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Material attractiveness evaluation of inert matrix fuel for nuclear security and non-proliferation

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ABSTRACT

The impacts of chemical stability of inert-matrix-fuel (IMF) on the material attractiveness for states and non-state actors were evaluated in an open transuranic fuel cycle employing high temperature gas cooled reactors. The methodology for material attractiveness evaluation was developed to assess material attractiveness for states and treat IMF which is chemically inert for nitric acid solution. The material attractiveness was relatively assessed with physical properties of material in each of three discrete phases in the development of a nuclear explosive device. The material attractiveness assessment for non-state actors revealed that the non-irradiated TRISO fuel particle and IMF kernel for high temperature gas cooled reactors and the non-irradiated mixed oxide fuel (MOX) powder for MOX light water reactors were the most vulnerable targets in each fuel cycles. The TRISO fuel particle and IMF kernel would have less material attractiveness than the MOX powder because of their greater processing time and complexity. The material attractiveness assessment for states aiming concealed diversion revealed that the TRISO fuel particle and IMF kernel have less material attractiveness than the MOX powder, and are regarded as irradiated uranium fuel grade in the material attractiveness.

1. Introduction

The utilization and destruction of the transuranic (TRU) nuclides in the form of ceramic-coated fuel particles through a high burnup irradiation (Deep burn) has been studied in light water reactors and high temperature gas cooled reactors (HTGRs) to reduce the volume of high level radioactive waste and concerns about nuclear proliferation especially for plutonium (Rodriguez et al., 2003; Hong et al., 2013; Jo et al., 2009; Sakai et al., 2010). In the study on deep-burning, utilization of inert matrix fuel (IMF) which is TRU diluted with neutronically inert matrix has been proposed for higher fuel burnup and better controllability by reducing self-shielding effect as a counter for ^{240}Pu accumulation along with burnup and high excess reactivity (Jo et al., 2009; Sakai et al., 2010; Aoki et al., 2018).

The IMF composed of a stable solid solution of TRU oxide and yttria stabilized zirconia in fluorite-type crystal structure is difficult to extract plutonium from that because of its specific crystal structure (Fukaya et al., 2014). Theft and diversion of nuclear material to produce the nuclear explosive device (NED) is one of the most significant threats in nuclear security and non-proliferation, respectively. The chemical stability of IMF is expected to enhance intrinsic resistance protecting nuclear material from the NED manufacturing by states and non-state actors, but there are no efforts to evaluate the intrinsic resistance of IMF against the nuclear security threat quantitatively. Impacts on IMF utilization on the proliferation resistance including the intrinsic resistance against NED manufacturing by states have been discussed with weighting factors dependent on expert knowledge and experiences (Akie et al., 1994). The intrinsic resistance of plutonium for states has been studied in terms of the heat content and the spontaneous fission emission rate hindering nominal yield of NED (Kimura et al., 2011, 2012). The material attractiveness, the relative utility of nuclear material for an adversary to assemble a NED, enables a comprehensive evaluation of the intrinsic resistance of nuclear material and an interface analysis based on a simplified threat basis system response analysis in malicious NED manufacturing process composed of acquisition, processing and utilization phases (Bathke and Inoue, 2013).

In the present paper, comprehensive and quantitative evaluations of the material attractiveness are performed for the IMF in its fuel cycle using HTGR assuming threats of NED manufacturing by states and non-state actors.

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2. Methodology

The material attractiveness is used to investigate an impact of the characteristics of IMF on the utility of nuclear material for states and non-state actors for a NED manufacturing. The measures and its scale of the material attractiveness were developed to assess a utility of nuclear material not only for non-state actors but for states. It should be noted that the material attractiveness is only available for relative assessment between nuclear materials. The material attractiveness is assessed for target materials in two fuel cycles to compare each other, as shown in Fig. 1; an open fuel cycle using the IMF kernel in an HTGR (*IMF_HTGR* fuel cycle) and a closed fuel cycle using mixed oxide (MOX) fuel in a LWR (*MOX_LWR* fuel cycle). This study investigates the nuclear material in the fuel fabrication facility, and in the fresh fuel (FF) and spent fuel (SF) storages in the nuclear power plant as target nuclear materials that could be diverted. The target materials assessed in the *IMF_HTGR* fuel cycle are the IMF kernel, TRISO fuel particle, and carbide fuel compact; and in the *MOX_LWR* fuel cycle, the MOX powder (U:Pu = 1:1 at.%) and the MOX rod. These target materials are located in the fuel fabrication facility. In addition, three types of storages in the nuclear power plant are considered for each of the following targets: the carbide FF block (Pu/Total = 0.94 kg/107 kg) and irradiated fuel (IF) block (Pu/Total = 0.43 kg/107 kg) in the *IMF_HTGR* fuel cycle, and the MOX FF assembly (Pu/Total = 6.0 kg/260 kg) and the MOX IF assembly (Pu/Total = 4.4 kg/260 kg) in the *MOX_LWR* fuel cycle. Target materials in the reactor core are excluded from this investigation because threats of utilization from the reactor core appear to be negligible because of the extremely high radiation exposure and other major obstacles to accessibility. The fuel composition and physical property of nuclear materials in IMF-HTGR fuel cycle referred to previous study (Aoki et al., 2018). The composition of the MOX FF assembly is derived from a paper, and the composition of the MOX IF assembly is calculated using an ORIGEN-ARP code and the JENDL3.3 library (Radition, 2018; Shibata and Kawano, 2002).

Fig. 2 shows the following three discrete phases in NED development from target materials, along with assessment measures applied to each phase. In the acquisition phase, the adversary seizes the target material from a nuclear facility regardless of physical protection. In the processing phase, the adversary converts the target material to a final plutonium metallic form. Finally, the adversary utilizes the final form to assemble a NED in the utilization phase.

First, the measures for each phase were evaluated quantitatively and qualitatively. Table 1 show the evaluation results of the measures for target materials in the fuel fabrication facility and the nuclear power plant. Subsequently, target materials were divided into four categories by applying the scales shown in Tables 2 and 3 to the assessment results. After the categorization, applicable measures were selected based on assumed threat characteristics described in sections 4.2 and 4.3 against NED manufacturing by non-state actors and state, respectively.

In the acquisition phase, the target material's net weight, acquisition time, and dose rate at 1 m were considered as measures of the material attractiveness. The net weight is the total weight of the target material necessary to obtain a Category I quantity of plu-

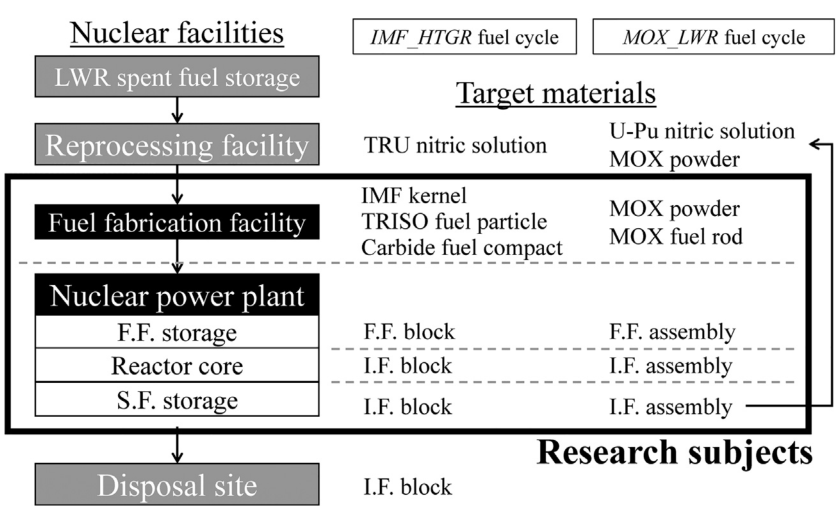

Fig. 1. Target materials in the nuclear fuel cycle for two process flows: the IMF use in a HTGR and the MOX fuel use in a LWR.

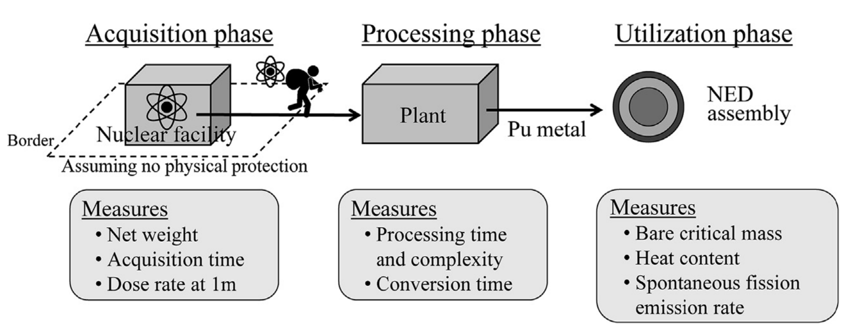

Fig. 2. Discrete phases in NED development and relevant measures applied in each phase.

Table 1
Measures of material attractiveness assessment for target materials.

Theft target	Acquisition phase			Processing phase		Utilization phase		
	Net weight [kg]	Acquisition time [min]	Dose rate at 1 m [Gy/h]	Processing time and complexity	Conversion time	BCM [kg]	Heat content [W/BCM]	SFN [neutrons/s/BCM]
Fuel compact	379 (9.3 kg × 41)	10	0	Conversion with TRISO and inert matrix removal	1–12 months (TRISO fuel and IMF)	21.2	259	7.6×10^6
TRISO fuel particle	127 (9.1 kg × 14)	10	0	Conversion with TRISO and inert matrix removal	1–12 months (TRISO fuel and IMF)	21.2	259	7.6×10^6
Fuel kernel	13 (13.2 kg × 1)	10	0	Conversion with TRISO and inert matrix removal	1–12 months (TRISO fuel and IMF)	21.2	259	7.6×10^6
F.F. block	320 (107 kg × 3)	40	0	Conversion with TRISO and inert matrix removal	1–12 months (TRISO fuel and IMF)	21.2	259	7.6×10^7
I.F. block	746 (107 kg × 7)	80	1	Conversion with TRISO and inert matrix removal	1–12 months (TRISO fuel and IMF)	25.4	2661	2.6×10^7
MOX fuel rod	118 (4 kg × 29)	10	0	Conversion in two or more steps (compounds)	1–3 weeks (Non-irradiated compound)	21.4	262	8.4×10^6
MOX powder	10 (12 kg × 1)	10	0	Conversion with relative difficult purification step (irradiated material)	1–3 weeks (Non-irradiated compound)	21.4	262	8.4×10^6
F.F. assembly	260 (260 kg × 1)	20	0	Conversion in two or more steps (compounds)	1–3 weeks (Non-irradiated compound)	21.4	262	8.4×10^6
I.F. assembly	260 (260 kg × 1)	20	1	Conversion with relative difficult purification step (irradiated material)	1–3 months (Irradiated material)	25.4	714	1.8×10^7

tonium metal. In present research, the Category I quantity of plutonium metal is defined as one eighth of the bare critical mass (BCM) of the plutonium metal, based on the relationship between the Category I quantity and the BCM of plutonium and ^{235}U (International Atomic Energy Agency, 2011; Ezoubtchenko et al., 2005). Target materials in the form of pellets, particles, or powder are assumed to be placed in cylindrical stainless-steel containers with a height of 40 cm, outer diameter of 19 cm, and weight of approximately 12 kg for easy management and transportation (Kawai, 2013). Target materials are categorized by net weight based on the capacity of a single person, a personal truck, and a commercial-sized truck (Bathke and Inoueve, 2013).

The acquisition time is the total time required to remove a Category 1 quantity of plutonium from the site. A 10 min operation time is assumed for the lifting by crane of a quantity not portable by man. An additional 10 min of operation time is assumed for removal of the target materials to the off-site. These were the only operation times accounted for in this study. Target materials are categorized by acquisition time based on the expected arrival time of response forces (Bathke and Inoueve, 2013).

The dose rate is defined as the dose rate in Gy/h at one meter from the theft target. In this study, the dose rate is assumed to be 0 for non-irradiated nuclear materials and 1 for irradiated nuclear materials, based on the International Atomic Energy Agency (IAEA) definition of IF in a reactor core (International Atomic Energy Agency, 2011). The threshold in the dose rate scale for materials portable by man is considered to be one order higher than that for materials not portable by man because portable targets are easier to shield and can be moved in a shorter time, allowing less exposure (Bathke and Inoueve, 2013).

In the processing phase, the conversion time and complexity are the only relevant measure of material attractiveness for non-state actors; it is a descriptive label corresponding to the most difficult process step that would be required to fashion the material into its final metallic form. Target materials are assessed and categorized for the measure of the conversion time and complexity based on the chemical processes and fission products necessary to obtain plutonium metal from the target material (Bathke and Inoueve, 2013). On the other hand, just the conversion time will be an effective barrier for proliferating states because they can ignore the complexity in processing phase due to their huge financial resources; it is defined as the time required to convert different forms of nuclear material to plutonium metallic components of a NED (IAEA, 2001). Target materials were categorized by conversion time based on the conversion times defined by IAEA (2001).

The process converting the TRISO fuel particle to the IMF kernel will need complex mechanical and combustion processes because the IMF kernel is protected by chemically inert and robust ceramics layers (Takei et al., 2003). And the plutonium extraction process from IMF kernel needs dissolution in nitric acid solution as well as irradiate fuels and will be difficult in the conventional PUREX process as the dissolution experiment of the IMF shows zirconium which is a host phase of plutonium dissolved only 2% even in concentrated nitric acid solution in 3 days (Fukaya et al., 2014). Then IMF will be quite difficult to extract plutonium completely even in several months and its conversion time should be greater than 3 months which is the conversion time estimated for the irradiated fuel. Therefore, the irradiated and non-irradiated TRISO fuel particles and IMF kernel are categorized in a level between that of irradiated material and a material that requires irradiation in the measures of conversion time and complexity, and conversion time in the processing phase.

In the utilization phase, the BCM, heat content, and SFN are considered as measures based on the physical properties of materials. The BCM is calculated at 0 °C using the MVP/GMVP II code and the JENDL-4.0 library (Nagaya et al., 2004; Shibata et al., 2011). The

Table 2
Scales of categorization for the material attractiveness for non-state actors.

Category	Acquisition phase			Processing phase	Utilization phase	
	Net weight [kg]	Acquisition time [min]	Dose rate at 1 m [Gy/h]	Processing time and complexity	BCM [kg]	Heat content [W/BCM]
1	<50	<15	<0.1 (1)[1]	Direct conversion in one step (metal form)	<80	<1292
2	50–3000	15–60	0.1 (1)-1 (10)	Conversion in two or more steps (compounds)	80–800	1292–6274
3	3000–6500	60–240	1 (10)–10 (100)	Conversion with relative difficult purification step (irradiated material)	800–4000	6274<
3/4				Conversion with TRISO and inert matrix removal		
4	6500<	240<	10 (100)<	Conversion requiring either irradiation or enrichment	4000<	

[1] The values in the brackets are for not man-portable materials.

Table 3
Scales of categorization for the material attractiveness for states.

Category	Processing phase	Utilization phase		
	Conversion time	BCM [kg]	Heat content [W/BCM]	SFN [neutrons/s/BCM]
1	1 week (Non-irradiated metal)	<80	<1292	$<1.56 \times 10^6$
2	1–3 weeks (Non-irradiated compound)	80–800	1292–6274	1.56×10^6-1.31×10^7
3	1–3 months (Irradiated material)	800–4000	6274<	1.31×10^7<
3/4	1–12 months (TRISO fuel and IMF)			
4	3–12 months (Low enrichment uranium)	4000<		

metallic phase of plutonium is assumed to be a delta phase, which is easy to compress and to insert excess reactivity in an implosion type of NED. Target materials are categorized by BCM with thresholds of 70, 20, and 10% of the BCM of U enrichment (Pellaud, 2002). The heat content is identified as the amount of heat generation that can be obtained from a unit BCM of the target material. Target materials are categorized by heat content with thresholds of 15 and 80% of the doping fraction of ^{238}Pu to ^{239}Pu, which is characterized by its thermal effect on the behavior of explosive material (Pellaud, 2002; Kimura et al., 2011). The SFN is identified as the SFN in neutron/s from a unit BCM of target material. Target materials are categorized by SFN with thresholds of 10 and 30% of the doping fraction of ^{240}Pu to ^{239}Pu (Kimura et al., 2012). The scale is determined based on the relationship between pre-detonation probability and the pressure propagation velocity in an implosion type of NED.

3. Results and discussion

3.1. Impacts on material attractiveness for non-state actors

For the material attractiveness assessment for non-state actors, non-state actors with the following characteristics are assumed as the adversaries who intend to divert the nuclear material into NEDs; (1) having no hesitation to commit suicide, (2) having no irradiation and enrichment capability, (3) having the capability to build a rudimentary NED, (4) having access to "garage-scale" processing capability, (5) accepting of any nuclear yield, (6) having support from one insider, and (7) strongly motivated to obtain a Category I quantity of target material in final metallic form (Bathke and Inoue, 2013). The processing time and complexity in the processing phase are considered in this assessment. The SFN is excluded in this assessment because non-state actors are not concerned with pre-detonation or the reliability of NEDs.

Fig. 3 shows assessment results based on measures for products and components in the fuel fabrication facility. Fig. 3(a) revealed that the carbide fuel compact in the *IMF_HTGR* fuel cycle had less material attractiveness than the MOX fuel rod because of the chemical stability of the TRISO fuel particle and the IMF kernel in the carbide fuel compact. Fig. 3(b) shows that the TRISO fuel particle and the IMF kernel had less material attractiveness than the MOX powder because of the former's larger net weight and/or increased processing time and complexity due to their chemical stability and small plutonium content.

Fig. 4 shows the results of material attractiveness assessment for non-state actors in the FF and SF storages in the *IMF_HTGR* and *MOX_LWR* fuel cycles. Fig. 4 shows that the irradiation of target materials in the *IMF_HTGR* fuel cycle decreased their material attractiveness, which was also the case in the *MOX_LWR* fuel cycle. The figure also revealed that the FF block had more material attractiveness than the FF assembly because of the former's significantly higher acquisition time, processing time and complexity, and heat

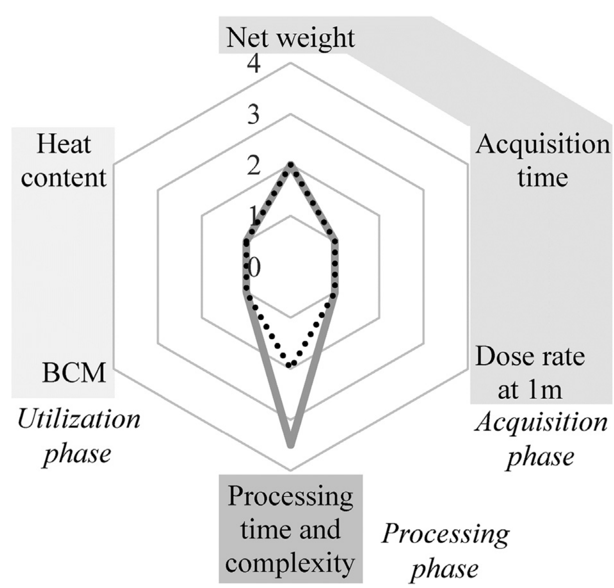

Fig. 3a. Material attractiveness for non-state actors of target materials in the fuel fabrication facility - Product.

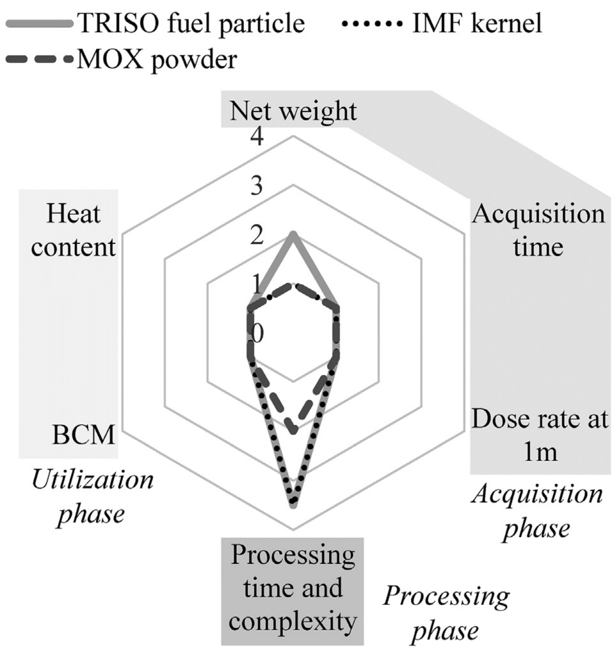

Fig. 3b. Material attractiveness for non-state actors of target materials in the fuel fabrication facility - Component.

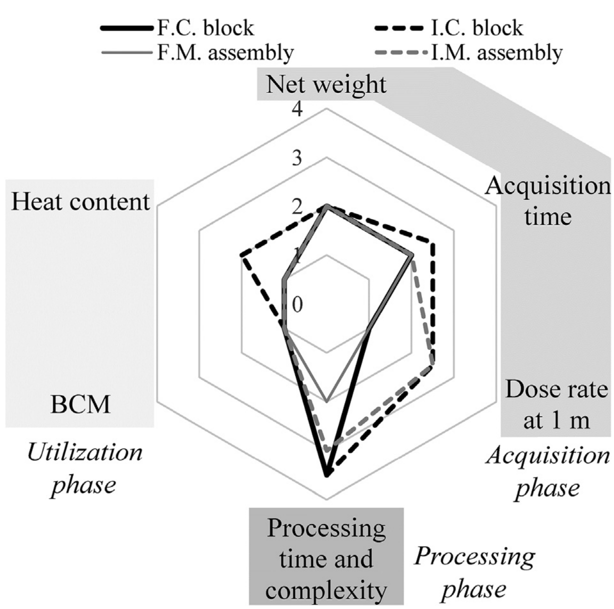

Fig. 4. Material attractiveness for non-state actors of target materials in the nuclear power plant.

content. The significantly higher acquisition time increases the adversary's risk of being interrupted by powerful response forces in the acquisition phase.

To discuss the material attractiveness throughout the fuel cycle, relative attractiveness is considered. Target materials are characterized by relative attractiveness indicating the lowest material attractiveness of a factor among the target materials in each phase. Fig. 5(a)–(c) shows the relative attractiveness of target materials in the *IMF_HTGR* and *MOX_LWR* fuel cycles in each discrete phase in NED development process. According to Fig. 3, the most vulnerable targets in the fuel fabrication facilities in the *IMF_HTGR* and *MOX_LWR* fuel cycles were the TRISO fuel particle and the IMF kernel, and the MOX powder, respectively. In the acquisition and utilization phases, there were no differences in the relative attractiveness of the most vulnerable targets in the *IMF_HTGR* and *MOX_LWR* fuel cycles. On the other hand, the TRISO fuel particle and the IMF kernel had less material attractiveness in the processing phase relative to the MOX powder because the chemical stability of the TRISO fuel particle and IMF kernel result in a greater processing time and complexity than for the MOX powder.

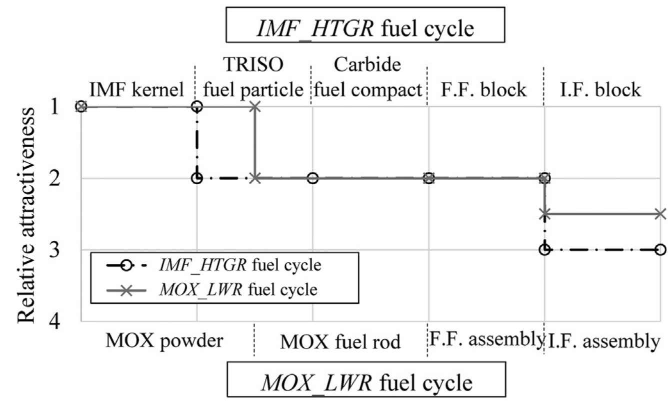

Fig. 5a. Relative attractiveness for the acquisition phase.

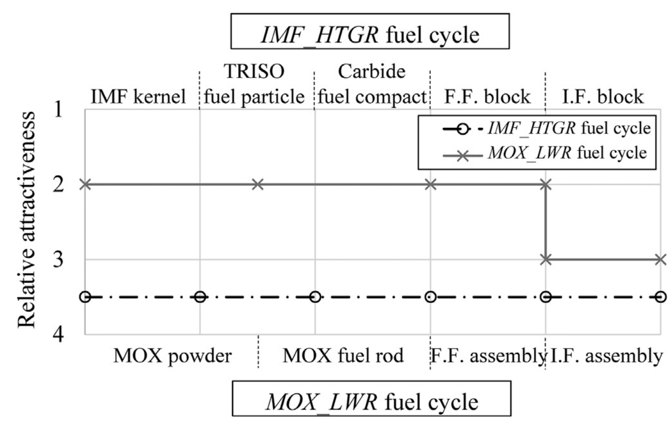

Fig. 5b. Relative attractiveness for the processing phase.

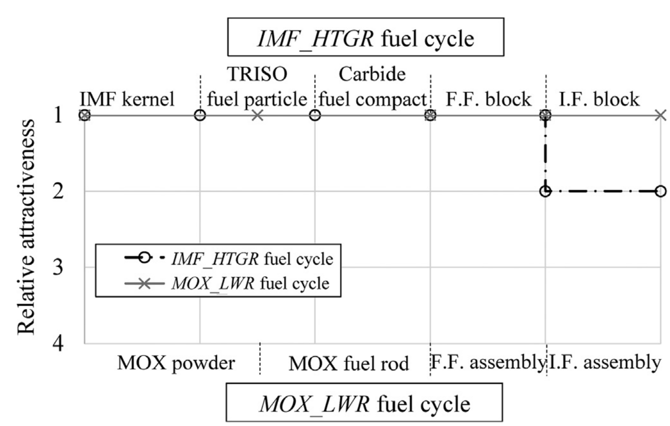

Fig. 5c. Relative attractiveness for the utilization phase.

Even assuming a 10% plutonium extraction rate from the chemically stable TRISO fuel particle and IMF kernel, the most vulnerable materials in the *IMF_HTGR* fuel cycle still had less material attractiveness than the MOX powder. Because a 10% plutonium extraction rate required approximately 10 times the amount of target material to obtain the Category I amount of plutonium metal. For the fuel block in the *IMF_HTGR* fuel cycle, acquiring 10 times the net weight of the target material in the acquisition phase required significantly more acquisition time, further diminishing its material attractiveness.

The material attractiveness assuming direct utilization of the TRISO fuel particle and the IMF kernel for a NED manufacturing was also studied. In this case, the adversary was assumed to intend to divert the TRISO fuel particle and the IMF kernel from the fuel fabrication facility to NEDs directly, without removal of the carbide coating and plutonium extraction in the processing phase. TRISO fuel particles and IMF kernels were assumed to be packed in a graphite matrix in a core of NEDs in the theoretically highest packing fraction (74%). In this scenario, the BCMs of TRISO fuel particles and IMF kernels mixed with the graphite matrix were approximately 3800 kg and 360 kg, respectively, and their heat contents were approximately 7800 W/BCM and 730 W/BCM, respectively. The BCM and heat content of the TRISO fuel particle and IMF kernel are relatively larger than that of the plutonium metal extracted from them. Relatively large BCMs increase the net weight in the acquisition phase. Therefore, the direct utilization of the TRISO fuel particle and the IMF kernel was shown to have less material attractiveness in the acquisition and utilization phases because of this significant increase in the BCM, heat content, and net weight over that of plutonium metal.

3.2. Impacts on material attractiveness for states

For material attractiveness assessment for states, a proliferating state intending to divert the safeguarded nuclear material into NEDs is assumed to have the following characteristics: (1) having advanced technology, well-developed industries, and abundant capital; (2) not having natural uranium resources; (3) accepting of the Non-Proliferation Treaty, comprehensive safeguards agreement, and additional safeguards agreements; (4) requiring 50% reliability for the NED; and (5) requiring production of one NED (Bari et al., 2011). Proliferating state's diversions may be tracked with various statistics, depending on each state's particular circumstances. In this study, concealed removal of declared material from a nuclear facility under international safeguards, or "concealed diversion," is considered as a representative statistic (Zentner et al., 2009).

From the concealed diversion statistic, it may not be difficult for proliferating states to access nuclear materials in their own nuclear facilities. After thus disregarding the acquisition phase, the conversion time in the processing phase, and the BCM, heat content, and SFN in the utilization phase are assessed because the proliferating state would be concerned over the risk of detection and the reliability of the NED. Fig. 6 shows the results of the material attractiveness assessment for states for both products and components in the fuel fabrication facilities in the *IMF_HTGR* and *MOX_LWR* fuel cycles. Fig. 7 shows the results for target materials stored in FF and SF storages in the *IMF_HTGR* and *MOX_LWR* fuel cycles. Figs. 6 and 7 revealed that the TRISO fuel particle, the IMF kernel and the carbide fuel compact in the *IMF_HTGR* fuel cycle and the MOX powder and MOX fuel rod in the *MOX_LWR* fuel cycle were the most vulnerable diversion targets. Comparing the results for the products of *IMF_HTGR* and *MOX_LWR* fuel cycles in Fig. 6(a), the carbide fuel compact had less material attractiveness than the MOX fuel rod from the perspective of conversion time in the processing phase because of the chemical stability of the TRISO fuel particle and the IMF kernel. Comparing the material attractiveness of these most vulnerable targets in each fuel cycle, Fig. 6 showed that the TRISO fuel particle, the IMF kernel and carbide fuel compact had less material attractiveness than the MOX powder and MOX fuel rod as a target because of their chemical stability and resultant

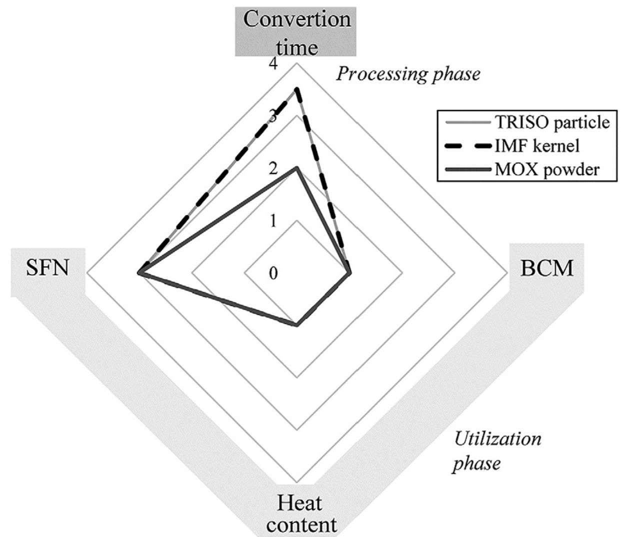

Fig. 6a. Material attractiveness for states of target materials in the fuel fabrication facility - Product.

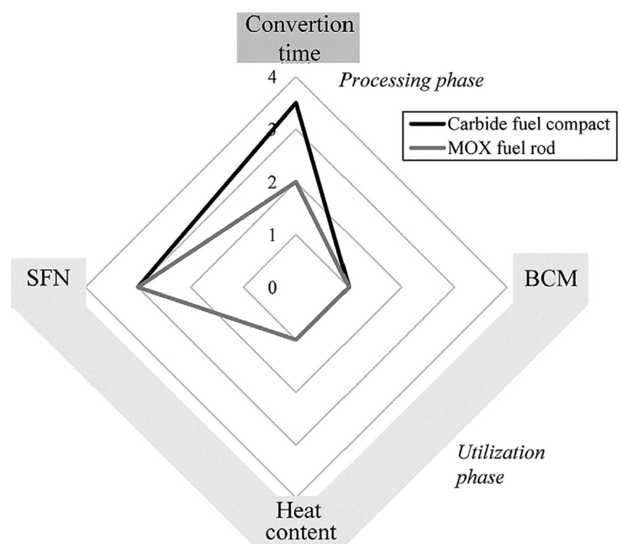

Fig. 6b. Material attractiveness for states of target materials in the fuel fabrication facility - Component.

greater conversion time relative to the MOX powder and MOX fuel rod. In a scenario assuming low plutonium extraction rate in processing phase and the utilization of large amount of target material, the material barrier in the utilization of nuclear materials will lower relatively. An effectiveness of the technical barriers such as detectability also have to be assessed in the case in future because the technical barrier can compensate for the material barrier's weakness. Here, the TRISO fuel particle and IMF kernel is made from TRU recovered from irradiate uranium fuels. The TRISO fuel particle and IMF kernel has nearly equal BCM, heat content and SFN in the utilization phase to those of the irradiated uranium fuel because of similar Pu isotopic composition. Their conversion time was estimated as longer than the irradiated uranium fuel as described. Then the material attractiveness of TRISO fuel particle and IMF kernel for state was less than that of the irradiated uranium fuel.

Fig. 7 revealed that the IF block had less material attractiveness than the FF block in the *IMF_HTGR* fuel cycle because of its higher decay heat generation from the plutonium metal in the utilization

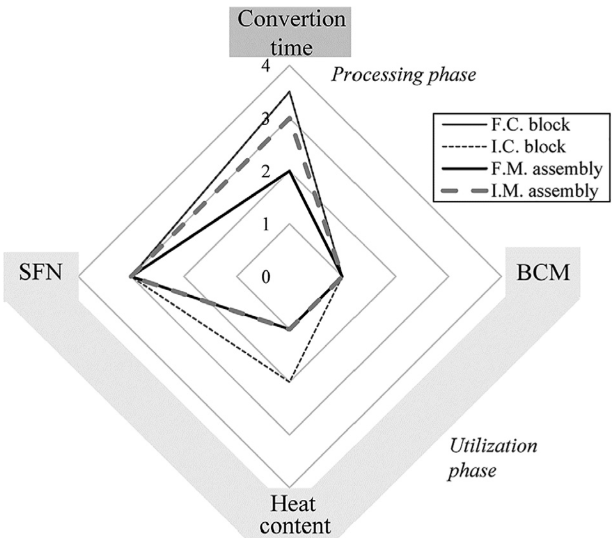

Fig. 7. Material attractiveness for states for target materials in the nuclear power plant.

phase, which hinder the manufacture of NEDs. The assessment results for the IF block and the IF assembly also revealed that the IF block had less material attractiveness than the IF assembly because the its higher decay heat generation hinders the manufacture of NEDs due to the high plutonium incineration ratio and buildup of ^{238}Pu in the HTGR, in addition to the chemical stability of the TRISO fuel particle and IMF kernel.

The impact of chemical stability of IMF on the material attractiveness were evaluated comparing with the MOX powder. The IMF utilization has some expected technical challenges in the material accountancy and the safeguards activity because radiation emission from IMF especially from minor actinides and lanthanides make it difficult to measure plutonium accurately by the radiation measurement.

4. Conclusion

The material attractiveness of inert matrix fuel was evaluated for states and non-state actors aiming for a nuclear explosive device manufacturing in the HTGR system using TRU oxide target to reveal the impact of its chemical stability.

The methodology for material attractiveness evaluation was developed to assess material attractiveness for states and treat IMF which is chemically inert for nitric acid solution. For the material attractiveness evaluation for non-state actors revealed that non-irradiated TRISO fuel particles and IMF kernels in the fuel cycle using IMF in the HTGR, and the non-irradiated MOX powder in the fuel cycle using MOX fuel in LWR were the most vulnerable targets. The TRISO fuel particle and the IMF kernel had less material attractiveness than the MOX powder for use because of their greater processing time and complexity due to their chemical stability. Even assuming a 10 wt% of plutonium extraction from the chemically stable IMF kernel, these still have less material attractiveness because approximately 10 times more net weight would be required in the acquisition phase. The irradiated fuel in HTGR had lower material attractiveness than unirradiated one because of higher decay heat hindering NED manufacturing in the utilization phase.

The evaluation of material attractiveness for states assuming the concealed diversion of declared material reveal that non-irradiated TRISO fuel particles and IMF kernels, the non-irradiated MOX powder were the most vulnerable targets in each fuel cycle. The TRISO fuel particle and the IMF kernel had less material attractiveness as a target than the MOX powder because of their greater conversion time due to their chemical stability, and were regarded as irradiated uranium fuel grade in material attractiveness. The irradiated fuel in the HTGR had lower material attractiveness than unirradiated one because of high decay heat generation hindering NED manufacturing and its nominal yield generation in the utilization phase.

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