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Technical Note

Effectiveness of the neutron-shield nanocomposites for a dual-purpose cask of Bushehr's Water—Water Energetic Reactor (VVER) 1000 nuclear-power-plant spent fuels



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ABSTRACT

In order to perform dry interim storage and transportation of the spent-fuel assemblies of the Bushehr Nuclear Power Plant, dual-purpose casks can be utilized. The effectiveness of different neutron-shield materials for the dual-purpose cask was analyzed through a set of calculations carried out using the Monte Carlo N-Particle (MCNP) code. The dose rate for the dual-purpose cask utilizing the recently developed materials of epoxy/clay/B₄C and epoxy/clay/B₄C/carbon fiber was less than the allowable radiation level of 2 mSv/h at any point and 0.1 mSv/h at 2 m from the external surface of the cask. By utilization of epoxy/clay/B₄C instead of an ethylene glycol/water mixture, the dose rates on the side surface of the cask due to neutron sources and consequent secondary gamma rays will be reduced by 17.5% and 10%, respectively. The overall dose rate in this case will be reduced by 11%.

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1. Introduction

The Bushehr Nuclear Power Plant (BNPP) is a Water—Water Energetic Reactor (VVER) 1000 type (model V-460) with an annual spent-fuel production of about 21 tons of heavy metals; the plant has been in full commercial operation since 2013 [1]. The spent-fuel assemblies (SFAs) will be stored for at least 3 years and usually for more than 5 years in the spent-fuel pool next to the core. The capacity of the pool for safe storage of SFAs is sufficient for about 8 years [2]. After this cooling time, the old SFAs shall be transported from the pool to provide enough capacity for new SFAs. A schema of the BNPP fuel assembly and its main characteristics is illustrated in Fig. 1 [2,3].

Conventionally, the TK-13 cask, which is a forged stainless-steel flask with an external neutron shielding of ethylene glycol mixed with water, is utilized for transporting 12 assemblies from the spent-fuel pools of VVER 1000 reactors [4]. The transportation systems using the TK-13 cask for these reactors were developed in the period of 1983–1991 [5]. A schematic view of this cask is presented in Fig. 2 [2].

Nowadays, optimization and improvement tasks are being performed in some countries to design and utilize new casks for

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the spent nuclear fuels of VVER 1000 reactors [6–9]. In view of global trends, these casks are designed as dual-purpose systems involving the storage of spent fuel, as well as on-site and off-site transportation, before and after storage [5,10]. One of the main technical aspects of a dual-purpose cask (DPC) is its shielding design, which has been considered as a research topic in different fields [11–17]. In this context, the most important modification of the newly designed casks for the VVER 1000 SFAs is the employment of solid neutron shields instead of liquid ones.

The use of liquid neutron shields may often complicate the cask design and operation. Expansion volumes and pressure relief systems must be provided to prevent overpressurization during normal and accident conditions. Additionally, the likelihood of retaining the liquid neutron shield in accident conditions is low. The loss of the entire neutron-shielding material in an accident results in a more difficult and potentially more hazardous accident-recovery procedure. In this study, the effectiveness of these solid neutron shields, especially the newly developed nanocomposite ones, for a DPC of the BNPP SFAs is investigated using Monte Carlo simulations.

2. Methodology

To evaluate radiation levels and analyze the effect of different neutron-shield materials, the Monte Carlo N-Particle (MCNP), as a

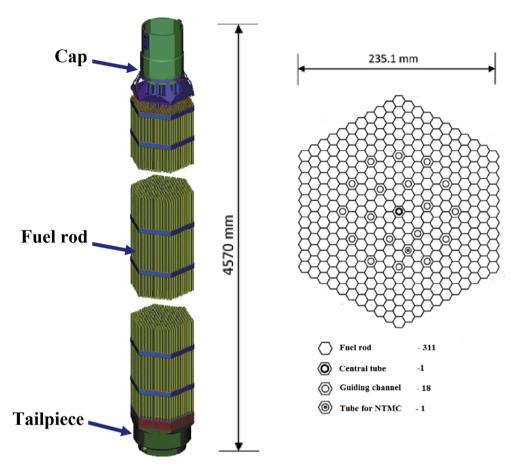


Fig. 1. Schema of the VVER 1000 fuel assemblies used in the Bushehr Nuclear Power Plant. NTMC, Neutron and Temperature Measurement Channel.

well-known and as the most widely used Monte Carlo code, was utilized [18]. A set of calculations was carried out, and, accordingly, the overall dose rates from gamma and neutron sources were determined on the cask surface and 2 m from the cask. According to the International Atomic Energy Agency (IAEA) regulations, the radiation level under routine conditions shall not exceed 2 mSv/h at any point on and 0.1 mSv/h at 2 m from the external surface of the cask [10,19]. In the case of accident conditions, the radiation level 1 m from the surface of the cask should not exceed 10 mSv/h [19].

The models of DPC and the SFAs treated in the shielding calculations are shown in Fig. 3. As shown in Fig. 3, the SFAs were modeled using three homogenized parts of fuel, cap, and tailpiece, with different radioactive sources. The radioactive-source specifications of the SFAs were determined using the ORIGEN2 code as well as data provided in the Final Safety Analysis Report (FSAR) of the BNPP [2,20].

The effects of different neutron-shield materials on the radiation levels were investigated by considering ethylene glycol mixed with water, polyethylene, and borated polyethylene as the common neutron-shield materials, as well as the recently developed neutron-shield materials of epoxy/clay/B₄C and epoxy/clay/B₄C/carbon fiber (CF) for the DPC [21]. Before the main calculations, benchmark calculations were performed by considering the assumptions of the BNPP FSAR for the TK-13 cask and the SFAs.

3. Modeling specifications and assumptions

Dose rates were calculated for primary gamma rays from fission and activation products in the spent fuel, and from activation products in the fuel-assembly cap and endpiece, and neutrons emitted by the actinides and (α, n) reactions in the spent fuel. In addition, secondary-gamma dose rates were considered in the calculation of the overall dose rates on the cask surface and 2 m from the cask.

The sources of gamma radiation from an SFA were provided in the BNPP FSAR for the most energy-critical SFA and 3-year cooling time. These gamma source specifications are presented in Table 1.

In addition to the FSAR data, ORIGEN2 calculations were carried out and gamma radiation was estimated in 18 groups of energy with a total activity of 4.25×10^{16} Bq [20]. The results were cross checked using other calculations presented in the literature [22–24]. As was expected, the neutron emission of the SFA came dominantly from the spontaneous fission of Cm²⁴⁴. Therefore, the neutron energy spectra were assumed to follow a Cm²⁴⁴ neutron distribution. The intensity of neutron sources in this study was 6.9×10^8 n/s for the burnup of 49 MWd/kgU [20].

The dose-rate calculations were made with the MCNP code. The major features of the shielding model are shown in Fig. 3. As shown in Fig. 3, the designed DPC for the BNPP SFAs [25,26]

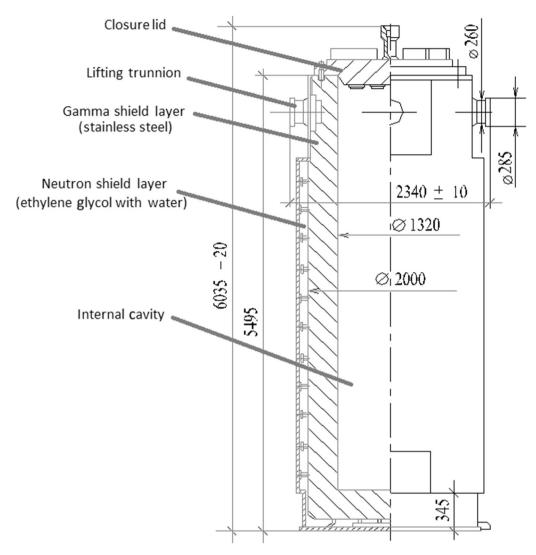


Fig. 2. Schematic view of the TK-13 cask.

consists of a cylinder body that is made of forged carbon steel with a wall thickness of 34 cm as the gamma shield. The neutronshield material, encased in stainless steel, was attached to the outer wall of the cask body. Also, the neutron-shield material was poured into the bottom plate of the cask, and a disk of neutronshield material encased in a steel shell was attached to the closure lid to provide neutron shielding at the bottom and top of the DPC, respectively. Air was assumed to be the medium surrounding the DPC.

In the MCNP model, cylindrical surfaces were placed outside the radial cask shield at different distances. These surfaces were further subdivided into segments by horizontal surfaces, and the F2 surface tally was used to obtain the averaged neutron and photon fluxes over each surface segment. These fluxes were converted to dose rates using the ANSI/ANS-6.1.1-1977 American Nuclear Society (ANS 1977) flux-to-dose-rate conversion factors. Since MCNP yields results normalized to one source particle, the FMn card is used to convert the MCNP calculated doses (mSv/particle) to the appropriate dose rates (mSv/h). For the gamma-ray doses, geometry splitting as a variance-reduction technique was employed to bias photon transport to the outside of the cask.

4. Specifications of neutron-shield materials

In recent years, the enhancement of the properties of the polymer matrix via the addition of nanoclay has received much attention, both in the academic area and in industry, because these additions often lead to an improvement in the mechanical and thermal properties of the resultant nanocomposite. Two neutronshield materials of epoxy/clay/B₄C and epoxy/clay/B₄C/CF were developed recently by the authors [21]. These nanocomposites were manufactured using an epoxy resin, SR 1500, and SD 2505 hardener filled with a commercially available nanoclay, natural montmorillonite modified with methyl, tallow, bis-2-hydroxy-ethyl material, and Cloisite 30B. The mixing ratio of resin/hardener was 100:33, and the formulation bases of the epoxy were bisphenols A and F. As shown in Fig. 4, the particle sizes are in a range of about $2-6~\mu m$. The main characteristics of the selected CF used in the material of epoxy/clay/B₄C/CF are summarized in Table 2. A scanning electron microscope image of the epoxy/clay/B4C/CF shield material is shown in Fig. 5.

To investigate the effectiveness of the recently developed materials of epoxy/clay/B₄C and epoxy/clay/B₄C/CF for the DPC,

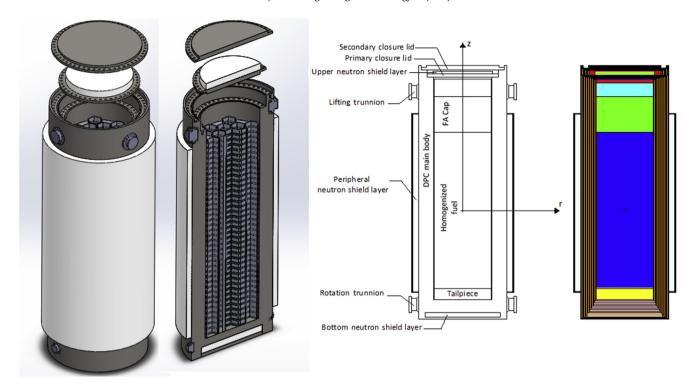


Fig. 3. Schematic view of the dual-purpose cask and the MCNP model. DPC, dual-purpose cask; FA, fuel assembly.

Table 1Intensity of gamma-radiation sources of a Bushehr Nuclear Power Plant spent-fuel assembly after 3-year cooling.

	•
Effective energy (MeV)	Volumetric intensity $(\gamma/cm^3/s)$
0.13	1.20E+09
0.18	1.80E+07
0.24	3.70E+07
0.3	2.50E+06
0.5	1.20E+09
0.64	1.20E+10
0.8	3.90E+09
1.23	5.70E+07
1.53	5.50E+07
2.18	6.60E+07

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Fig. 4. SEM image of epoxy/clay/ B_4C neutron-shield material. SEM, scanning electron microscope.

Table 2Main characteristics of the carbon fiber used in the neutron-shield material.

Main characteristic	Value
Tensile strength (MPa) Modulus (GPa) Elongation (%) Density (g/cm³) Thickness (mm)	3,530 230 1.70 1.76 0.23

polyethylene and borated polyethylene are also considered in this study. These materials are the most widely used ones for neutron shielding in commercial casks. The specifications of the solid neutron-shield materials considered in this study are presented in Table 3. The material-composition data of polyethylene and borated

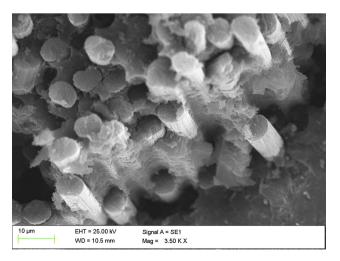


Fig. 5. SEM image of epoxy/clay/B₄C/carbon fiber neutron-shield material. SEM, scanning electron microscope; EHT, Extra High Tension Voltage; WD, Working Distance; Mag, Magnification.

 Table 3

 Specification of various neutron-shield materials considered in this study.

Material	Density (g/cm³)	Chemical composition (wt %)
Polyethylene Borated polyethylene Epoxy/clay/B ₄ C	0.93 1.00 1.4	H (14.4), C (85.6) H (12.5), C (77.5), B (10.0) C (75.5), H (7.1), O (10.1), Si (2.1), Al (1.2), Mg (0.09), B (3.9)
Epoxy/clay/B ₄ C/carbon fiber	1.315	C (85.3), H (4.3), O (6.0), Si (1.3), Al (0.7), Mg (0.05), B (2.3)

Table 4Maximum dose rates of the TK-13 cask.

	Bushehr Nuclear Power Plant Final Safety Analysis Report (mSv/hr)	This study (mSv/hr)
Trunnion surface	0.7	0.67
Cask side surface	0.2	0.29
One meter from side surface	0.2	0.22

polyethylene are based on the Pacific Northwest National Laboratory data [27].

5. Results and discussion

The dose rates on the cask side surface, trunnion surface, and 1 m from the cask were determined by considering the assumptions of the BNPP FSAR for the TK-13 cask and the BNPP spent fuels. The results of this calculation are compared with the BNPP FSAR data in Table 4. According to the BNPP FSAR and other references [4], the neutron-shield material for the conventional TK-13 cask is ethylene glycol (67%) mixed with water. The differences between the results and the BNPP FSAR data shown in Table 4 arise from modeling assumptions, which are not clearly determined in the BNPP FSAR, as well as the utilized code and cross-sectional data.

For the DPC with the gamma shield of carbon steel, the replacement of the liquid neutron shield (ethylene glycol mixed

with water) by the solid neutron-shield material of polyethylene was investigated. Based on the performed calculations, the utilization of polyethylene will reduce the dose rates from neutron sources by 45%, decreasing the overall dose rates by 10%. The dose rates from the neutron sources and the consequent secondary gamma rays on the DPC side surface for a neutron-shield material of ethylene glycol mixed with water and polyethylene are shown in Figs. 6 and 7, respectively.

The main portion of the total dose rate on the side surface of the DPC is related to the secondary gamma emissions caused by neutron absorption in the shielding materials. To reduce the dose rates of the secondary gamma, borated materials can be used. The utilization of borated polyethylene for the DPC, in comparison with using ethylene glycol mixed with water, will reduce the dose rates from neutron sources and the consequent secondary gamma emissions by 55% and 6.5%, respectively. As shown in Fig. 8, the dose rates from neutron sources and consequent secondary gamma rays in this case will be reduced by 16%.

For the DPC with neutron-shield materials of epoxy/clay/B₄C and epoxy/clay/B₄C/CF, the overall dose rates from the primary gamma rays, neutrons, and secondary gamma rays were determined. As illustrated in Fig. 9, the maximum dose rate was determined at a point 271 cm above the DPC center (in the *z* direction in Fig. 3), which is related to the trunnion surface. This matter is also the same in the other cases. To reduce the dose rate in this vicinity, the trunnions should be manufactured to be hollow, so as to allow pouring of the neutron-shield material into them.

In the case of the DPC with the neutron-shield material of epoxy/clay/B₄C, in comparison with ethylene glycol mixed with water, the dose rates from the neutron sources and the consequent secondary gamma will be reduced by 17.5% and 10%, respectively. As shown in Fig. 10, the dose rates from the neutron sources and consequent secondary gamma rays in this case will be reduced by 11%. In the case of the DPC with the neutron-shield material of epoxy/clay/B₄C/CF, the overall dose rate is the highest. However, the calculated dose rate in this case is also less than the allowable radiation level of 2 mSv/h at any point on the external surface of the DPC.

For an easy comparison, the dose rates on the DPC side surface and at the center (z=0 in Fig. 3) for different neutron shields are provided in Table 5.

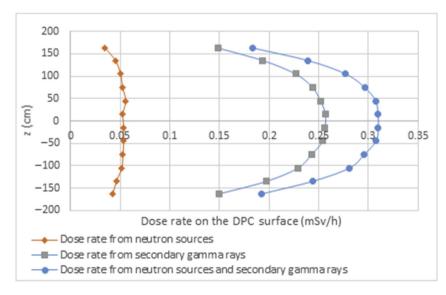


Fig. 6. Dose rates from neutron sources and consequent secondary gamma rays on the surface of the dual-purpose cask (DPC) with neutron-shield material of ethylene glycol mixed with water.

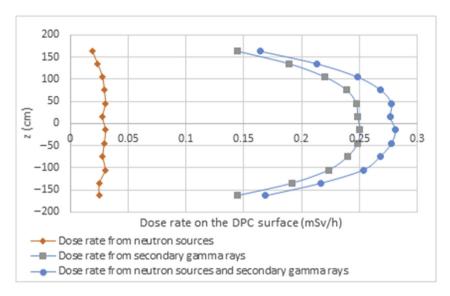


Fig. 7. Dose rates from neutron sources and consequent secondary gamma rays on the surface of the dual-purpose cask (DPC) with neutron-shield material of polyethylene.

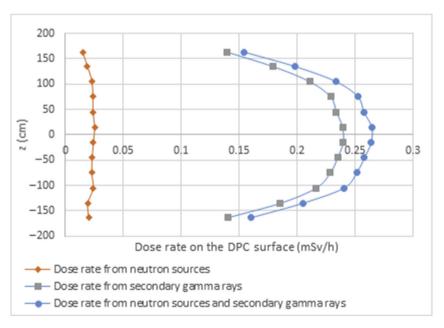


Fig. 8. Dose rates from neutron sources and consequent secondary gamma rays on the surface of the dual-purpose cask (DPC) with neutron-shield material of borated polyethylene.

6. Conclusion

The effectiveness of different neutron-shield materials for a DPC of Bushehr's VVER 1000 nuclear-power-plant spent fuels was investigated through a set of calculations carried out using the MCNP code. Neutron-shield materials of polyethylene, borated polyethylene, and the recently developed materials of epoxy/clay/B₄C and epoxy/clay/B₄C/CF were employed. Based on the performed calculations, the replacement of the liquid neutron shield (ethylene glycol mixed with water) by a solid neutron-shield material of polyethylene will reduce the overall dose rates by 10%.

The utilization of borated polyethylene for the DPC, in place of ethylene glycol mixed with water, will reduce the dose rates from neutron sources and the consequent secondary gamma by 55% and

6.5% , respectively. The overall dose rates in this case will be reduced by 16%.

The recently developed materials of epoxy/clay/B₄C and epoxy/clay/B₄C/CF can be used as the neutron shield of the DPC. The calculated dose rates for both cases are less than the allowable radiation level of 2 mSv/h at any point on the external surface of the DPC. In the case of the DPC with the neutron-shield material of epoxy/clay/B₄C, the overall dose rates will be reduced by 11% in comparison with the dose rate for ethylene glycol mixed with water.

The maximum dose rate for all cases was determined 271 cm above the DPC center, which is related to the trunnion surface. To reduce the dose rate in this vicinity, the trunnions should be manufactured to be hollow, so as to allow pouring of the neutron-shield material into them.

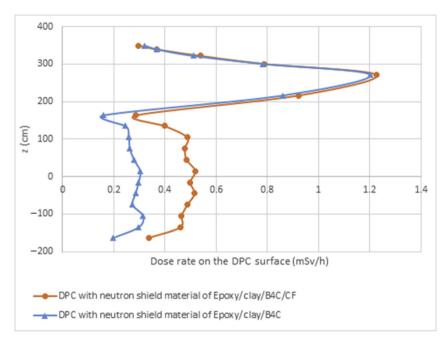
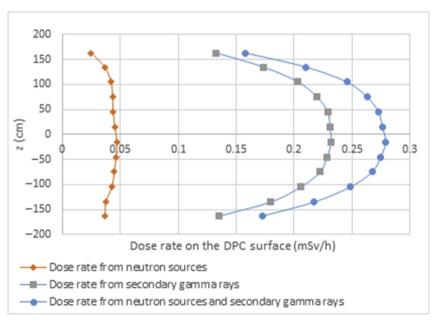


Fig. 9. Overall dose rates on the surface of the dual-purpose cask (DPC) with neutron-shield materials of epoxy/clay/B₄C and epoxy/clay/B₄C/carbon fiber.



 $\textbf{Fig. 10.} \ \ \text{Dose rates from neutron sources and consequent secondary gamma rays on the surface of the dual-purpose cask (DPC) with neutron-shield material of epoxy/clay/B_4C.}$

Table 5Dose rates on the side surface of the dual-purpose cask with different neutron shields.

Material of neutron shield	Maximum dose rates on the side surface and at $z = 0$ (mSv/hr)			
	From neutron sources	From secondary gamma rays	From neutron sources and secondary gamma rays	
Ethylene glycol mixed with water	0.053	0.257	0.310	
Polyethylene	0.029	0.250	0.279	
Borated polyethylene	0.025	0.239	0.264	
Epoxy/clay/B ₄ C	0.046	0.231	0.277	
Epoxy/clay/B ₄ C/carbon fiber	0.186	0.301	0.487	

Conflicts of interest

There is no conflict of interest.

References

- IAEA, Reference Data Series, Nuclear Power Reactors in the World, No. 2, International Atomic Energy Agency. 2015.
- [2] Design and Engineering Survey Institute, Safety Analysis Report for the Bushehr Nuclear Power Plant, Atomenergoergoproekt, Moscow, 2003.
- [3] Y. Semchenkov, Y. Styrin, Advancing of VVER Reactor Core, 2010. Bulgaria, Varna.
- [4] V.I. Koulikov, T.F. Makarchuk, N.S. Tikhonov, Implementation of burnup credit in spent fuel management systems, Proceedings of an Advisory Group meeting held in Vienna, 20–24 October, IAEA, Vienna, 1997.
- [5] IAEA TECDOC, Operation and Maintenance of Spent Fuel Storage and Transportation Casks/Containers, No. 1532, Vienna, Austria, International Atomic Energy Agency (IAEA), 2007 [CD-ROM].
- [6] T. Funke, Ch. Henig, CASTOR® 1000/19: development and design of a new transport and storage cask, in: Proceedings of the International Youth Nuclear Congress, 20 – 26 September, Interlaken, Switzerland, 2008.
- [7] S.A. Mazur, Development of the Container Fleet for SNF Transportation, OJSC Federal Centre for Nuclear and Radiation Safety, Moscow, 2014.
- [8] T. Makarchuk, Experience of cask technology for SNF management, in: International Conference on the Management of Spent Fuel in Nuclear Power Reactors, IAEA, Vienna, 2015.
- [9] A.M. Kirkin, A.V. Kuryndin, A.A. Stroganov, Implementation of guidance for an integrated transport and storage safety case for dual purpose casks for spent nuclear fuel in Russian regulatory document, in: International Conference on Management of Spent Fuel from Nuclear Power Reactors: An Integrated Approach to the Back End of the Fuel Cycle, IAEA, Vienna, 2015.
- [10] IAEA TECDOC, Guidance for Preparation of a Safety Case for a Dual Purpose Cask Containing Spent Fuel, International Atomic Energy Agency (IAEA), Vienna, 2013.
- [11] J.H. Ko, et al., Shielding analysis of dual purpose casks for spent nuclear fuel under normal storage conditions, Nucl. Eng. Technol. 46 (2014) 547–556.
- [12] B. Droste, Overview of preparation of a safety case for a dual purpose cask for storage and transport of spent fuel and recommendations to WASSC and TRANSSC from WASSC/TRANSSC Working Group, in: International Workshop on the Development and Application of a Safety Case for Dual Purpose Casks for Nuclear Spent Fuel, IAEA, Vienna, 2014.

- [13] M. Asami, K. Sawada, A. Konnai, N. Odano, Application of dose evaluation of the MCNP code for the spent fuel transport cask, Prog. Nucl. Sci. Technol. 2 (2011) 855–859.
- [14] C.J. Park, C.J. Jeong, D.K. Min, H.Y. Kang, W.S. Choi, J.C. Lee, G.S. Bang, K.S. Seo, Shielding performance of a newly designed transport cask in the advanced conditioning spent fuel pyroprocess facility, Nucl. Eng. Technol. 40 (2008) 319–326
- [15] I.J. Cho, J.N. Jang, G.S. You, J.S. Yoon, Neutron and gamma shielding estimation for the KN-12 cask, J. Nucl. Sci. Technol. 45 (2008) 54–57.
- [16] M. Asami, H. Sawamura, K. Nishimura, Monte Carlo shielding calculations for a spent fuel transport cask with automated Monte Carlo variance reduction, J. Nucl. Sci. Technol. 2 (2011) 860–865.
- [17] J.K. Shultis, Radiation Analysis of a Spent-fuel Storage Cask, Department of Mechanical and Nuclear Engineering, Kansas State University, Manhattan, USA, 2000.
- [18] Los Alamos National Laboratory, User Manual Monte Carlo N-particle Transport Code System, Los Alamos National Laboratory, New Mexico, 2000.
 [19] IAEA Safety Standards, Regulations for the Safe Transport of Radioactive
- [19] IAEA Safety Standards, Regulations for the Safe Transport of Radioactive Material, Specific Safety Requirements No. SSR-6, International Atomic Energy Agency, 2012.
- [20] M. Rezaeian, J. Kamali, Radioactive source specification of Bushehr's VVER-1000 spent fuels, Sci. Technol. Nucl. Ins. 2016 (2016) 4579738, http:// dx.doi.org/10.1155/2016/4579738.
- [21] M.A. Kiani, S.J. Ahmadi, M. Outokesh, Fabrication of "Boron—Clay—Polymer" and "Lead—Clay—Polymer" Nanocomposites for Radiation Shielding of Neutron and Gamma Rays, M.S. thesis, Sharif University of Technology, Tehran, Iran, 2014.
- [22] M.B. Emmett, Calculational Benchmark Problems for VVER-1000 Mixed Oxide Fuel Cycle, Oak Ridge National Laboratory: ORNL/TM-1999/207, USA, 2000.
- [23] I.I. Linge, E.F. Mitenkova, N.V. Novikov, End-to-end calculation of the radiation characteristics of VVER-1000 spent fuel assemblies, Phys. At. Nucl. 75 (2012) 1603—1615.
- [24] P. Petrov, Calculation of the neutron sources in the spent nuclear fuel from WWER-1000, Compt. Rend. Acad. Bulg. Sci. 61 (2008) 885–890.
- [25] M. Rezaeian, J. Kamali, Basket criticality design of a dual purpose cask for VVER 1000 spent fuel assemblies, Kerntechnik 81 (2016) 640–646.
- [26] M. Rezaeian, J. Kamali, Effect of a dual-purpose cask payload increment of spent fuel assemblies from VVER 1000 Bushehr Nuclear Power Plant on basket criticality, Appl. Radiat. Isot 119 (2017) 80–85.
- [27] R.G. Williams, C.J. Gesh, R.T. Pagh, Compendium of Material Composition Data for Radiation Transport Modeling, Pacific Northwest National Laboratory: PNNL-15870 Rev. 1, USA, 2011.