Natural rubber block as gamma radiation shielding for medical applications

O Onjun¹, N Buasuwan¹, T Rungseesumran², N Kamwang², J Channuie² and P Sinkaew²

¹Department of Science Service, Ratchathewi, Bangkok, 10400, Thailand
²Thailand Institute of Nuclear Technology, Chatuchak, Bangkok, 10900, Thailand
E-mail: orasa@dss.go.th

Abstract: In this study, a prototype of commercialized-style radiation shielding blocks based on natural rubber mixed with radiation shielding substance (i.e. lead and tungsten compound) was developed. The gamma shielding test was carried out using ¹³⁷Cs (662 keV) source and ⁶⁰Co (1173, 1332 keV) source. The results reveal that based on the gamma attenuation property and forming capability, the optimum formula was a shielding block with 60% lead oxide, which was able to completely shield gamma radiation (> 99%). In comparison with a radiation shielding block with lead oxide, a radiation shielding block with tungsten oxide showed similar characteristics. Due to the higher price of tungsten oxide, natural rubber block with lead oxide is preferred. However, with an environmental concern, natural rubber block with tungsten oxide is selected. Moreover, it was found that the mechanical properties of these radiation shielding blocks (including hardness, tensile strength and elongation at break) were reduced with increasing of the amount of radiation shielding substance mixed in the radiation shielding blocks. However, the reduction of these mechanical properties does not affect the actual utilization since these radiation shielding blocks are normally not subjected to any strong force or pressure. In addition, the SEM images showed the uniform dispersion of radiation shielding substances in the rubber block texture.

1. Introduction
Gamma rays are widely used in medicine; for example, in the area of oncology that malignant and cancerous tumors are treated by the gamma radiation, which this process is called “gamma knife surgery.” [1-3] However, it is known that an exposure to gamma rays could potentially lead to cells becoming cancerous due to the highly penetrable capability of gamma rays. Therefore, radiation protection must be required for a basic safety measure and regulation. Generally, radiation protection aims to protect humans, animals, and the environment from the harmful effects of ionizing radiation, especially a radiation of high energy electromagnetic radiation, like gamma or x-ray. The development of materials with high radiation shielding capability is essentially needed due to the safety regulation for gamma utilization in various applications and activities.

It is known that lead is the most widely used substance for this purpose due to being high Z material and, consequently, high gamma shielding capability. In literature, shielding materials with different lead oxide content and composition have been intensively studied by many research teams [4-7]. Several advantages and disadvantage were shown and discussed. However, due to engineering and economic
aspects, shielding material using lead substance has been commonly utilized. Recently, new materials, like tungsten, have emerged as a potentially suitable material since it is more environmental friendly and safety production [8-9]. In this work, the study of two metal compounds (lead oxide and tungsten oxide) mixed with natural rubber as a shielding block for the attenuation of gamma radiation is investigated with the inclusion of design for practical utilization.

2. Experimental Set up

2.1. Shielding block preparation
A gamma radiation shielding block is developed using natural rubber as a matrix. Different amounts of the metal compound, either lead oxide or tungsten oxide, are mixed with natural rubber in order to fabricate into a shielding block. Three steps are used in this preparation of a shielding block. The first step is a “Mixing” step, which is done by using a two-roll mill and internal mixer (kneader) in order to mix natural rubber, metal compound, and other necessary chemical compounds. The second step is the rubber vulcanization step, which is done by using a sulfur compound. The final step is a “Forming” step, which is done by using a compression moulding machine.

The formula used for each shielding block is shown in Table 1, where the ratio of main chemical ingredients (natural rubber: lead oxide: tungsten oxide) are described for each formula. It is worth noting that the name of each formula is in the first column, which will be used to call them throughout this work. Also, the number appeared in the formula shown in the first column indicates percentage of the shielding metal weight to the total weight. It is noted that the percentage calculation was done based on the percentage of shielding metal to the weight of shielding substances. The content of lead metal in lead oxide is equal to 86.6% while the content of tungsten metal in tungsten oxide is equal to 79.3%. As a result, the 80% tungsten formula (SGT80) is impossible to be prepared.

| Formula | Natural Rubber | Lead oxide | Tungsten oxide |
|---------|----------------|------------|---------------|
| RBk     | 100            | -          | -             |
| SGL5    | 100            | 13         | -             |
| SGL10   | 100            | 25         | -             |
| SGL20   | 100            | 45         | -             |
| SGL40   | 100            | 100        | -             |
| SGL60   | 100            | 270        | -             |
| SGL80   | 100            | 1500       | -             |
| SGT5    | 100            | -          | 14            |
| SGT10   | 100            | -          | 26            |
| SGT20   | 100            | -          | 50            |
| SGT40   | 100            | -          | 122           |
| SGT60   | 100            | -          | 375           |

2.2. Radiation measurements
In this work, there are 3 energy levels of gamma radiation, including 662 keV, 1173 keV, and 1332 keV, used to test the shielding capability. The gamma radiation of 662 keV is obtained from Caesium-137; while the gamma radiation of 1173 keV and 1332 keV is obtained from Cobalt-60. A LaBr3(Ce) detector is used for measuring gamma ray intensity. A simple diagram is shown in Figure 1 below:
2.3. Mechanical property testing
Physical properties of each formula in Table 1 were evaluated in order to ensure the durability of rubber shielding blocks. Hardness and tensile properties (tensile strength and elongation at break) were measured according to the standard methods of ISO 7619-1:2010 and ISO 37:2017, respectively.

2.4. Particle distribution
Scanning electron microscope (SEM) was used to characterize the distribution of shielding substance in rubber block. Beam accelerating voltage of 10 kV was applied in order to obtain the image of 1,000x magnification.

3. Results and discussion

3.1. Gamma attenuation
Figure 2 shows the 662 keV gamma attenuation as a function of a block thickness for a pure lead shielding block and a series of natural rubber shielding blocks those are mixed with different contents of either (a) lead oxide or (b) tungsten oxide. Gamma attenuation was calculated from gamma ray intensity measured by experimental setup in Figure 1 using equation (1), where $I_0$ is incident gamma intensity and $I_x$ is transmitted gamma intensity.

$$\text{Gamma attenuation} = \left( \frac{I_0 - I_x}{I_0} \right) \times 100 \quad (1)$$

It is worth noting that the thickness considered in this work is up to 10 cm. In addition, all radiation measurement is obtained from a LaBr$_3$(Ce) detector, which is set for 180 seconds measurement. It can be seen that the natural blocks mixed with both lead oxide and tungsten oxide can reduce the 662 keV gamma radiations similar to that with a pure lead shielding block. The amount of gamma reduction clearly depends on the thickness of shielding blocks. However, it can be seen that the absorption efficiency of all natural rubber shielding block is not as good as that of a pure lead shielding block, which is not surprising since the natural rubber shielding block’s density is much lower. Therefore, all natural rubber shielding blocks must require a thicker block to be able to shield gamma radiation similar to that for a pure lead shield block. In addition, it can be seen that as the content of either lead oxide or tungsten oxide increases, the shielding capability increases and get closer to the pure lead shielding block.
The gamma attenuation of natural rubber shielding blocks mixed with a (lead oxide) and b (tungsten oxide) for 662 keV gamma radiation obtained from $^{137}$Cs.

Figure 3. Trends of gamma radiation shielding properties of SGL60 and SGT60 rubber block using $^{137}$Cs source.

It can be seen in Figure 3 that the pure lead shielding block at least 5 cm thickness can completely absorb all gamma radiation with the energy of 662 keV. Note that “completely absorb” means above 99% attenuation [10-12]. For a natural rubber shielding block of SGL80 (80% lead), it requires at least 8 cm thickness; while a natural rubber shielding block of SLG60 (60% lead), it is estimated to require at least 16 cm thickness. It is worth noting that a physical property of the SGL80 natural rubber shielding block is not suitable for fabrication since it is highly brittle. This is caused by the insufficient amount of natural rubber in the shielding block. As a result, for overall consideration, a SGL60 natural rubber shielding block is more suitable for fabrication. In the case of a natural shielding block with mixing tungsten, similar trends with the case of shielding block with lead oxide was observed. It was found that the highest attenuation is a natural rubber block with 375 phr of tungsten oxide (SGT60). In addition, it is found that the shielding efficiency of SGL60 natural rubber shielding block is almost similar to that with SGT60 natural rubber shield block.

Figure 4 shows the 1173 keV and 1332 keV gamma attenuation as a function of a block thickness for a pure lead shielding block and a series of natural rubber shielding blocks those are mixed with different contents of either (a) lead oxide or (b) tungsten oxide. It can be seen that the attenuation of 1173 keV and 1332 keV gamma is lower comparing to that for 662 keV gamma. As a result, a thicker block is needed.

As seen from Figure 5, the pure lead shielding block with 9 cm thickness can completely absorb the gamma radiation with the energy of 1173 keV and 1332 keV. For a SGL80 natural rubber shield block, the attenuation is about 98% at 10 cm thickness; while the attenuation is about 84% at 10 cm thickness for a SGL60 natural rubber shielding block. Based on calculation, the thickness to complete absorb 1173 keV and 1332 keV gamma is about 26 cm and 30 cm for SGL60 and SLT60, respectively. It was also found that the shielding efficiency of the SGL60 natural rubber shielding block is almost similar to the SGT60 shielding block.
Figure 4. The gamma attenuation curve for $^{60}$Co (1,173, 1,332 keV) of natural rubber shielding blocks mixed with (a) lead oxide and (b) tungsten oxide

Figure 5. Trends of gamma radiation shielding properties of SGL60 and SGT60 rubber block using $^{60}$Co source

It is worth reminding here that as percentage of shielding substance weight to the total weight increases, a shielding block becomes more brittle. As a result, rubber block forming tends to be more difficult. At some certain point, forming cannot be made, like in the case of SGL80 and SGT80. It is better to develop a shielding block at 60% shielding substance by weight. Thus, it is more suitable to consider SGL60 and SGT60 for further development. However, since both SGL60 and SGT60 shielding block tends to yield similar shielding efficiency, but tungsten is more environmental friendly, the better choice for utilization is a SGT60 shielding block.

Based on the gamma attenuation property, the optimal formula was a shielding block with 60% content of shielding substances (SGL60 and SGT60). Since both SGL60 and SGT60 have the same range of shielding capability, the economical aspect is the main issues of consideration. Due to the higher price of tungsten oxide, natural rubber block with lead oxide (SGL60) is preferred. However, with an environmental concern due to potential release to environment, natural rubber block with tungsten oxide (SGT60) is more suitable. It should be addressed here that inclusion of lead compound has been prohibited in many products. Thus, it is better to avoid using lead in the product.

3.2. Mechanical properties of the rubber shielding block

It is expected that some mechanical properties of a natural rubber block would change as a shielding substance is mixed. In Figure 6, some mechanical properties of the developed natural rubber blocks are shown for each formula developed in this work. The mechanical properties shown here include hardness and tensile strength. It is worth reminding that the number appeared in each formula indicates percentage of the shielding material weight to the total weight. It can be seen from Figure 6 that mixing lead oxide or tungsten oxide tends to lower the hardness of a rubber shielding block. However, at high percentage of shielding material weight to the total weight, it appears that the hardness for SGL80 is higher than that for SGL60. This could be explained that the SGL80 shielding block is almost lead oxide block, where only 6.2% is natural rubber content. Thus, the tensile properties, both tensile strength and elongation at break, obtained is dramatically drop. It appears that for small amount of shielding
substance, the tensile strength slightly increases as amount of lead oxide or tungsten oxide increases. This could be explained that more cross linking is formed by the metal oxide (lead oxide or tungsten oxide) when that shielding substance is mixed [13-14]. However, as a fraction of shielding substance reaches some certain point, the tensile strength is getting lower.

![Figure 6](image1.png)

**Figure 6.** Physical properties of the rubber shielding block, including hardness and tensile strength for a shielding block mixed with (a) lead oxide or (b) tungsten oxide

### 3.3. Particle distribution

The particle distribution is one of the key issues for a gamma shielding. A uniform distribution of metal compound can result in better gamma shielding performance. In this work, the Scanning Electron Microscope (SEM) technique is used to investigate the distribution of metal compounds in rubber matrix. Figure 7 shows the SEM image of the fracture surface on the natural rubber shielding blocks mixed with (a) lead oxide and (b) tungsten oxide. It can be clearly seen that both lead oxide and tungsten oxide are uniform distributed.

![Figure 7](image2.png)

**Figure 7.** SEM images of the fracture surfaces of natural rubber shielding blocks mixed with (a) lead oxide and (b) tungsten oxide with magnification of 1,000
3.4. Designs of rubber shielding blocks

In this work, a new design of rubber shielding block was developed based on two criteria: radiation shielding ability and installation ability. One key requirement for shielding block installation is that there must not be any gap between each shielding block that the radiation can penetrate through, resulting in the reduction of gamma attenuation. A conceptual design of radiation shielding blocks is shown in Fig. 8. It can be seen that there is no gap between between each monoblock due to an interlock between all sides.

![Figure 8](image)

**Figure 8.** A conceptual design of radiation shielding blocks: an interlocked monoblock (a), constructed simple wall (b), and a radiation shielding wall for front view (c) and top view (d).

Figure 9 shows different types of monoblock, which can be used to constructed radiation shielding wall without any gap, resulting in the reduction of gamma attenuation and, consequently, excellent radiation shielding property.

![Figure 9](image)

**Figure 9.** Different types of radiation shielding blocks (a) are shown and how to put them together to construct radiation shielding wall (b).

Figure 10 shows an example of a monoblock of natural rubber shielding block developed and a constructed radiation shielding wall setting up from those monoblocks developed. It can be seen that radiation shielding wall can be setup as a wall and a room corner.

![Figure 10](image)

**Figure 10.** A radiation shielding block (a) and constructed radiation shielding wall (b)
4. Conclusion
A gamma radiation shielding block based on natural rubber mixed with a radiation shielding substance was developed and evaluated at 3 energy levels of gamma radiation (662 keV, 1173 keV and 1332 keV gamma rays). It was found that the formula for a natural rubber shielding block with 80% lead oxide (SGL80) was the best among the shielding blocks with lead oxide developed. It was able to completely shield 662 keV gamma radiation within 8 cm thickness. Meanwhile, a natural rubber shielding block with 60% lead oxide (SGL60), the gamma attenuation test showed that it was estimated to completely shield 662 keV gamma radiation within 16 cm thickness. In comparison with a radiation shielding block with lead oxide, a radiation shielding block with tungsten oxide showed similar characteristics. In addition, it was found that the hardness of these natural rubber shielding blocks tends to be lower with increasing of the amount of radiation shielding substance mixed in the radiation shielding blocks. On the other hand, at low fraction of shielding substance, the tensile strength tends to increase with an increase of shielding substance. However, as a fraction of shielding substance reaches some certain points, the tensile strength is getting lower. The reduction of these mechanical properties does not strongly affect the actual utilization of these shielding blocks since these radiation shielding blocks are normally not subjected to any strong force or pressure. In addition, the uniform dispersion of radiation shielding substances was found in the rubber shielding block. Due to an economical constraint, natural rubber block with lead oxide (SGL60) is preferred. However, with an environmental constraint, natural rubber block with tungsten oxide (SGT60) is more suitable.

Acknowledgements
The authors thank Department of Science Service (DSS) and Thailand Institute of Nuclear Technology (TINT) for financial support.

References
[1] Chilton AB, Shultis JK and Faw RE 1984 Principles of Radiation Shielding (Englewood Cliffs, New Jersey: Prentice-Hall)
[2] Shultis JK and Faw RE 1996 Radiation Shielding (Englewood Cliffs, New Jersey: Prentice-Hall)
[3] Price BT, Horten CC and Spinnery KT 1957 Radiation Shielding (Elmsford, New York: Pergamon)
[4] Harada Y and Nakahara H 1989 INPADOC (Japan: JP patent application) pp 1-146620
[5] El-Khatib A, Kassem M and Ezzat AA 1990 J. Polym. Mater. 7
[6] Matsuda K, Nishikowa H and Harada H 1988 INPADOC (Japan: JP patent application) pp 63-716
[7] Kraus WB, Glasgow MB, Kim MY, Olmeijer DL, Kiefer RL, Orwoll RA and Thibeault SA 1933 Anon-205th ACS National Meeting (Polymer 23) (USA: Washington) p 273
[8] Broder DL, Popkov KK, Trofimov IN and Tsvetkova SA 1975 Distribution of fast neutrons in iron-water shielding Atomic energy 38 44-46.
[9] Celli M, Grazzi F and Zoppi M 2006 A new ceramic material for shielding pulsed neutron scattering instruments Nuclear Instruments and Methods in Physics Research A 565 (Italy) p 861
[10] Santoro RT and Barnes JM 1981 Monte carlo calculations of neutron and gamma – ray energy spectra for fusion reactor shield design: Comparison with experiment Nuclear Science and Engineering (USA: Oak Ridge National Laboratory, Tennessee 37830) 615: 574 – 6084.
[11] Schmidt FAR 1971 Nuclear Engineering Design Vol 15 p 209
[12] Stoddard DH and Hootman HE 1971 The Cf-252 Shielding Guide (USA: Savannah River Laboratory) Report DP-1246
[13] Chokanandsombat Y and Sirisinha C 2013 J. Appl. Polym. Sci. 128 2533-40
[14] Soares BG 2017 Progress in Rubber Nanocomposites (London: Woodhead Publishing) pp 285-318