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GAMMA RAY RADIATION RESEARCH IN BUILDING MATERIALS

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ABSTRACT

Building materials used in the construction industry, such as marble, granite, ceramics, travertine, and cimstone, are of crustal origin and contain natural radiation. While these materials act as a source of natural radiation inside the building, they actually serve as a shield by blocking external radiation. Therefore, these building materials can also be considered as radiation-shielding materials. In the first part of this study, activity concentrations of radioactive isotopes in materials used in construction industry were determined via gamma spectrometry. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radioactive isotopes in marble samples were measured as 10.5,14.3 and 584.6 Bqkg^{-1}, respectively. In granite samples, the activity concentrations of radioactive isotopes were found to be 72.3, 63.5, 1024.7 Bqkg^{-1}, respectively. In ceramic samples , the activity concentrations of radioactive isotopes were found to be 39.8, 68.5, 1338.7 Bqkg^{-1}, respectively. In the experiment performed using travertine samples, the activity concentrations of radioactive isotopes were calculated to be 5.9, 9.9 and 1338.7 Bqkg^{-1}, respectively. The activity concentrations of radioactive isotopes in cimstone samples were measured as 6.78, 26.9 and 649.7 Bqkg^{-1}, respectively. Radium equivalent activity caused by these activities was 21.6, 101.46, 32.29, 91.7, and 47.51 Bqkg^{-1} for marble, granite, travertine, ceramic, and cimstone, respectively. The second part of the study focused on the use of building materials as shielding materials. Mass attenuation coefficient μ/ρ (cm^2/gr), and half value layer (HVL) values, representing the radiation interaction with the material used for shielding, were examined individually for marble, granite, travertine, ceramic, and cimstone samples. The attenuation coefficients were measured for gamma energies from ^{152}Eu, ^{137}Cs, and ^{60}Co sources. The obtained experimental data were compared with the theoretical values calculated using WinXCom software, and the results were found to be compatible.

KEYWORDS:
Natural radioactivity, radiation shielding, mass attenuation coefficient, building material, XRF

INTRODUCTION

Radiation is a phenomenon that exists in the earth's crust, air, and water. People are continuously exposed to terrestrial and cosmic radiations that constitute natural radiation sources as a result of internal and external irradiations, depending on the geological structure of the region where they live and the quality of their lives. Because people spend most of their time in closed buildings such as homes and workplaces, it is important to know the radiation dose they are exposed to and determine the level of radiation in buildings. Studies show that the amount of radiation inside a building is relatively small compared to the amount of radiation outside a building, and the dose rates of radiation absorbed inside the building particularly depend on the location of the building and the type of construction material used. In buildings with a stone, brick, or concrete exterior, gamma rays from external sources are greatly absorbed by the walls, while the dose rate absorbed inside the buildings depends on the activity concentration of natural radionuclides in the construction materials used [1]. In the literature, there are studies for the investigation of radioactivity in the construction industry [2-16]. With the rapid development of nuclear technology worldwide, nuclear activities have increased, causing the development of anti-radiation measures to be inevitable. However, in medicine, it is impossible to completely abandon radiation, which is used in diagnostics and treatment and provides benefit in this aspect. Therefore, in the field of medicine, methods have been developed to reduce the harmful effects of radiation while utilizing its beneficial effects. The most effective way of protection from the harmful effects of gamma rays emitted by radiation sources is shielding. The material that is used in shields to reduce the dose of radiation is called shield material; the choice of the material depends on the type of radiation. The greater the density of the shield material, the greater its radiation-shielding effect. There are several studies on the shielding properties of building materials [17-23].

MATERIALS AND METHODS

Radioactivity measurements. Forty-four building materials procured from different factories and enterprises in Turkey were used in the experiments. ORTEC's 905-4 (3" × 3") model NaI (Tl) scintillation detector-based gamma spectrometer system was used for measurements. The resolution of the system is 2% at 0.5 MeV and 1.3% at 2 MeV (for 1 µCi ^{137}Cs at a distance of 10 cm). The thickness of the detector's Al container is 0.5 mm. Standard RGU-1 uranium, RGTh-1 thorium, RGK-1 potassium sources, and point ^{60}Co source with known gamma energies, as referenced were used for calibration of the system. The activity concentration of ^{226}Ra radioactive isotope was calculated by selecting photo peak of ^{226}Ra (186 keV), ^{214}Pb (295 and 351 keV), and ^{214}Bi (609 keV) energy. The activity concentration of ^{232}Th radioactive isotope was calculated using photo peak of ^{228}Ac (911 keV) and ^{208}Tl (583 keV) gamma energies. The activity of ^{40}K radioactive isotope was calculated using photo peak of 1460 keV energy specific to this radioactive isotope. The activity concentration values obtained as a result of measurements are shown in Table 1.

TABLE 1
Activity concentration values measured in samples

Sample ID		Activity concentration in (Bqkg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
Marble	Range	1.17-28.64	4.7-30.7	492.1-700.4
	Average	10.53	14.3	584.5
Granite	Range	4.9-290.9	6.5-128.5	698.6-1886.0
	Average	72.3	63.5	1024.7
Ceramic	Range	6.7-93.9	43.1-133.8	1154.3-1793.9
	Average	39.8	68.2	1383.7
Travertine	Range	3.1-8.0	5.5-26.8	317.2-812.2
	Average	5.9	9.9	410.6
Cimstone	Range	3.9-57.0	6.9-65.0	495.9-1088.1
	Average	6.7	26.9	649.7

Radium equivalent activity. Radium equivalent activity (Ra$_{eq}$) is a parameter used for evaluating radioactivity caused by radium, thorium, and potassium. The radium equivalent activity was calculated using Eq.(1) [24-25].

$$Ra_{eq}=C_{Ra}+1,423C_{Th}+0.077C_K \qquad (1)$$

where C_{Ra}, C_{Th}, and C_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K radioactive isotopes in Bqkg^{-1}.

External and internal hazard index. To determine gamma radiation hazard, external (Iɣ) and internal hazard indices were calculated using the activity concentrations of ^{226}Ra in uranium decay series, and the activity concentrations of ^{232}Th and ^{40}K in thorium decay series. For building materials, external hazard index (Iɣ) was calculated using Eq.(2)

$$I\gamma = \frac{CTh}{200} + \frac{CRa}{300} + \frac{CK}{3000} \qquad (2)$$

To evaluate the additional alpha radiation caused by inhalation of ^{222}Rn gas, internal hazard index (Iα) was calculated using Eq. (3) [26].

$$I\alpha = \frac{CRa}{200} \qquad (3)$$

According to European Commission (EC) , the external hazard index (Iɣ) and the internal hazard index (Iα) used to evaluate the amount of radiation being exposed to due to the inhalation of radon gas must be less than 1 in order to meet the safety requirements of building materials. This can be achieved if the activity of ^{226}Ra is less than 200 Bqkg^{-1} [27]. The external and internal hazard indices and equivalent radium activity calculated using measured activities are shown in Table 2.

TABLE 2
Calculated radiological parameters

Sample	Ra$_{eq}$ (Bqkg^{-1})	Activity index	
		I ɣ	Iα
Marble	75.93	0.26	0.05
Granite	240.04	0.85	0.36
Ceramic	241.89	0.93	0.19
Travertine	51.56	0.18	0.02
Cimstone	94.95	0.36	0.03

Gamma transmission technique. Using gamma transmission technique, the radiation attenuation effect of materials can be measured experimentally. This technique is based on the interaction of gamma rays with the material through which they pass [28]. Gamma rays lose part of their energy when they interact with materials; the linear attenuation coefficient (μ) of the material against gamma ray, mass attenuation coefficient (μ/ρ), and minimum thickness value (HVL) that the material must have to reduce the intensity of radiation by half are calculated and the count value is obtained as a function of energy and the density of the material. The radiation permeability of the material varies depending on its chemical content. The chemical content of ceramic, granite, and marble used in this study was determined using XRF technique and as shown in Table 3. The chemical content of cimstone and travertine was taken from the articles mentioned in Table 3. In this part of the study, the radiation attenuation effect of marble, granite, ceramics, travertine, and cimstone was calculated using gamma energies of 121.78, 244.7, 344.28, 661.64, 778.9, 964.08, 1085.9, 1173.2, 1332.5, and 1408 keV obtained from Eu-152, Cs-137, and Co-60 point standard sources. The experimental results obtained were compared with the theoretical values obtained using WinXCom software [29]. Experimental apparatus based on gamma transmission technique is described in Figure 1, so that there would be a distance of 70 mm between the detector and the sample, and a distance of 175 mm between the standard point gamma ray source and the sample. To obtain a thin beam in apparatus, the collimator was designed to have a diameter 0f 30 mm. The materials were placed between the gamma ray source and the NaI(Tl) detector.

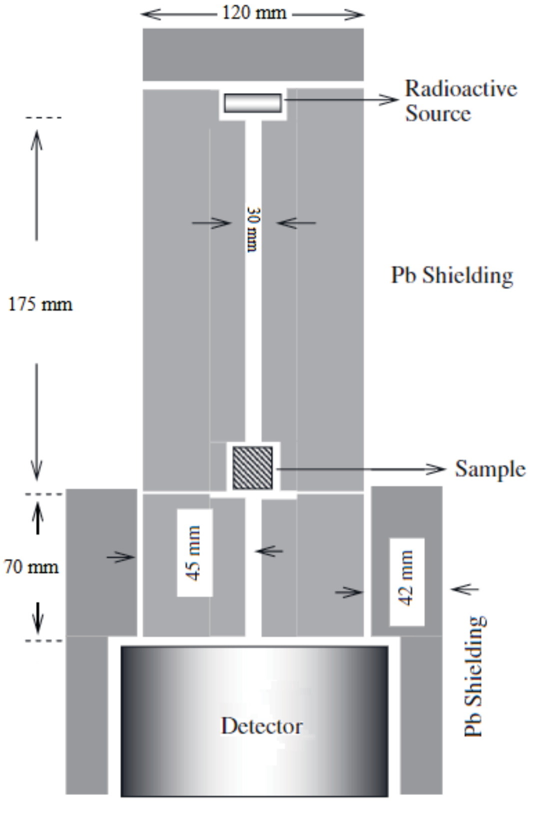

FIGURE 1
Schematic view of the experimental setup

RESULT AND DISCUSSION

Radiological evaluations. In the experiments performed using eight different marble samples, it was found that the activity concentration of ^{226}Ra ranged from 1.17 to 28.64 Bqkg^{-1}, while the average activity concentration of ^{226}Ra was 10.53 Bqkg^{-1}. In the same experiment, it was determined that the average activity concentration of ^{232}Th was 14.37 Bqkg^{-1}, while the average activity concentration of ^{40}K was 584.54 Bqkg^{-1}. In the experiments performed using ten different granite samples, it was determined that the average activity concentration of ^{226}Ra ranged between 4.91–290.08 Bqkg^{-1}, and the average activity concentration was 72.39 Bqkg^{-1}. It was found that the average activity concentration of ^{232}Th was 63.55 Bqkg^{-1}, ranging from 6.56 to 128.5 Bqkg^{-1}. In the granite samples, the average activity concentration of ^{40}K radioactive isotope was found to be 1024.73 Bqkg^{-1}.In the experiment performed using seven different ceramic samples, the average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K radioactive isotopes were found to be 39.80 Bqkg^{-1}, 68.20 Bqkg^{-1}, and 1383.70 Bqkg^{-1}, respectively. In the experiment performed using nine different travertine samples, the average activity concentrations of ^{226}Ra and ^{232}Th radioactive trackballs were determined as 5.96 Bqkg^{-1} and 9.93 Bqkg^{-1}, respectively. In the same samples, the average activity concentration of ^{40}K radioactive isotope was found to be 410.67 Bqkg^{-1}.In the experiment performed using ten different cimstone samples, the average activity concentration of ^{226}Ra and ^{232}Th radioactive isotopes were found to be 6.78 Bqkg^{-1} and 26.9 Bqkg^{-1}, respectively, and the average activity concentration of ^{40}K radioisotope was found 649.77 Bqkg^{-1}. Based on UNSCEAR 2000 report values, it can be said that the average activity concentrations of ^{226}Ra and ^{232}Th radioactive isotopes detected for marble, ceramics, travertine, and cimstone were considerably smaller than the world average [30]. The granite measurements indicated that the average activity concentration of ^{226}Ra radioactive isotope was twice the world average, while the average activity concentration of ^{232}Th radioactive isotope was 1.5 times the world average. The measurements for ceramics and travertine indicated that the average activity concentration of ^{40}K radioactive isotope was below the world average. In the measurements for marble and cimstone, the average activity concentration of ^{40}K radioactive isotope was slightly above the world average, and the average concentration of activity of ^{40}K radioactive isotope was 2.5 times the world average. Radium equivalent activity caused by these activities was 21.6, 101.46, 32.29, 91.7, and 47.51 Bqkg^{-1} for marble, granite, travertine, ceramic, and cimstone, respectively. Radium equivalent activity value is 370 Bqkg^{-1}, as reported by the Nuclear Energy Agency (NEA) [31] . As it is seen in Table 2, the external hazard index (Iɣ) and internal hazard index (Iα) indices are less than 1. In many countries, there are studies aimed at determining natural radioactivity levels in building materials. Table 4 summarizes the studies conducted in some countries. Comparing with the results of studies in different countries, it can be said that the average radioactivity concentration for many materials used in the construction industry in Turkey is lower than the limit values.

TABLE 3
Chemical contents (%by weight) of building materials

	Na_2O	MgO	$A_{l2}O_3$	SiO_2	P_2O_5	SO_3	Cl	K_2O	CaO	TiO_2	Fe_2O_3	Loss H_2O	Total
Ceramic T1200011390	2.4	0.85	18.14	71.44	-	-	-	3.62	1.02	0.55	-	1.75	0.2
Cimstone [32]	0,15	0.40	0.02	96.51	0.06	0.19	0.01	0.10	1.48	0.93	-	0.09	-
Granite T120001139	3.26	0.45	13.24	73.32	-	-	-	5.26	1.21	0.316	-	2.59	0.34
Marble T120001139	0.01	3.85	0.01	0.01	-	-	-	0.01	51.72	0.01	-	0.196	42.48
Travertine [33]	0.2	0.2	0.1	0.4	0.0	-	-	0.1	52.6	0.0	0.0	0.2	44.94

TABLE 4
Similar studies conducted in different countries

Sample	Country	Activity concentration in $Bqkg^{-1}$			Ra_{eq} ($Bqkg^{-1}$)	References
		^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	
Marble	India	9.25	61.63	1366.85	202.63	[5]
	Saudi Arabia	5.8	2.3	33.7	11.6	[6]
	Turkey	5.4	4.9	49.7	54.88	[7]
	Tunis	33.24	8.01	116.98	-	[15]
	This paper	10.5	14.3	584.61	73	
Granite	India	6.17	4.62	39.17	23.49	[5]
	Egyptian	32.46	47.46	1314.82	-	[8]
	Saudi Arabia	54.5	43.4	667.7	168.7	[6]
	This paper	72.3	63.5	1024.7	240.0	
Cermaic	Egypt	51.12	40.52	682.6	-	[8]
	Saudi Arabia	47.18	80.7	590.2	207.2	[6]
	Turkey,	31±5	28±12	358±31	81	[10]
	Algerian	136	52.4	491.33	252.31	[9]
	Italy	50±2	59±2	520±22	-	[16]
	This paper	39.8	68.2	1338.7	242.8	
Travertine	Turkey	0.8	0.9	4.1	-	[7]
	Italy	5.4	13.5	65.6	75.2	[11]
	This paper	5.9	9.9	410.6	51.6	
Cimstone	Turkey	2.4	4.6	47.8	-	[7]
	This paper	6.78	26.9	649.7	99.9	

Investigation of mass attenuation coefficients. In this part of the study, gamma spectrometer system was used for attenuation coefficients measurements. The sample was placed between the gamma source and the detector. The radiation attenuation of ceramic, cimstone, granite, marble, and travertine samples produced in Turkey, were investigated. These materials are widely used as construction materials in public and commercial buildings. In this study, ceramic, cimstone, granite, marble, and travertine tiles measured 3–4 cm × 3–4 cm × 0.5–2 cm and their masses varied from 15 to 50 g. Content analysis of ceramic, granite, and marble was done using XRF technique with codes T1200011390, T1200011392, and T1200011391, respectively. The chemical contents of cimstone and travertine were taken from the studies indicated in Table 3 [32-33]. The mean density of ceramic, cimstone, granite, marble, travertine respectively is 2.36 $grcm^{-3}$, 2.15 $grcm^{-3}$, 2.6 $grcm^{-3}$, 2.78 $grcm^{-3}$, 2.44 $grcm^{-3}$.In this part of study, we investigate the gamma attenuation of building materials. Comparison of mass attenuation coefficients of gamma ray sources is as shown in Figures 2,3,4,5 and 6, respectively. The solid line shows the calculated results with WinXCom; the points shows experimental results obtained in this study. In most cases, experimental and theoretical calculations were in good agreement. The differences between the mass attenuation coefficients are thought to be related to the chemical composition of the material. It can be said that the mass attenuation coefficient obtained experimentally decreases with increasing energy. With this study, experimental calculations showed that building materials can be used as an alternative material against radiation, especially at low energies.

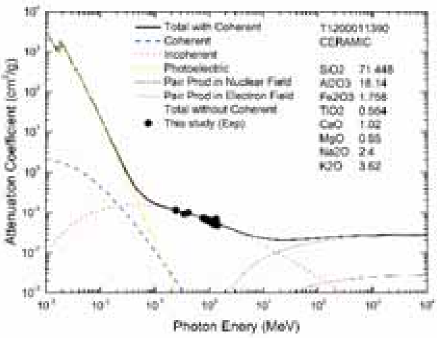

FIGURE 2
Comparison of mass attenuation coefficients of gamma ray sources of ceramic samples

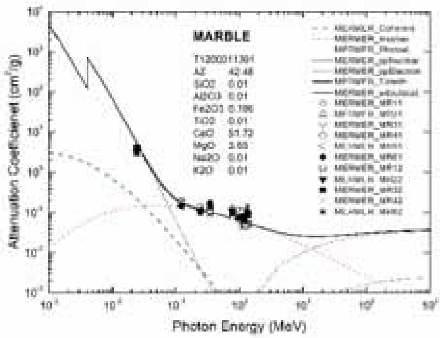

FIGURE 5
Comparison of mass attenuation coefficients of gamma ray sources of marble samples

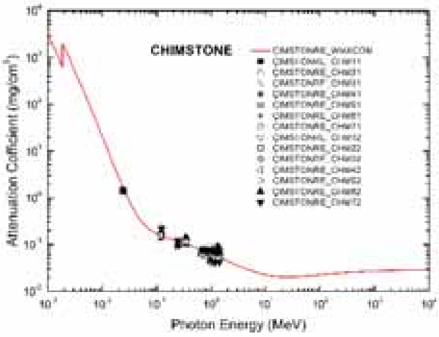

FIGURE 3
Comparison of mass attenuation coefficients of gamma ray sources of cimstone samples

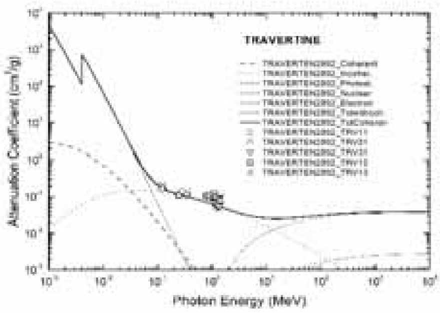

FIGURE 6
Comparison of mass attenuation coefficients of gamma ray sources of travertine samples

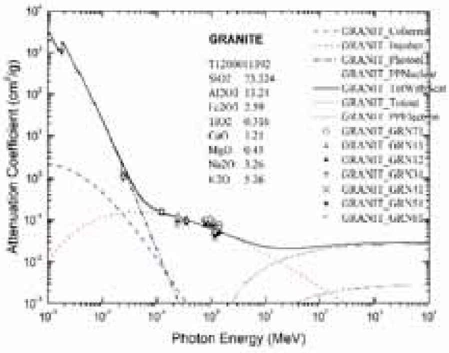

FIGURE 4
Comparison of mass attenuation coefficients of gamma ray sources of granite samples

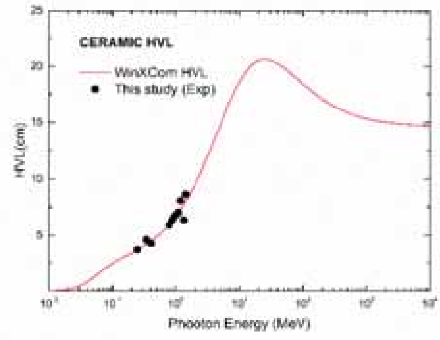

FIGURE 7
The plot of half value layers of ceramic.

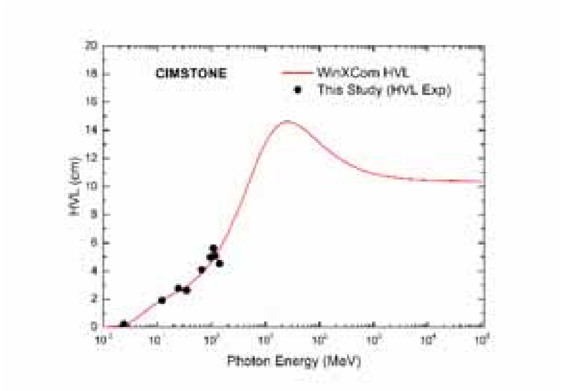

FIGURE 8
The plot of half value layers of çimstone.

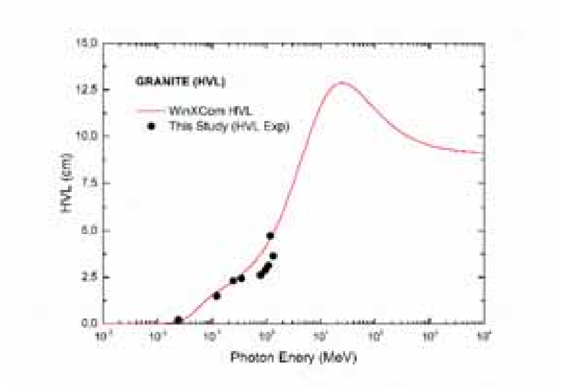

FIGURE 9
The plot of half value layers of granite

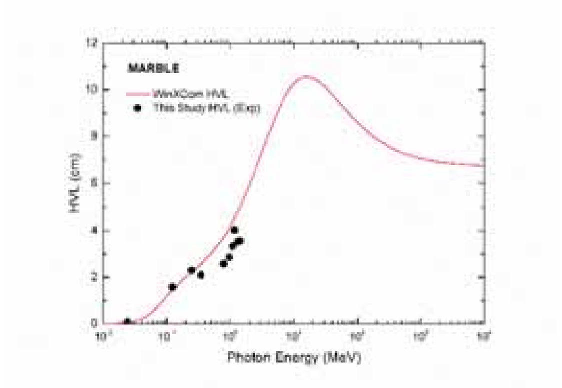

FIGURE 10
The plot of half value layers of marble

The measured and calculated half value layers (HVL) results for building materials are given in Figures 7,8,9,10 and 11, respectively. It can be seen that the experimental and theoretical values fit well as shown in these figures.

CONCLUSIONS

The activity concentrations of radionuclides building materials used in construction industry were determined using gamma spectrometric method. Radium equivalent activity indices as well as external and internal activity indices, which contribute to the protection of human health in the long term, were calculated based on the determined activity results. The results of the experiment were evaluated based on the standard data adopted by the UNSCEAR and EC commissions. The results of the study in different countries suggest that the activity and radiological parameter values in most building materials were lower than the limit values. In this study, the radium equivalent activity calculated for selected building materials was found less than the limit value of 370 Bqkg^{-1}. It was also seen that the materials used in the experiment met the requirement for the hazard index to be less than 1. On the contrary, it was seen that granite materials contained potassium activity above the limit value, which was because of its geological structure.

Experimental and theoretical studies were conducted to have an idea about the radiation permeability of building materials. Obtained results proved that the radiation permeability of materials exposed to different gamma energies shows similar changes.

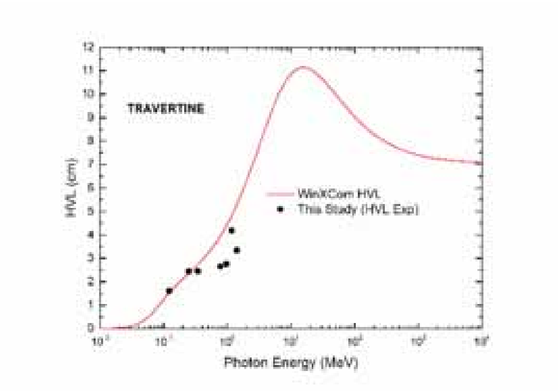

FIGURE 11
The plot of half value layers of travertine

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