

Development Of Silicone Rubber-Molybdenum Composites As Lead-Free Thyroid Shielding Materials For X-Ray Radiation Protection

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Abstract– Exposure to radiation of the thyroid gland continues to be a major concern in diagnostic and interventional radiology, due to the high sensitivity of the thyroid gland to radiation. Lead-based (Pb) thyroid shields are effective in reducing radiation exposure, but there are several limitations, including heavy weight, rigidity, and toxicity, which can compromise the comfort and safety of the user. The aim of the present study is to develop a silicone rubber-molybdenum (SR-Mo) composite as a lead-free alternative for thyroid shields which is lighter, more flexible and shows effective radiation absorption performance. The composite was synthesised with silicone rubber matrix with molybdenum filler at concentrations of 0, 1, 3, 5, 7 and 9 wt%. Characterisation was by X-ray attenuation test at 77 kVp. The results show that the mass of SR-Mo composite is significantly reduced compared to conventional Pb-based thyroid shields, and its radiation attenuation is increased, as indicated by the linear attenuation coefficient increase from 0.375 cm^{-1} to 1.259 cm^{-1} , the increase in Radiation Attenuation Ratio (RAR) from 20% to 53%, and the decrease in Half Value Layer (HVL) from 1.846 cm to 0.551 cm at 9 wt% Mo concentration. With increasing Mo content, the radiopacity increased, but the visibility of the underlying objects was not obstructed. The findings indicate that SR-Mo composites could be promising candidates for lightweight, flexible and safer lead-free thyroid shielding materials for radiation protection applications.

Keywords: Radiation Attenuation, Molybdenum, Silicone Rubber, Thyroid Shielding, X-ray.

I. INTRODUCTION

Thyroid shielding is an important part of radiation protection in diagnostic and interventional radiology due to the high sensitivity of the thyroid gland to ionising radiation, especially X-rays [1], [2]. Stochastic effects, such as thyroid cancer, can be induced by even low doses of radiation when received repeatedly, especially in children and young people. Importantly, Saenko and Mitsutake reported that radiation exposure below 100 mGy may still contribute to increased risk of thyroid cancer from both medical and environmental sources [3]. However, there are some limitations in the use of lead (Pb) based thyroid shielding. According to Mohd Ridzwan et al., some radiation workers are reluctant to use thyroid shielding because of the heavy weight, rigidity and reduced mobility of Pb-based materials [4]. Moreover, lead is naturally brittle and is easily cracked or damaged during its use and storage. This has resulted in the development of more flexible and ergonomic shielding materials [4]. In previous studies, Sutanto et al. prepared SR-Pb composite shielding with a thickness of 0.6 cm, which showed better performance than the earlier works of Irdawati et al [5], [6].

On the other hand, lead toxicity is a serious concern because it can bioaccumulate in human body and cause neurological, haematological and renal disorders, even at low exposure levels [7]–[9]. Molybdenum (Mo) is less toxic than lead and is a safer alternative for shielding materials [10]–[12]. Moreover, the high weight of Pb-based shielding has been associated with a higher incidence of musculoskeletal disorders in long-term use [13], [14]. Thus, the development of lead-free shielding materials has become more relevant in the context of modern radiation protection.

Molybdenum (Mo) has been chosen as a filler because of its relatively high density (10.28 g/cm^3) and atomic number ($Z = 42$) that improve the photon interaction within the material. The NIST X-ray mass attenuation coefficient data shows that Mo has good attenuation properties in the low to medium energy X-ray range [15]. Previous studies have also shown that incorporation of Mo and MoO_3 in glass and concrete shielding materials improves the mass attenuation coefficient and reduces the Half Value Layer (HVL) [16]–[18].

However, the application of molybdenum in flexible thyroid shielding materials is still limited. Therefore, this study is focused on developing silicone rubber-molybdenum (SR-Mo) composites as lead-free thyroid shielding materials that are lightweight, flexible, and comfortable to wear while maintaining the performance of radiation attenuation. The use of silicone rubber (SR) as a matrix is expected to improve flexibility and durability preventing cracking or damage during use and storage.

II. THEORETICAL BACKGROUND

1. Thyroid Shielding

Thyroid shielding is used to protect the thyroid gland from exposure to scattered radiation during diagnostic radiographic procedures. Its use is an important part of the implementation of the ALARA principle, since the thyroid is a radiosensitive organ susceptible to ionising radiation [19], [20]. The effectiveness of thyroid shielding in reducing dose was reported by Bawazeer et al., who showed dose reduction up to 73% in panoramic radiography procedures [21]. Lead (Pb) is the most common shielding material because of its high attenuation ability against X-rays. It is usually used with a minimum thickness of 0.35-0.5 mm Pb according to radiation protection standards [22]. However, lead has several drawbacks such as high weight, rigidity and toxicity which have led to the development of other materials that are more flexible and comfortable to use [4], [23]. Molybdenum (Mo) is a promising candidate as alternative filler material due to its relatively high density of 10.28 g/cm^3 and good radiation absorption properties [24], [25]. Therefore, molybdenum-silicone rubber-based composites are expected to be developed to yield thyroid shielding materials which are lighter, more flexible and still effective in X-ray radiation attenuation.

2. Molybdenum

Molybdenum (Mo) is a metal with atomic number 42 and relatively high density of 10.28 g/cm^3 , and thus it is a promising candidate as a radiation shielding filler material. Mo has good chemical stability, corrosion resistance, and effective radiation attenuation ability in the medium energy X-ray and gamma-ray range [25]. The addition of Mo or MoO_3 was found to improve radiation absorption performance by increasing the attenuation coefficient and decreasing the Half Value Layer (HVL) in glass, concrete and polymer-based materials [17], [26], [27].

The addition of 15 wt% MoO_3 into HDPE composites was found to have a higher linear attenuation coefficient (LAC) and a lower HVL than conventional concrete, according to a study by Oto et al. found that the mass attenuation coefficient and effective atomic number (Z_{eff}) increased as the weight fraction of Mo increased, showing better shielding performance against gamma radiation [25].

Molybdenum is less toxic than lead (Pb) and therefore a safer alternative for radiation shielding applications. It is expected that molybdenum in combination with an elastomeric matrix such as silicone rubber will produce shielding materials which are lighter, more flexible and still effective in attenuation of X-ray radiation. Due to these properties, molybdenum has a high potential as an alternative filler in silicone rubber-based composites to develop safer, more flexible and effective lead-free thyroid shielding materials.

3. Silicone Rubber

Silicone rubber is a thermoset elastomer, which is widely used due to its high flexibility, excellent elasticity and stability under extreme temperature and chemical environments [28], [29]. It is also low in toxicity and very biocompatible which makes it suitable for medical applications in which it will be in direct contact with the human body. These properties make silicone rubber a promising matrix material for flexible shielding applications, especially for radiosensitive organs such as the thyroid. Silicone rubber is a thermoset polymer. Thermoset polymer is a polymer that cures to form permanent crosslinked networks, which leads to better thermal and mechanical stability. The material's elastic structure contributes to its flexibility and comfort during use, and also allows for effective interaction with inorganic fillers like heavy metals and metal oxides [30]. Moreover, the density of silicone rubber is relatively low, which contributes to realise the lighter shielding materials than the conventional lead-based shielding.

In this study, silicone rubber was used as the composite matrix, owing to its ability to provide flexible, lightweight and structurally stable materials, as well as to achieve a more homogeneous dispersion of molybdenum filler. Such combined materials are expected to provide thyroid shielding materials with improved user comfort without loss of X-ray radiation attenuation performance [29], [30].

4. Composites

Composites are materials consisting of two or more distinct constituents combined to obtain better mechanical and physical properties than those of the individual components [31], [32]. The composite structure in general consists of a matrix as the continuous phase and a filler as the reinforcing phase. The matrix serves as a binder and distributes the applied load, and the filler enhances the mechanical, thermal and radiation attenuation properties of the material [33].

The Polymer Matrix Composite (PMC) is one of the most widely developed composites and uses a polymer as the matrix. PMCs possess advantages of low weight, flexibility, corrosion resistance and ease of fabrication [34]. In this study, silicone rubber is chosen as the matrix because it is a thermoset polymer with high elasticity, good thermal stability and the ability to interact well with metallic fillers such as molybdenum [30], [35].

The filler used in this work consists of particles of molybdenum, therefore the material is a particle reinforced composite. Advantages of using particulate fillers include more homogeneous reinforcement distribution, easier fabrication processes and lower production cost compared to fibre reinforced composites [36], [37]. Moreover, the addition of heavy metal particles can improve the X-ray attenuation property significantly, apart from the increase in material density. Therefore, the combination of silicone rubber and molybdenum is expected to produce a composite material with flexibility, light weight, ease of processing and effective radiation attenuation appropriate for thyroid shielding applications.

III. METHODOLOGY

1. Fabrication of SR-Mo Composite Materials for Thyroid Radiation Shielding

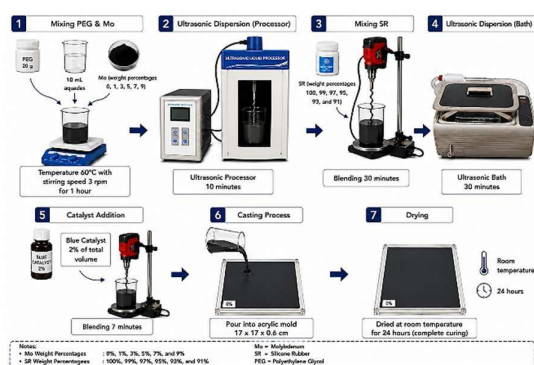


Figure 1. Synthesis Process of SR-Mo Based Thyroid Shielding Composite Samples

The elastomeric matrix was silicone rubber (SR) and the radiation absorbing filler was molybdenum (Mo) to make silicone rubber-molybdenum (SR-Mo) based thyroid shielding composites. The effect of increasing the Mo content on the radiation attenuation performance and the flexibility of the material was investigated by varying the filler concentrations at 0, 1, 3, 5, 7 and 9 wt%. This range of concentrations was chosen to find the optimum composition to balance radiation protection performance and user comfort for wearable shielding applications [38].

The composite synthesis was carried out by a combination of blending and ultrasonic dispersion methods as shown in Fig. 1. This method was chosen because of its efficiency to improve the homogeneity of the distribution of the filler particles in the silicone rubber matrix and to reduce the agglomeration of molybdenum particles which can induce negative effects on the mechanical properties and the radiation absorption performance of the material. Moreover, ultrasonic dispersion promotes interfacial interactions between the filler and matrix to achieve composites with enhanced stability and more uniform structural features [5], [6], [39], [40].

2. Sample Characterization

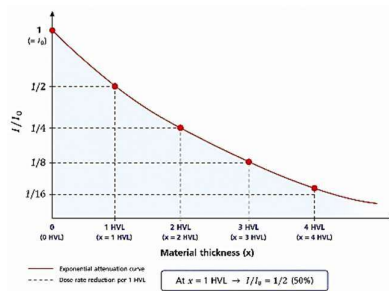


Figure 2. Graph of radiation attenuation in materials, adapted from [41]

The characterisation of silicone rubber-molybdenum (SR-Mo) composite materials was performed to evaluate their suitability as lead free thyroid shielding by assessing their physical properties, radiation absorption capacity and mechanical performance. The tests performed were: mass measurement, X-ray radiation attenuation measurement and mechanical measurement in the form of elongation at break. The mass measurements were carried out to assess the effect of inclusion of molybdenum filler on the physical characteristics of the material and to compare the mass of the SR-Mo composites with the conventional lead-based thyroid shielding. This parameter is of great importance because the material weight directly influences the comfort of the user during long radiological procedures [13].

Radiation absorption tests were conducted with an X-ray radiography system. The attenuation capacity of the material was studied using the Lambert-Beer law which describes the exponential decrease of the intensity of radiation through a material of a certain thickness, as expressed in equation (1) [15]:

$$I = I_0 e^{-\mu x} \quad (1)$$

where I_0 is the initial radiation intensity, I is the transmitted radiation intensity, μ is the linear attenuation coefficient (cm^{-1}) and x is the material thickness (cm). The absorption parameters analysed based on this equation are the linear attenuation coefficient, Half Value Layer (HVL), and Radiation Attenuation Ratio (RAR). In addition, the qualitative analysis of radiographic images was performed for the evaluation of radiopacity changes caused by the incorporation of molybdenum filler.

The mechanical characterisation was performed using a tensile test in order to determine the elongation at break and the force. The test is to assess the ductility of the material and effect of filler addition on the deformation behaviour of SR-Mo composites as flexible thyroid shielding materials [42]–[44].

IV. RESULTS ANALYSIS

1. Mass Analysis of SR-Mo Based Thyroid Shielding Composite Material Samples

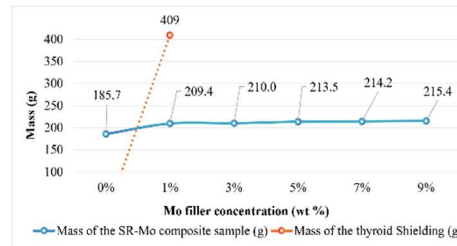


Figure 3. Comparison Chart of Sample Mass and Conventional Thyroid Shielding

As seen in Figure 3, the mass of the SR-Mo composite increases gradually with increasing concentration of the molybdenum filler. The sample mass increases from 185.7 g at 0 wt% to 215.4 g at 9 wt%. The increase is attributed to the inclusion of molybdenum particles which are high density and thus increase the overall composite density. However, the increase is still relatively small, and tends to saturate above 1 wt% indicating that the addition of Mo filler does not substantially change the total mass of the composite material. SR-Mo composites demonstrate significantly lower mass values in comparison to the conventional lead based thyroid shielding (mass of ~409 g). The composite mass at the highest concentration (9 wt%) is only about 52.7% of the conventional shielding mass. These results show the potential of SR-Mo composites to develop much lighter shielding materials, thus improving the comfort of the user particularly for the radiology personnel who wear shielding for a long time. The reduced mass of the shielding also helps to reduce the static load on the neck and shoulders, a well known drawback of the lead-based shielding. Thus, the SR-Mo composite improves the ability of radiation absorption with the increase in molybdenum content, but preserves its lightweight nature which is required for ergonomic performance and occupational safety in thyroid shielding applications.

2. Analysis of X-Ray Radiation Absorption Data in SR-Mo-Based Thyroid Shielding Composite Samples

X-ray radiation absorption testing was carried out to evaluate the effectiveness of silicone rubber-molybdenum (SR-Mo) composite materials in attenuating the intensity of the transmitted radiation. This test was carried out to investigate the effect of molybdenum filler addition on the improvement of X-ray attenuation capability of silicone rubber-based thyroid shielding materials.

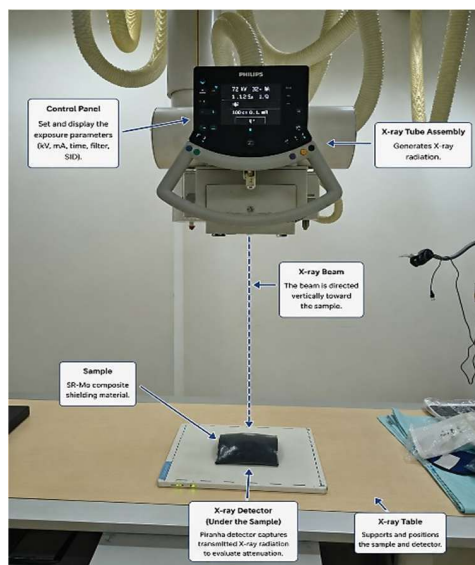


Figure 4. X-ray Radiation Absorption Measurement Setup for SR-Mo based Thyroid Shielding Composite Samples

In the experimental setup shown in Figure 4, the composite sample was placed on a digital detector and irradiated using a diagnostic X-ray system, while the radiation dose was measured using a Piranha detector as a dosimetry instrument. The interaction between X-ray radiation and the composite material was evaluated through changes in radiographic image intensity and the measured radiation dose after transmission through the sample. The data obtained from the general radiography system are presented in Table 1.

Table 1. X-ray Radiation Absorption Data of SR-Mo-Based Thyroid Shielding Composite Samples

No	Sample (% wt Mo)	D_1/D_0	μ (cm^{-1})	μ_m (cm^2/g)	HVL (cm)	RAR (%)
1.	0	0.798	0.375	0.351	1.846	20
2.	1	0.739	0.503	0.416	1.378	26
3.	3	0.681	0.639	0.528	1.084	31
4.	5	0.640	0.743	0.603	0.933	36
5.	7	0.523	1.081	0.875	0.641	48
6.	9	0.470	1.259	1.014	0.551	53
7.	Shielding Tiroid 0.5 mm Pb	0.026	-	-	-	97

Table 1 shows the results of the radiographic test, which reveal that the radiation absorption capacity of the silicone rubber-Molybdenum (SR-Mo) composites is significantly affected by the increase in the concentration of molybdenum (Mo) filler. As the Mo concentrations increase, the radiographs brightness (grayscale intensity) continuously increase, as shown in Figure 5.

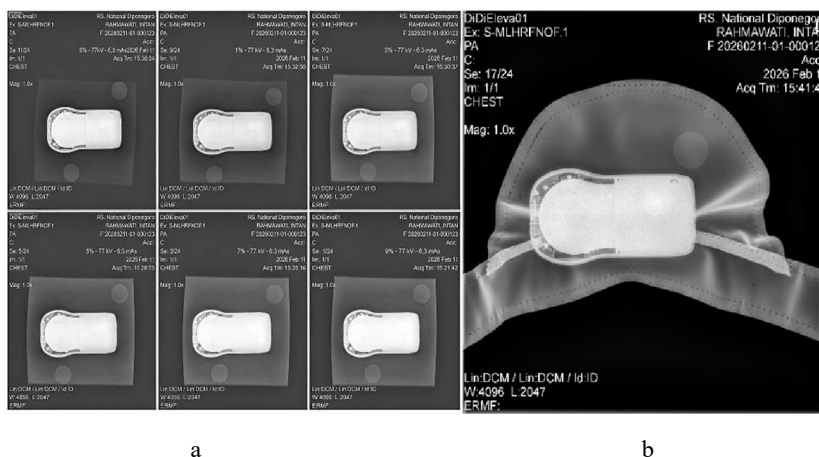


Figure 5. Radiographic Images of (a) SR-Mo-Based Thyroid Shielding Composite Samples and (b) 0.5 mm Pb Thyroid Shielding

As shown in Figure 5(a), the 0 wt% sample is the darkest, which means that most of the radiation can still pass through the material. In contrast, the increase in radiopacity of 7 wt% and 9 wt% samples is more considerable. This means that the addition of molybdenum improves the ability of the material to decrease the transmission of X-rays. Moreover, the SR-Mo composites have a lower radiopacity than the traditional Pb-based thyroid shields, allowing the radiographic objects to be more clearly visualized in the images, as shown in Figure 5(b). This feature might enhance the diagnostic accuracy as the anatomical details are not totally obscured by the shielding material.

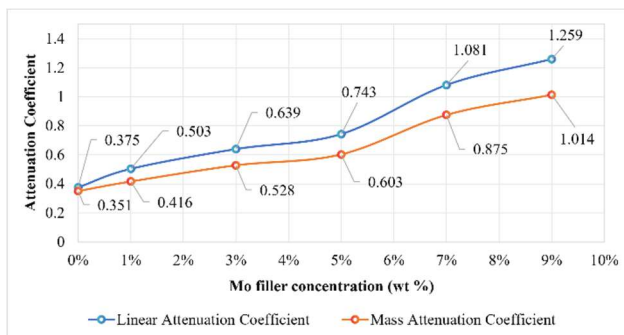


Figure 6. Effect of Mo Filler Concentration on the Attenuation Coefficient

The enhancement in shielding performance is supported by the increase in the linear attenuation coefficient (μ) from 0.375 cm^{-1} for the 0 wt% sample to 1.259 cm^{-1} at 9 wt%. Similarly, the mass attenuation coefficient (μ_m) increases from $0.351 \text{ cm}^2/\text{g}$ to $1.014 \text{ cm}^2/\text{g}$. The increase in both parameters indicates a higher probability of X-ray photon interactions within the material, resulting from the increased number of Mo atoms incorporated into the SR matrix. In addition to the relatively high atomic number of molybdenum, the increase in material density from 1.071 g/cm^3 to 1.242 g/cm^3 further contributes to the improved radiation absorption efficiency.

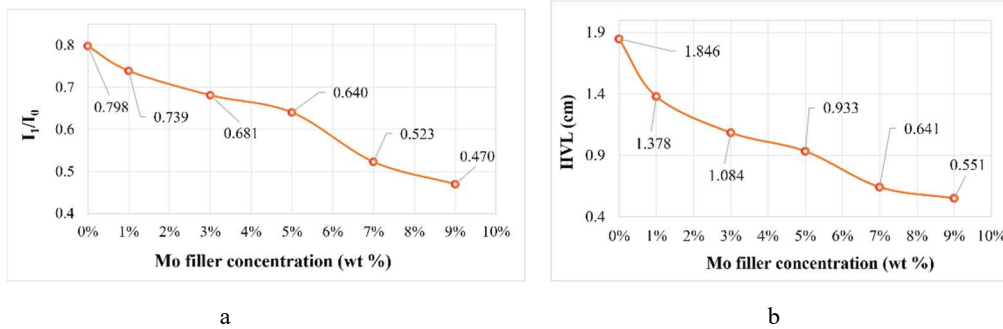


Figure 7. Effect of Molybdenum Filler Concentration on Dose Rate and HVL: (a) Dose Rate and (b) Half Value Layer

The HVL value decreases from 1,846 cm to 0,551 cm with the increasing of filler concentration. This reduction demonstrates that a smaller material thickness is required for the attenuation of the radiation intensity by 50% which improves the overall shielding efficiency. A similar trend is observed for the ratio of dose rates I/I_0 which decreases from 0,798 to 0,470, which indicates that a smaller fraction of radiation passes through the material.

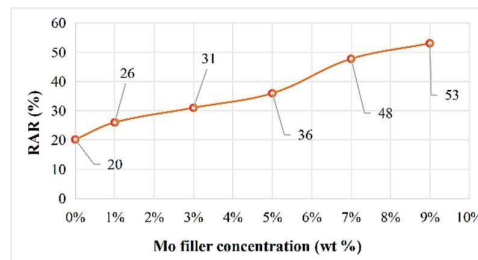


Figure 8. Effect of Mo Filler Concentration on Radiation Attenuation Ratio (RAR)

This is also evidenced by the Radiation Attenuation Ratio (RAR) that increases from 20% in the 0 wt% sample to 53% at 9 wt% showing the efficiency of radiation absorption. The improvement suggests that the addition of Mo filler increases the absorption ability by about 2,5 times compared with the pure silicone rubber. However, this performance is still lower compared to commercially available 0,5 mm Pb based thyroid shielding which appears significantly more radiopaque in radiographic images and is able to almost completely block the radiation transmission.

V. DISCUSSION

In this work, we show that the incorporation of molybdenum (Mo) filler to the silicone rubber (SR) matrix significantly improves the ability of the material to absorb X-ray radiation without much reduction of its flexibility. The increase of Mo concentration results in increase of linear attenuation coefficient and mass attenuation coefficient, and decrease of half value layer (HVL) and transmitted radiation dose rate. These results suggest that the presence of Mo atoms in the SR matrix enhances the probability of X-ray photons interacting with the composite, thereby improving its ability in radiation attenuation.

The improved shielding performance of the SR-Mo composite is in agreement with various previous studies. Almuqrin et al. showed that the addition of MoO_3 to HDPE composites increased the value of the linear attenuation coefficient and decreased the value of the HVL in comparison with conventional materials [17]. Oto et al., reported similar results, where increasing the weight fraction of Mo leads to higher values of mass attenuation coefficients and effective atomic numbers (Z_{eff}) for brick-based shielding materials [25]. The consistency of these results confirms the role of molybdenum as an effective filler for enhancing the radiation absorption properties of various matrix types, including silicone rubber.

However, the SR-Mo composite still has a lower radiation attenuation capability than the commercial Pb-based thyroid shields with the equivalent thickness of 0,5 mm Pb. This discrepancy is mainly attributed to the atomic number of Pb ($Z=82$) much higher than Mo ($Z=42$), which leads to a higher probability of photoelectric interactions in the diagnostic X-ray energy range. But molybdenum does have a number of significant advantages, especially when it comes to safety and ergonomics. Mo is less toxic than toxic Pb and results in shielding materials with a significantly lighter mass. This is especially important because it has been reported that the long-term use of Pb-based shielding makes radiology staff uncomfortable, physically tired, and suffering from musculoskeletal disorders due to the weight of the material [13], [14].

The SR-Mo composite also has better mechanical properties than some heavy-metal-based shielding materials besides the attenuation performance. Demirel and Yücel observed that the shielding effectiveness increased with the increase in the bismuth oxide filler content but was accompanied by an increase in the material stiffness. In contrast, the addition of Mo in this study enhanced the radiation absorption capacity without a significant loss of flexibility. This advantage is also complemented by the mass of the SR-Mo composite, ranging from 185,7 to 215,4 g, significantly lower than the ~409 g mass of conventional Pb-based thyroid shields. These results show that silicone rubber is a good matrix to make protective materials that are light, flexible and still have good radiation attenuation capabilities. Hence, the present study aims at enhancing the shielding performance and, more importantly, to address the ergonomic and comfort issues of the user which have been neglected in many previous studies.

In conclusion, the results of this study suggest the SR-Mo composite is a promising candidate as a lead free thyroid shielding material. The attenuation efficiency of this composite is not yet fully comparable to that of lead-based shields, but it represents a good compromise between radiation absorption ability, mechanical flexibility, light weight and material safety. Such a combination of properties makes the SR-Mo composite as a potential candidate for further implementation as a wearable radiation shield for radiology personnel and other diagnostic radiation protection applications.

VI. CONCLUSION

This research was successful in the development of silicone rubber-molybdenum (SR-Mo) composites as low weight, flexible and suitable lead-free thyroid shielding materials for contemporary radiation protection application. The addition of molybdenum filler improves the X-ray radiation absorption property as the linear attenuation coefficient and Radiation Attenuation Ratio (RAR) increase and the Half Value Layer (HVL) decreases with the increasing of Mo concentration. The SR-Mo composites provide not only improved shielding performance, but also reduced mass relative to traditional Pb-based thyroid shielding, which can improve comfort for the user and reduce ergonomic load for radiology personnel. Overall, results indicate that SR-Mo composites are a promising alternative to traditional lead-based thyroid shielding, with a safer, lighter and more flexible solution for radiation protection applications.

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