF values summarized in Table 1. In particular, the influence of sampling the reactor coolant in the following locations: reactor vessel (middle curve), reactor building sump (lower curve), and auxiliary building sump (upper curve) is illustrated. The results of Fig. 2 suggest that the sampling location has a significant impact on the $^{131}I/^{137}Cs$ ratio. A proper assessment of a recriticality must consider the influence of the sampling location when comparing measurements and calculations.

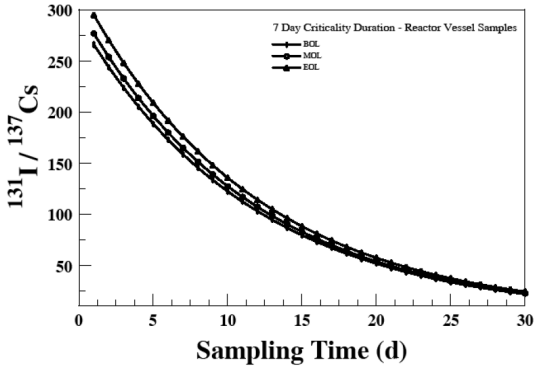

Figure 3. Calculated $^{131}I/^{137}Cs$ activity ratio for an assumed criticality based on reactor vessel samples and a criticality duration of 7 d. The upper, middle, and lower curves are based on EOL, MOL, and BOL core operating histories, respectively.

The influence of the core operating history is illustrated in Fig. 3. Three operating configurations are assumed and include the BOL (lower curve), MOL (middle curve), and EOL (upper curve) fuel compositions described previously. A 7 d criticality duration and reactor coolant sampling in the reactor vessel are assumed to permit comparison to the Fig. 1 results. The results demonstrate that the core operating history affects the calculated ratio, but the effect is not as significant as the sampling location or the duration of the criticality. In addition, the effects of core operating history become minimal after about 30 d post criticality

For completeness, other isotope ratios are reviewed. In particular, $^{131}I/^{90}Sr$ (upper curve), $^{131}I/^{137}Cs$ (middle curve), and $^{131}I/^{134}Cs$ (lower curve) isotope ratios are provided in Fig. 4 for reactor vessel samples. The ^{90}Sr and ^{134}Cs calculations provide a comparison to the previous ^{137}Cs results. The trends in Fig. 4 are under-

standable based on the physical half-lives and SLRF values noted previously. The $^{131}I/^{90}Sr$ ratio has the largest value since the Sr SLRF for the reactor vessel (Table 1) is a factor of 25 times smaller than the Cs factor. The $^{131}I/^{137}Cs$ ratio is larger than the $^{131}I/^{134}Cs$ ratio because the ^{134}Cs half-life is shorter than the ^{137}Cs half-life.

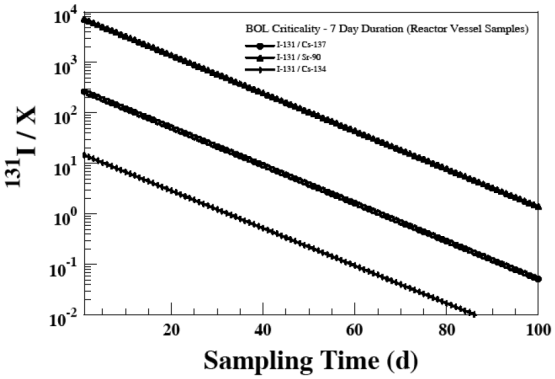

Figure 4. Calculated $^{131}I/X$ activity ratio for an assumed BOL criticality based on reactor vessel samples and a criticality duration of 7 d (X = ^{90}Sr, ^{137}Cs, and ^{134}Cs). The upper, middle, and lower curves are the $^{131}I/^{90}Sr$, $^{131}I/^{137}Cs$, and $^{131}I/^{134}Cs$ isotope ratios, respectively.

The $^{131}I/^{90}Sr$, $^{131}I/^{137}Cs$, and $^{131}I/^{134}Cs$ isotope ratios provide additional information to analyze a recriticality event. Using samples from a variety of locations and evaluating various isotopic ratios can provide insight into the nature of a recriticality following a severe reactor accident. However, the analysis must be carefully performed and consider a variety of factors to determine if a recriticality occurred.

5.0 Conclusions

Recriticality concerns have been expressed following the TMI-2 and Fukushima Daiichi accidents. These severe accidents complicate recriticality analysis because fuel melting and core relocation limits the effectiveness of nuclear instrumentation. If neutron detectors are unavailable or inoperable, a recriticality can be evaluated by considering fission product isotope activity ratios. However, the analysis must consider the sampling location, core operating history, criticality duration, and sampling time post criticality to provide the most accurate assessment of the event.

References

1. GPU Nuclear Corporation Three Mile Island Nuclear Station Unit 2 Defueling Completion Report, Revision 4. Middletown, PA: GPU Nuclear Corporation (1990).
2. Bevelacqua, J. J. Applicability of Health Physics Lessons Learned from the Three Mile Island Unit-2 Accident to the Fukushima Daiichi Accident, Journal of Environmental Radioactivity 105, 6 (2012).
3. Matsui, T. Deciphering the Measured Ratios of Iodine-131 to Cesium-137 at the Fukushima Reactors (2011), available at arXiv:1105.0242v3 [nucl-th] accessed on August 5, 2012.
4. U. S. Nuclear Regulatory Commission. Recriticality in a BWR Following a Core Damage Event. Washington DC, U. S. Government Printing Office, NUREG/CR- 5653 (1990).
5. The National Diet of Japan, The Fukushima Nuclear Accident Independent Investigation Commission, The Na-tional Diet of Japan, Tokyo (2012).
6. Bevelacqua, J. J. Contemporary Health Physics: Problems and Solutions. 2nd edition Weinheim, Wiley-VCH (2009).
7. Bevelacqua, J. J. Basic Health Physics: Problems and Solutions. 2nd edition Weinheim, Wiley-VCH (2010).
8. Knief R. A. Nuclear engineering—theory and technology of commercial nuclear power. 2nd edition. Washington, DC, Hemisphere Publishing Corporation (1992).
9. U. S. Nuclear Regulatory Commission. Reactor Safety Study: An assessment of Accident Risks in U.S. Commer-cial nuclear Power Plants. Washington DC, U. S. Government Printing Office, WASH-1400, (1975).
10. International Nuclear Data Committee Report, Handbook of Nuclear Data for Safeguards Database Extensions, August 2008, INDC(NDS)-0534, International Atomic Energy Agency, Vienna (2008).
11. Glasstone, S. Energy Deskbook, DOE/IR/05114-1. US Department of Energy, Washington, DC (1982).
12. IAEA Report, Thorium fuel cycle – Potential benefits and challenges, IAEATECDOC-1450, International Atomic Energy Agency, Vienna (2005).
13. IAEA Report, International Evaluation of Neutron Cross-Section Standards, STI/PUB/1291, International Atomic Energy Agency, Vienna (2007).
14. Olander, D. R. Fundamental Aspects of Nuclear Reactor Fuel Elements, TID-26711- P1. US Department of En-ergy, Washington, DC (1976).