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Performance Evaluation According to Polymer Encapsulation Characteristics of Eco-Friendly Plastic Gamma-Ray Shield

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Abstract: To eliminate the exposure of medical staff to radiopharmaceuticals during nuclear medicine tests, a new process technology was proposed for manufacturing gamma-ray shields used in nuclear medicine. In the case of manufacturing the existing gamma-ray shield, a method of increasing the content of the shielding material in the mixed material is used to improve the shielding performance. However, it is impossible to improve the shielding performance by simply increasing the content of the shielding material. Therefore, this study aims to present the optimal conditions for improving the miscibility between composite materials. Following the additional mixing of barium sulfate and bismuth oxide with tungsten, a syringe shield was developed via a plastic injection mold process. When tungsten was solely used or in combination with other shielding materials, polymer encapsulation occurred, and miscibility between composite materials was observed. Based on these results, the optimal conditions in terms of eco-friendly materials, economic feasibility, and improvement in shielding performance were determined. The findings of this study reveal that when tungsten and the polymers are combined, the polymer encapsulation is optimal, the particles are uniformly dispersed, and the shielding performance is significantly improved. With a ^{99m}Tc source, a 6.9% improvement in the shielding performance is achieved compared with the use of lead.

Keywords: barium sulfate; bismuth oxide; tungsten; polymer composite material; gamma-ray shielding

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1. Introduction

X-rays and γ-rays are used during diagnosis and treatment in medical institutions. Currently, X-rays, which are located in the low-energy region of the electromagnetic spectrum, are used for diagnostic imaging, whereas γ-rays, which are located in the high-energy region, are used in nuclear medicine diagnosis and treatment [1]. As nuclear medicine tests are conducted such that radiopharmaceuticals are directly administered to patients, facility staff and personnel surrounding the patient are readily exposed to these rays [2,3]. Therefore, a shield for active defense is required.

For X-rays, the representative shield is an apron, which is made of a 0.25 mmPb lead equivalent and used as clothing to protect the front side of the body [4]. When shielding γ-rays, a Pb apron with a thickness within the range of 0.3–0.5 mmPb is typically worn and used to prevent scattering rays from the human body before and after the injection of radiopharmaceuticals [5]. Direct radiation occurs during the direct injection of radiopharmaceuticals, and exposure often occurs when patients are injected with radionuclides.

Although the quantity of radiopharmaceuticals used in nuclear medicine diagnostic areas is significantly low, the dangers of radiation exposure due to radionuclide synthesis, distribution, and injection of radionuclides by radiologists and medical personnel should be considered [6].

Currently, representative nuclides used in nuclear medicine include ^{99m}Tc, ^{131}I, ^{18}F, ^{123}I, ^{201}Tl, ^{67}Ga, ^{111}In, and ^{89}Sr. Due to the differences between the types of radiation, energy intensity, and half-life of the nuclide, caution should be exercised [7,8]. The International Commission on Radiological Protection recommends the use of a syringe shield to reduce

exposure to the hands of medical staff when injecting radionuclides. Currently, most shields used in medical institutions contain lead as the primary component [9]. Lead is an excellent material for manufacturing various types of shields due to its excellent processability and economic efficiency. However, given that heavy metals pose a risk of exposure, which is harmful to the human body, and contamination during disposal, shields used in medical institutions are replaced with lead-free materials [10,11]. Currently, tungsten, bismuth oxide, and barium sulfate have been proposed as substitutes for lead and eco-friendly materials. However, tungsten exhibits limited processability as it starts melting at 3400 °C or higher temperatures [12]. When bismuth oxide or barium sulfate is used as a shielding material, the thickness of the shield should be increased significantly to achieve an effective shielding performance; this limits its application [13]. Therefore, a syringe shield made of an eco-friendly material instead of lead can only be commercialized when new shielding materials or mixtures are realized, which requires an improvement in the process technology.

Therefore, in this study, a syringe shield was fabricated using either tungsten or a composite material of tungsten, bismuth oxide, and barium sulfate, and its shielding properties were investigated. The mixing characteristics between the particles of the shielding material and the miscibility between the polymer and the shielding material of a syringe-type shield manufactured via an injection mold process were investigated [14]. The correlation between the differences in polymer encapsulation with respect to the characteristics of particle dispersion, the dispersion state between particles, and the shielding performance during the mixing process of the shielding particles and the polymer material were analyzed, in addition to the injection process of the mixture. Moreover, new characteristics of pinholes and the affinity of the two materials, which are dependent on the characteristics of the shielding material particles when mixed with polymer materials, were reported. Barium sulfate and bismuth oxide are economical among eco-friendly shielding materials, and tungsten exhibits a similar shielding performance as that of lead due to its high density [15]. When manufacturing a radiation shield, various materials can be mixed and used considering the economic efficiency and shielding performance [16]. Therefore, in this study, the shielding performance and degree of particle dispersion were evaluated with respect to the miscibility of the materials used.

To increase accessibility and convenience for nuclear medicine practitioners, an injection process technology that could mix plastic and shielding materials was developed to manufacture a syringe shield. In addition, this process replaced the use of lead, which is a heavy metal, by an eco-friendly shielding material [17]. The eco-friendly shielding materials used in this study were tungsten, which has a higher density than lead and an excellent shielding performance; and barium sulfate and bismuth oxide, which have economic advantages [18]. In addition, a polyamide nylon resin (PA66), which exhibits excellent mechanical strength, was selected as a polymer material to be mixed during the mold injection of this composite material [19,20].

The aim of this study was to compare the shielding performances, affinities with polymer, and internal particle distributions of a shield manufactured using a single material and another shield manufactured using a composite material. Therefore, a shield containing only tungsten; a shield containing tungsten and bismuth oxide; a shield containing tungsten and barium sulfate; and a shield containing tungsten, bismuth oxide, and barium sulfate were fabricated. In addition, after evaluating the shielding performances of the four prepared samples, two optimal samples were used to prepare a syringe shield for comparison with a lead-based syringe shield.

The shielding performances of four samples, as well as the shielding performance and lead equivalent of a plastic syringe shield developed accordingly, were investigated. In addition, the internal particle dispersibility and polymer encapsulation of the shields prepared using single materials and composite materials were visually observed using a field-emission scanning electron microscope. The findings of this study can serve as a basis for the realization of favorable process conditions for injection molds, such as properties

with respect to the combination of materials and shielding performance according to polymer encapsulation when manufacturing a gamma-ray shield. Moreover, the findings of this study can be used as basic data for the development of an eco-friendly shield that helps reduce exposure to workers when injecting radionuclides in the nuclear medicine field.

2. Materials and Methods

The shielding effectiveness of X-rays and γ-rays is indicated by a decrease in the intensity of radiation when it passes through the shield [21]. Therefore, with an increase in the density of the shield or the thickness of the shield, the radiation intensity decreases [22,23]. The intensity of the transmitted photons can be expressed using Equation (1).

$$I = I_0 exp(-\mu x), \quad (1)$$

where I is the intensity of the photons after transmission; I_0 is the intensity of the incident photon energy; μ is the linear attenuation coefficient; and x is the thickness of the shield.

When radiation penetrates a shield made of tungsten particles, the intensity attenuates according to the thickness of the shield, as expressed by Equation (2).

$$-\frac{dI}{dx} = I\sigma N, \quad (2)$$

where σ is the unit area where the mass attenuation occurs and N represents the number of elements per unit volume of the tungsten shield. In particular, the intensity of radiation decreases as the probability of interaction increases [24].

Therefore, $I\sigma N = \mu_m(\mathrm{cm^2 g^{-1}})\rho(\mathrm{g{\cdot}cm^{-3}})$ can be expressed as the product of the mass attenuation coefficient (μ_m) and the density (ρ). By distributing the particles over a large area, increasing the interaction cross-sectional area (σ) and increasing the thickness of the shield can induce attenuation of the transmission intensity, as expressed by Equation (3) [25].

$$I = I_0 exp(-\mu_m \rho x). \quad (3)$$

Based on these characteristics, by adjusting both the distribution area of the shielding material with a high number of atoms and the thickness of the shield, the shielding performance can be improved [26].

A dispersion of tungsten particles was obtained using a process technology wherein a polyamide nylon resin (PA66) in a solid state was powdered and mixed with tungsten particles. The process was modified for simpler stirring when PA66 was mixed in a liquid state. This modification of the process technology increases the dispersing power of particles in the injection-molding process. This is because when the polymer is in a solid powder state, the miscibility between materials is superior to that in a liquid state. Rather than mixing high-density liquid-state polymers with metal particles, the tendency to be easily distributed between particles is higher when solid-state polymers are used [27,28].

In this study, a 10 mL syringe was used, and the syringe shield was designed with a radius, length, and outer wall thickness of 20 mm, 90 mm, and 3 mm, respectively, to fabricate a mold frame. The four panels realized for comparing the syringe shield and shielding performance had dimensions of 150 mm × 150 mm × 3 mm (150 ± 0.3 g).

The content of the shielding material was set within the range of 85–90 wt% as a standard, and after preprocessing by applying the premixing process technology, it was injected into the mold using a screw extruder. At this time, the output temperature of the injection machine was within the range of 170–220 °C. Moreover, the screw speed (0.2 l bs/h) inside the injection machine varied with temperature; therefore, it was adjusted at approximately 10% considering the sample size. The specific gravity of tungsten varies depending on the specific gravity of the entire mixed material, and the shielding performance was evaluated while other conditions were kept the same. The size of the tungsten particles used was 4.36 μm, and the sizes of barium sulfate and bismuth oxide particles were in the range of

40–50 μm. The reason that the particle size of the tungsten particles and other materials was different was to reduce the voids between the particles during mixing.

The fabricated sample panel is shown in Figure 1. A component analysis of the samples was performed using an X-ray fluorescence (XRF) analyzer (XRF-1800, Shimadzu, Japan). XRF is an analysis method that uses fluorescence X-rays generated when X-rays are injected into a sample to identify specific peaks according to the elements present [29]. In addition, to analyze the degree of dispersion inside the shield, the sample shield was visually observed using field-emission scanning electron microscopy (FESEM) via an optical microscope (S-4800, Hitachi, Japan) [30].

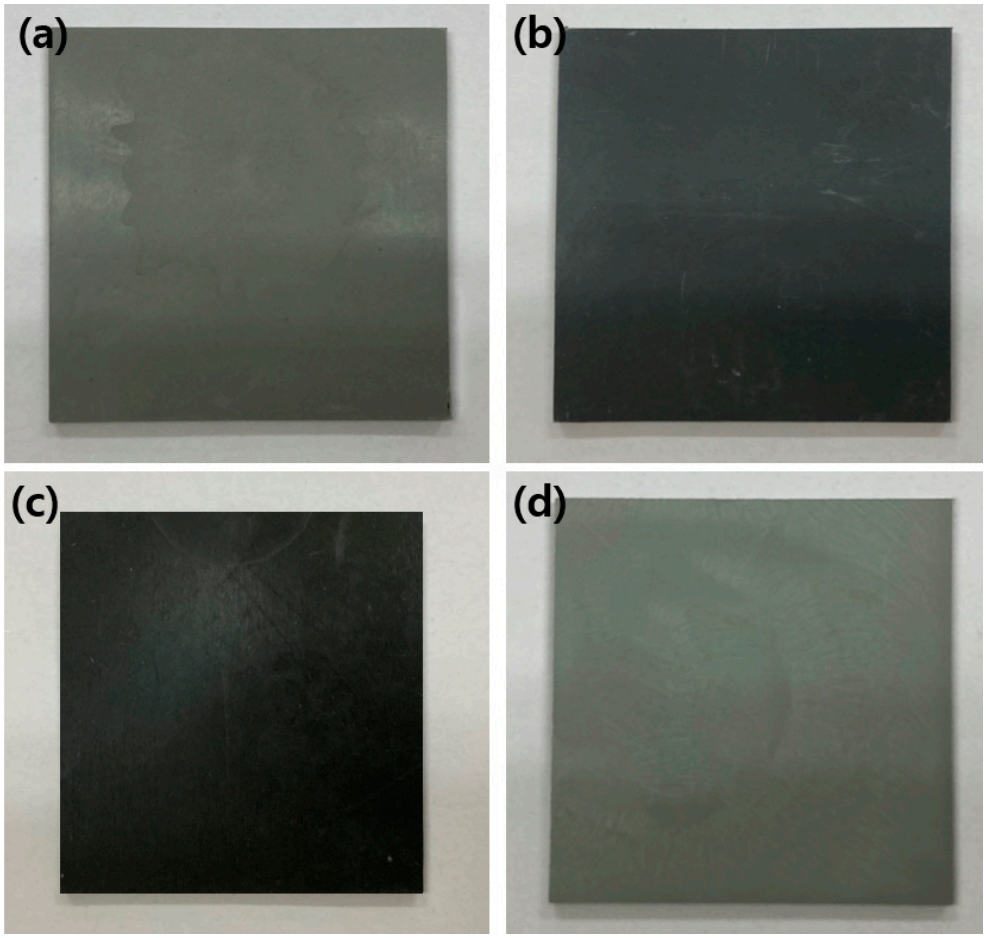


Figure 1. Mold samples through shielding material mixing composition (150 mm × 150 mm × 3 mm (150 ± 0.3 g)): (**a**) PWBa-80 (W, Ba), (**b**) PWBi-80 (W, Bi), (**c**) PW-85 (W), and (**d**) PWBaBi-80 (W, Ba, Bi).

In this study, ^{99m}Tc (200 μCi), ^{131}I (300 μCi), and ^{18}F (300 μCi) were used as radioactive isotopes in the experiment to evaluate the shielding performance according to the composition of the polymer composite shielding material. A dose calibrator (CRC-15R, Mirion Technologies (Capintec), Inc., Florham Park, NJ, USA) was used to measure the amount of radioactivity, and the transmitted dose that passed through the sample shield was measured using a survey meter (Inspector USB, SE international, Inc., Summertown, TN, USA). The measurement was performed in a uterine cancer treatment radiation room, and the geometric structure was set as shown in Figure 2 [31,32]. The position of the radioisotope was moved and measured. The distance between the sample shield and the source was set as 0.3 m and 0.5 m. In addition, for the two samples in which the particles were almost uniformly distributed, the shielding performance was evaluated at a distance of 0.3 m from the source, as shown in Figure 3, by manufacturing a syringe shield with the same weight as a lead product.

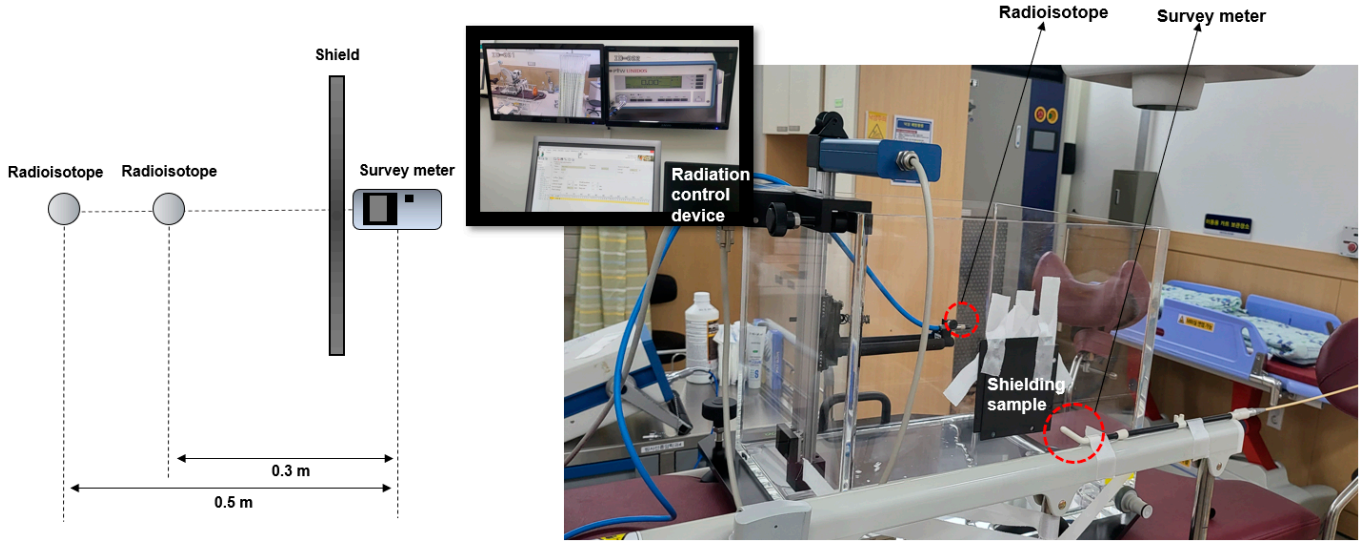


Figure 2. Geometric composition of the evaluation of the γ-ray shielding of samples.

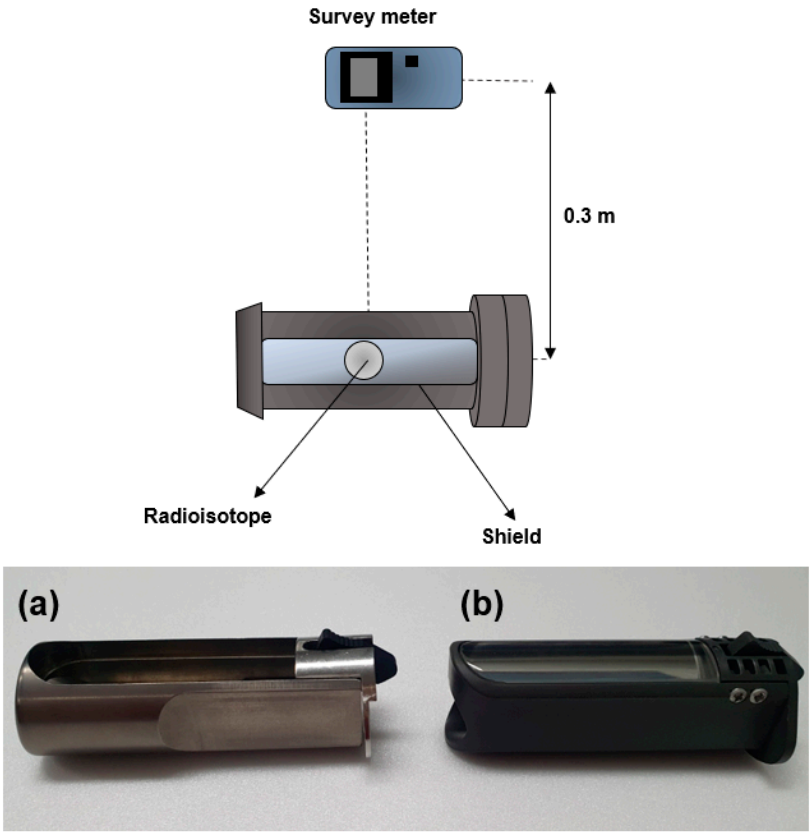


Figure 3. Appearance and measurement method of the manufactured radiation syringe shield: (**a**) existing lead syringe shield and (**b**) plastic syringe shield.

3. Results

Figure 4 presents the component analysis results of the prepared samples for the particle dispersion state of the polymer composite materials. The polymer composite materials used comprised barium sulfate, bismuth oxide, and tungsten. The samples were prepared by mixing the powder states of barium sulfate and bismuth oxide with tungsten, which exhibited an excellent shielding performance, to form a panel through an injection machine. Figure 5 presents the cross-sectional view of the fabricated samples captured

using an electron microscope. As shown in Figure 5c, wherein only tungsten was used as a single material, the cross-sectional view of the electron micrograph revealed that the distribution of particles therein was more uniform than those of the other samples. As shown in Figure 5d, wherein the composite material comprised tungsten, barium sulfate, and bismuth oxide, multiple-sized particles were scattered in the interparticle distribution. In addition, a comparison of Figure 5a,c revealed that the polymer material did not surround the shielding material particles and was aggregated in a conformal shape between the particles. Figure 5b reveals that the bismuth oxide particles were independently composed, and the miscibility was insufficient due to aggregation between the polymers.

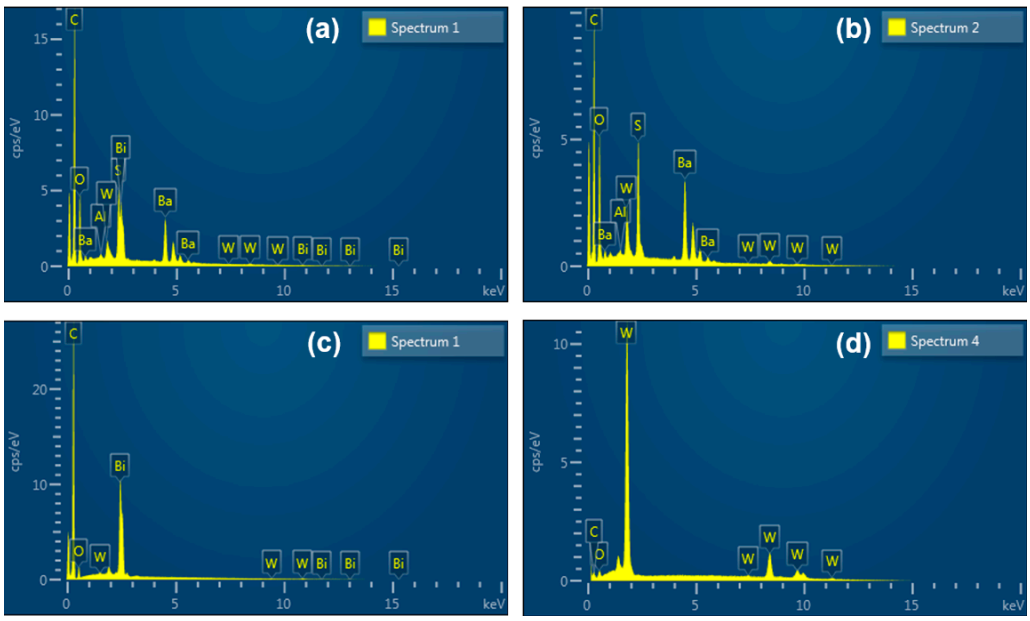


Figure 4. Component analysis results of the shield samples: (**a**) PWBaBi-80 (W, Ba, Bi), (**b**) PWBa-80 (W, Ba), (**c**) PWBi-80 (W, Bi), and (**d**) PW-85 (W).

Figure 6 presents an image of the compatibility between the shielding materials observed on a polymer basis in the polymer mixture for the production of γ-ray shielding. To increase the shielding effect, the voids should be reduced by improving the uniform distribution between the particles of the shielding material. Therefore, as shown in Figure 6a, when a single tungsten material was used, the compatibility with the powdery polymer was excellent, but in Figure 6b, the uniform distribution was insufficient despite the use of composite materials of various sizes. Therefore, the shielding performance could be further improved when the voids between the particles were fewer or the polymer was encapsulated by coating around the shielding particles. As shown in Figure 7, a constant gap between the tungsten particles was maintained due to the polymer encapsulation of tungsten, thereby reducing the number of voids.

In this study, ^{99}mTc, ^{131}I, and ^{18}F were used as radiation sources to evaluate the nuclear medicine gamma-ray shielding performances of the shield sample and the syringe shield manufactured by the injection molding process. Table 1 lists the characteristics of the sources used.

The aforementioned sources are the most commonly used for nuclear medicine testing and treatment. They are primarily introduced into the body in the form of injection. To verify the gamma-ray shielding effect, the doses and shielding rates of the four samples using ^{99m}Tc, ^{131}I, and ^{18}F sources are shown in Tables 2–4.

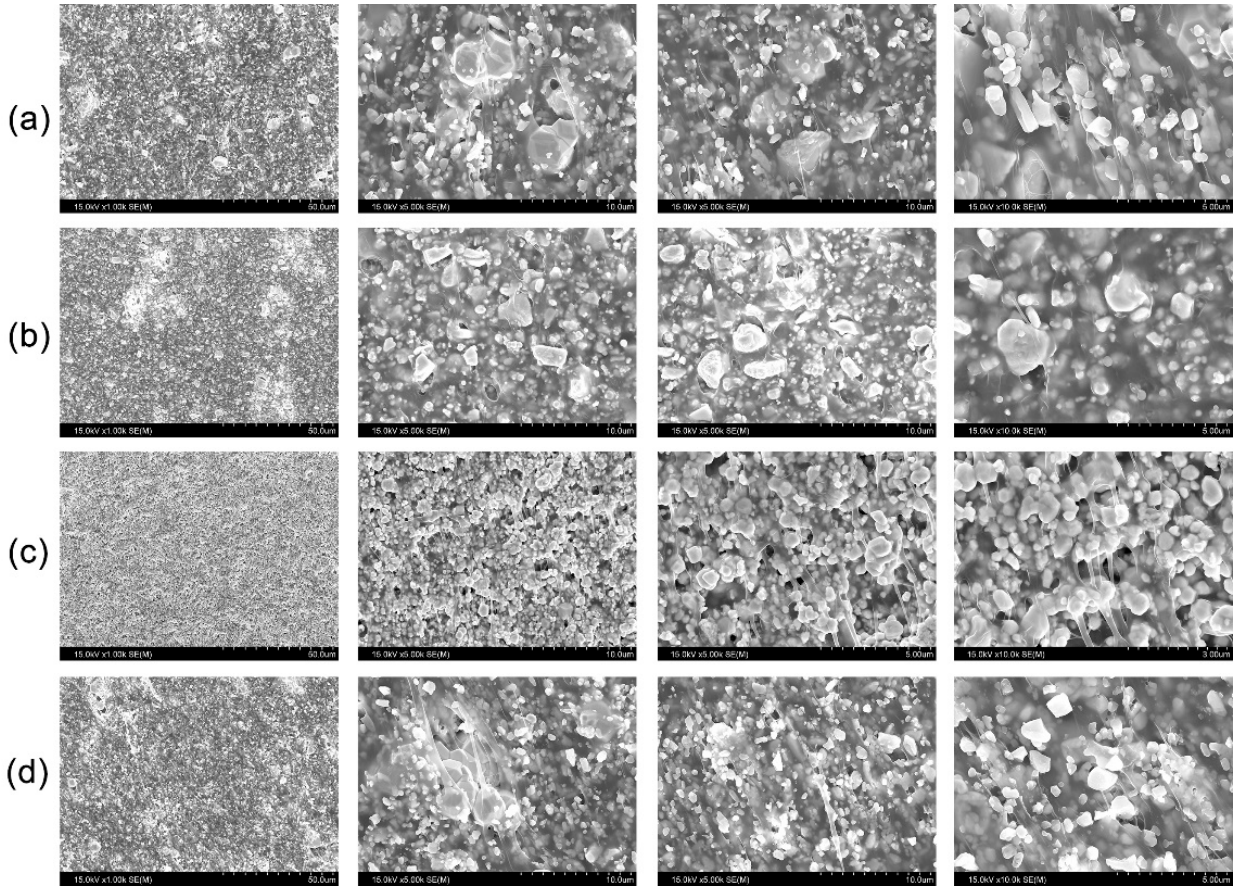


Figure 5. Cross-sectional particle analysis results of shield samples: (**a**) PWBa-80 (W, Ba), (**b**) PWBi-80 (W, Bi), (**c**) PW-85 (W), and (**d**) PWBaBi-80 (W, Ba, Bi).

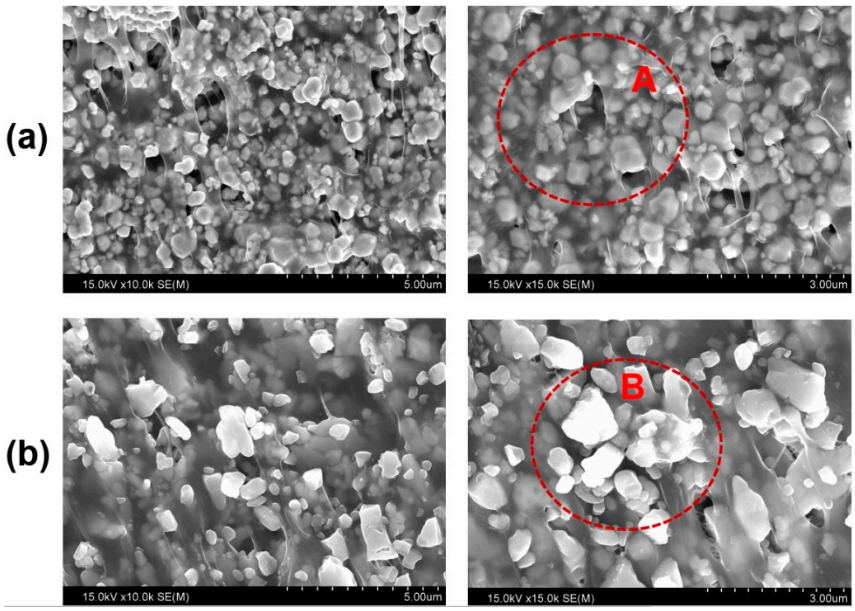


Figure 6. Comparative analysis of the cross-sectional images of single and composite materials of shield samples: (**a**) PW-85 (W) and (**b**) PWBaBi-80 (W, Ba, Bi). The circled portion labeled A exhibited the encapsulation phenomenon progressed by polymer coating around the particles of the shielding material, and B presents the state of the agglomeration between the particles of the composite material.

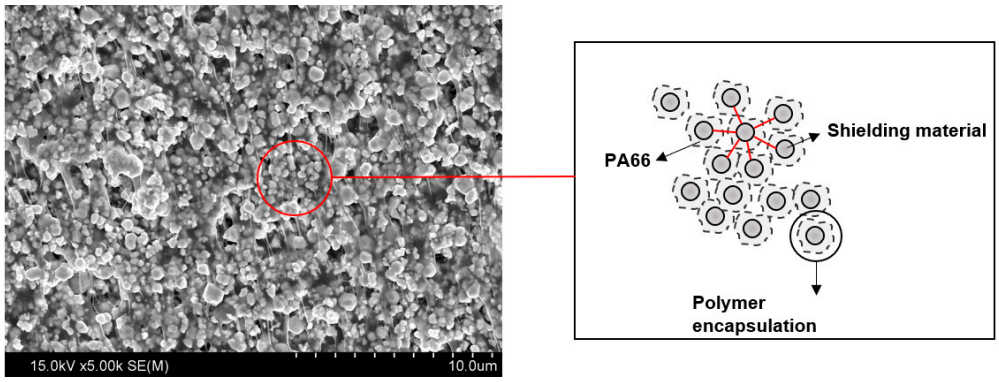


Figure 7. Description of the polymer encapsulation structure of tungsten particles.

Table 1. Characteristics of nuclear medicine sailors.

Characteristic		Radionuclide (keV)		
		^{99m}Tc	^{131}I	^{18}F
Half-life		6.01 h	8.04 d	109.9 min
Emitting Energy	ɤ emission	140.5 (0.89) 18.4 (0.04) 18.3 (0.02)	364 (0.81)	511.0 (1.94)
	β max emission	-	191 (0.89)	633.5 (0.97)

Table 2. Transmission dose and shielding rate of ^{99m}Tc.

Shield	Transmission Dose (mR/h)		Shielding Rate (%)	
	0.3 m	0.5 m	0.3 m	0.5 m
PW	2.012	1.622	88.9	93.7
PWBa	2.328	1.898	47.8	52.4
PWBi	2.220	1.769	68.2	73.0
PWBaBi	2.051	1.651	86.4	90.5

Table 3. Transmission dose and shielding rate of ^{131}I.

Shield	Transmission Dose (mR/h)		Shielding Rate (%)	
	0.3 m	0.5 m	0.3 m	0.5 m
PW	3.421	2.592	28.5	36.4
PWBa	4.495	3.364	8.9	10.9
PWBi	4.012	3.312	12.2	14.5
PWBaBi	3.644	2.814	26.5	29.1

Table 4. Transmission dose and shielding rate of ^{18}F.

Shield	Transmission Dose (mR/h)		Shielding Rate (%)	
	0.3 m	0.5 m	0.3 m	0.5 m
PW	19.012	15.021	18.2	20.7
PWBa	25.823	17.254	4.8	6.0
PWBi	22.151	16.982	7.2	9.5
PWBaBi	19.425	15.412	15.8	19.2

For the ^{99m}Tc source, the shielding performance of the shield made of a single tungsten material was the highest, and the shielding performance of that made of a mixture of barium sulfate and tungsten was the lowest. This phenomenon was almost the same for the other

sources. Moreover, regarding the uniform dispersibility of the shield cross-section particles, with an increase in the uniformity of the particle distribution, the shielding performance was improved. Consequently, given that barium sulfate and bismuth oxide demonstrated a lower shielding performance than that of tungsten, and due to the lack of interparticle miscibility, a direct relationship with the shielding performance was indicated. Therefore, when a single material was used instead of a composite material, the polymer encapsulation was uniformly distributed, and the dispersion degree of particles was improved. Finally, a syringe shield was manufactured using a sample in which all three materials were mixed and a sample with only tungsten. A comparison of the shielding performances of the two syringe shields with that of 2 mm-thick lead is presented in Table 5. Notably, the shielding performance of the 3 mm-thick plastic syringe shields were slightly superior. In particular, for the ^{131}I source, the plastic syringe shield exhibited a considerably high shielding performance. Although the syringe shield developed in this study was not manufactured with the same thickness as lead, it was manufactured with the same weight, so it is expected that the accessibility of medical staff will be the same.

Table 5. Comparison of shielding rate between syringe lead shield and tungsten-based plastic shields (based on a distance of 0.3 m).

Shield (150 ± 0.3 g)		**Shielding Rate (%)**		
		^{99m}Tc	**^{131}I**	**^{18}F**
Lead (2 mm)		82.02	3.25	26.41
Plastic (3 mm)	PW	88.9	28.5	18.2
	PWBaBi	86.4	26.5	15.8

4. Discussion

Nuclear medicine departments in medical institutions primarily use gamma rays; therefore, radioactive-protective aprons are used to reduce the radiation exposure risk of workers. These aprons are manufactured per the relevant standards IEC 61331-3;2003. However, the lead equivalent in these shields is based on the standard for diagnostic X-rays [33]. Therefore, it is necessary to increase the amount of lead equivalent for shields used for gamma-ray shielding in the actual medical field and to develop lightweight and eco-friendly materials.

In this study, a syringe shield product made of a polymer composite material was manufactured using eco-friendly materials such as tungsten, bismuth oxide, and barium sulfate instead of lead, and the gamma-ray shielding effect was evaluated. In addition, the shielding effect was evaluated according to the composition of single and composite materials by examining the mixed-particle structure of the polymer composite materials.

By maintaining the dense structure of the particle arrangement of the shielding material, the shielding performance is improved [34]. Generally, uniform dispersion cannot readily be achieved because shielding materials form agglomeration in the mixing process, or polymer materials are agglomerated due to insufficient affinity between the polymer and shielding materials [35]. In this study, to address this issue, a compounding process technology of a powdery plastic polymer with high processability and a shielding material was used.

This allows the polymer to be dispersed more effectively than when it is in a liquid state, thereby enabling the encapsulation of the polymer during the injection process. A thin layer of polymer surrounds the particles of the shielding material, which allows for strong adhesion with other shielding materials, irrespective of agitation during the injection process. Therefore, the spacing between particles can be reduced, thus resulting in uniform dispersion [36].

Given that the developed syringe shield in this experiment is intended for direct contact with the human body, an eco-friendly shielding material should be used rather than

lead, which is a heavy metal. Moreover, the reproducibility of the shielding performance should be considered. The shield manufactured by injection molding using the process technology presented in this study is highly accessible and can reduce the risk of lead for medical staff [37]. In this process, the standardization of manufacturing technology can be promoted by maintaining the reproducibility of the shielding performance and introducing a circular mold injection method. In particular, the reproducibility of the shielding performance is dependent on the content of the shielding material and the dispersion technology. Moreover, the stability of the dispersion technology can be secured through powder mixing and polymer encapsulation.

To secure the price competitiveness of the syringe shield, an appropriate mixed material should be composed, and the affinity between the materials should be considered. In addition, in the case of a polymer composite material, the particle dispersion of the shielding material should be considered by reducing the aggregation between the shielding materials and the polymer materials.

With reference to ORAMED Project Work package 4, ^{99m}Tc, ^{131}I, and ^{18}F sources are the most frequently used in nuclear medicine, and the minimum allowable syringe shield thickness is 2 mm or 5 mm. However, the ^{18}F shield requires a considerable thickness [38]. In particular, to secure the effectiveness of shielding gamma rays instead of X-rays, appropriate shielding thickness and shielding performance are required depending on the source of the radiation. Therefore, a study on shield thickness control and the content of the mixture is necessary [39]. If injection molding is performed in consideration of the characteristics of the polymer composite materials presented in this study, the suggested problems can be solved. This study was limited in that although a polymer composite material was used, the investigation of the affinities of various shielding materials according to the characteristics of the polymer was not comprehensive.

The composite materials used in this study were prepared by adding an eco-friendly gamma-ray shielding material, namely tungsten (a metal particle) in a state where its content was mixed at 50 wt% or greater, followed by secondary stirring inside the injection machine. Thereafter, injection molding was performed to prepare samples. Using the existing method, the affinity between particles cannot readily be examined by mixing the polymer in the liquid state. However, this process technology can be used to analyze the affinity between the composite materials by applying the composite material to an injection machine. When using only tungsten, polymer encapsulation is more advantageous, and this encapsulation phenomenon is more effective in dispersion. To increase the gamma-ray shielding efficiency, the efficient dispersion of the shielding material is critical process technology, and the process technology presented in this study can be used to manufacture an effective shield in addition to the syringe shield.

5. Conclusions

As confirmed, the properties of composite materials affect the shielding performance of gamma-ray plastic shields that can be used in nuclear medicine. When a composite shielding material is mixed with a polymer, the affinity and miscibility between the shielding material particles according to the particle characteristics affect the shielding performance. In this study, the influence on the shielding performance due to an increase in the affinity between shielding materials through polymer encapsulation was analyzed. The method of improving the shielding performance by compounding the shielding material is affected by the encapsulation of the polymer corresponding to the properties of the particles rather than the properties such as the density of the shielding material, and it has been confirmed that this affects the dispersion of the particles. The composite material-based plastic shields fabricated in this study showed 86.4%–88.9% shielding ability at ^{99m}Tc compared to ^{131}I and ^{18}F. Moreover, it showed the exhibited a shielding rate of 18.2%–15.8% in ^{18}F. Polymer encapsulation occurred more effectively when a single material was used instead of a composite material, and the shielding performance was approximately 2%–3% higher.

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