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Asphaltene or Polyvinylchloride Waste Blended with Cement to Produce a Sustainable Material Used in Nuclear Safety

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Abstract: The current research uses sustainable methods to preserve the environment, such as exploiting municipal or industrial waste that may harm the environment. The wreckage of polyvinyl chloride (PVC) pipes and asphaltene are used as additives to cement to improve its mechanical properties, while stabilizing the radioactive waste resulting from the peaceful uses of nuclear materials, or enhancing its radiation shielding efficiency. New composites of Portland cement with ground PVC or asphaltene up to 50% are investigated. Fast neutron removal cross-section (Σ_R) and gamma shielding parameters, such as mass attenuation coefficient (MAC), half-value layer (HVL), effective atomic number (Z_{eff}), and exposure build-up factor (EBF) at wide energy range and thickness, are determined. The compressive strength and apparent porosity of the examined composites are examined to test the durability of the prepared composites as stabilizers for radioactive waste. The obtained results show that the bulk density of hardened cementitious composites was slightly increased by increasing the additive amount of PVC or asphaltene. The compressive strength of cement composites reached more than 4.5 MP at 50 wt.% PVC and 8.8 MPa at 50 wt.% asphaltene. These values are significantly higher than those recommended by the US Nuclear Regulatory Commission (3.4 MPa). Additionally, the obtained results demonstrate that although the gamma MAC is slightly decreased by adding asphaltene or PVC, the neutron removal cross-section was highly increased, reaching 171% in the case of 50 wt.% asphaltene and 304% in the case of 50 wt.% PVC. We can conclude that cement composites with PVC or asphaltene have optimized radiation shielding properties and can stabilize radioactive waste.

Keywords: asphaltene; cement; polyvinylchloride; radiation shielding; waste immobilization

1. Introduction

Radiation shielding and radioactive waste stabilization are two critical requirements for safe peaceful nuclear applications. The various applications of radioactive materials have adhere to general radiation protection principles, such as justification, optimization, and dose and risk limits [1]. The reduction in personnel dose is a fundamental principle of radiation protection. To reduce the occupational radiation hazard, the level of radiation must be assessed, and three parameters are typically emphasized in controlling the radiation hazard: time, distance, and shielding [1].

The stabilization of radioactive waste reduces the possibility of radioactive contamination, migration, or the dispersing of radionuclides. Stabilization is defined as the transformation of waste into another form through solidification, embedding, or encapsulation

for safe handling, transportation, storage, and disposal in designated landfills. Radioactive waste is encapsulated by physically surrounding it in materials such as bitumen or cement, which isolates it and retains radionuclides. Packaging or immobilization protects the radioactive substances, and prevents them from being accessed by the environment or escaping into the environment [2]. Furthermore, radioactive materials constantly irradiate through the immobilizing medium, sometimes at high levels of radiation. Radionuclides with long half-lives have extended irradiation periods and make the immobilization process a problem with no simple solution, necessitating the use of another material with high radiation shielding properties.

In the solidification/stabilization process, radioactive waste is converted into a stable solid form that is insoluble by cementation or vitrification, so as to prevent the radio contamination of the surrounding environment [3].

The excellent compressive strength and cost-effectiveness of cement allow it to be widely used as a stabilizer for hazardous and radioactive waste produced initially or as a by-product from treatment operations [4,5]. Cementation technology is used for the immobilization of radioactive waste. The practice of encapsulating radioactive waste in Portland cement began during the early years of the nuclear industry due to its low cost, availability, and compatibility with aqueous waste. The properties of cement vary depending on the type of cement used and the additives used, as well as the desired characteristics that the slurry design should have in order to maintain the durability of the cement. To achieve this goal, several studies and experiments have been carried out with the target of increasing the cement's stability, lifetime, and quality, while decreasing the cement's deterioration. Portland cement can be modified using a range of additives. These may include cellulosic waste [6,7], bitumen [8,9], glass [10], polymers [11–14], nanomaterials [15,16] and cement wastes [17,18].

With the growing problem of the high levels of problematic plastic waste produced today, polyvinylchloride (PVC) is the third most-produced synthetic polymer on a global scale. Annually, approximately 39.3 million tons are consumed worldwide, with a 3.2% increase in demand per year [19]. The recycling of PVC waste, including mechanical, chemical, re-extrusion, and burning, has been a leading cause of the presence of dioxin in incinerators when they are burned, and poses a harmful waste problem [20]. PVC wastes require recycling to overcome the many troubles of waste disposal and attain a sustainable solution for various industries, including nuclear applications. PVC wastes can be mixed with cement as a sustainable additive to produce cementitious products suitable for the immobilization of radioactive waste or as a shielding construction material. This technique is an environmentally friendly and economic method for developing economies such as Egypt, due to the great need to dispose of PVC and radioactive waste at the same time.

Another type of problematic material is asphaltene, since it can form dense flocculation and deposits in reservoirs, wellbores, and transportation pipelines, and thus can cause severe operational and production problems. Fortunately, asphaltene is a low-cost hydrocarbon byproduct produced during deep oil refining, with no significant uses in the pavement industry. Recently, asphaltenes were chemically modified for use as a novel thermal conductivity enhancer for liquid paraffin [21], and have been used with cement as additives to asphalt emulsion-stabilized layers [22]. Presently, asphaltene is characterized by its proper ability to mix with cement and produce a new composite of reasonable radiation shielding properties.

This research offers a comparative study to evaluate the mechanical properties, gamma attenuation and fast neutron removal cross-section of two cement composites, namely, a cement–PVC composite and a cement–asphaltene composite. This research reports on a systematic study demonstrating the effects of the incorporation of various ratios of grated PVC waste or asphaltene emulsion into Portland cement to improve the durability and mechanical properties of the produced composite, which will be used as a solidifying agent for radioactive waste, to enhance the attenuation coefficient required to achieve radiation safety in a shielding construction material, or to achieve the two purposes at the same

time. This study aimed to investigate the suitability of some cement additives of definite compositions for producing proper composites with good performances and high densities, using different types of aggregates (asphaltene or PVC) that could enhance the shielding efficiency or achieve the safe stabilization of radioactive waste. The most significant issue in the present study is the elimination of two problematic wastes (asphaltene and PVC), and the production of an innovative product with reasonable efficiency that can be used in the immobilization of radioactive waste or radiation shielding construction, with eco-sustainability, low energy demands, and cost minimization.

2. Methodology of Research

2.1. Materials

2.1.1. Portland Cement

A commercially available Portland cement supplied by El Sewedy cement company, CEM1(42.5 N), was used in this study as the binding material, developed according to Egyptian Standard Specifications, ES 4756-1/2005 [23], and the British Standard Institution (BSI), EN 197-1/2011 [24].

2.1.2. Polyvinylchloride (PVC)

Scraps of polyvinylchloride pipes were collected as municipal solid waste and cleaned with tap water. The large pieces were crushed and sieved to obtain fine particles of polyvinyl waste appropriate for the subsequent blending with cement [25].

2.1.3. Asphaltene

The asphaltene used in this study was of commercial grade. Asphaltene is a fraction of a hydrocarbon fuel, consisting of condensed polynuclear aromatic ring systems at the center bearing alkyl side chains with hetero elements, the FT-IR spectrum has been presented previously in the literature [26].

2.2. Preparation of Cementitious Samples

Portland cement, the major component used in this study, was hydrated with 35% tap water without any additives to prepare the blank samples, or was blended with various additives, such as asphaltene or PVC, to prepare the different composites at various additive ratios.

Each group of compositions contained more than six specimens, three of which were subjected to compressive strength tests, three to porosity and water absorption measurements, and the remaining were subjected to attenuation testing. The pastes of various groups were poured into cylindrical bottles that were internally coated with Vaseline, manually compacted, tightly closed, and safely stored in the laboratory at room temperature (25.5 °C in wet conditions) for 28 days to allow complete reaction and proper solidification. After curing, the solidified samples were demolded and subjected to mechanical integrity tests, permeability spectroscopic analysis, and attenuation measurements.

2.3. Assessment of Mechanical Properties

The mechanical integrity is here expressed as compressive strength in MPa for cementitious samples—either cement mixed with asphaltene or PVC, or cement hydrated without any additives. In the Italian testing machine, Ma test E 159 SP, cylindrical samples were loaded axially between the loading plates to determine the maximum load before sample cracking, determined via the ASTM C109 standard test method [27]. Furthermore, the boiling water technique was used to measure porosity, bulk density, and water absorption following ASTM C 20-00 [28]. Using the water displacement method, which is based on Archimedes' saturation technique, the masses of porous specimens, both dry and in water, were recorded. The following two equations can be used to calculate apparent porosity.

$$P = [(W - D)/V] \times 100 \tag{1}$$

$$V = W - S \tag{2}$$

P—apparent porosity (%), W—saturated mass of the specimen (g), D—dry mass of the specimen (g), S—suspended weight of the specimen (g), V—exterior volume of the specimen (cm^3).

Then, the bulk density (r) can be calculated from the volume and saturated mass of the sample, as:

$$r = D/V \tag{3}$$

2.4. Determination of Gamma-Ray Shielding by Theoretical Aspects

The attenuation of γ-rays by a medium can be described by the well-known Lambert–Beer formula [29]:

$$I = B \times I_0 \times e^{-\mu_m x \rho} \tag{4}$$

where I and I_0 are, respectively, the transmitted and initial intensities of photons; B (X, E) is the build-up factor, depending on the penetration depth x of the material and energy E of the incident photon; μ_m is the mass attenuation coefficient (cm^2/g) and ρ is the mass density (g/cm^3). The mass attenuation coefficients (MAC) of the prepared samples at an energy range of 0.015–15 MeV were calculated using Phy-X/PSD, a user-friendly online software. This program, published by the National Institute of Standards and Technology, includes the coefficients of attenuation of all elements in the periodic table at various energies (NIS) [30].

The radiation protection efficiency (RPE), which is provided by the following equation, may be used to study the shielding efficiency of an absorber sample.

$$I = \left(1 - \frac{I}{I_0}\right) \times 100 \tag{5}$$

The effective atomic number (Z_{eff}) of a sample containing a mixture of elements is the ratio of its effective atomic cross-section (σ_a) to its electronic cross-section (σ_e), and can be calculated using the following formula [31];

$$Z_{eff} = \frac{\sigma_a}{\sigma_e} = \frac{\sum_i f_i A_i (\mu_m)_i}{\sum_i f_i \frac{A_i}{Z_i} (\mu_m)_i} \tag{6}$$

where A_i and Z_i are the atomic weight and the atomic number, respectively, of the *i*-element inside the sample. The term f_i refers to the fractional abundance. The Auto-Z_{eff} program is a user-friendly software presented recently by Taylor that can be used for the quick computation of Z_{eff} [32].

The equivalent atomic number (Z_{eq}) of the prepared sample at a specific energy may be calculated using the fractional mass attenuation coefficient owing to Compton scattering $R = (\mu_m)_{comp}/(\mu_m)_{total}$. This may be accomplished by matching R_1 and R_2 for two pure elements with two consecutive atomic numbers Z_1 and Z_2 at the same energy, resulting in R being positioned between R_1 and R_2 (near to R). The logarithmic interpolation formula of Z_{eq} is as follows [33]:

$$Z_{eq} = \frac{Z_1 (\log R_2 - \log R) + Z_2 (\log R - \log R_1)}{\log R_2 - \log R_1} \tag{7}$$

Harima et al. created the Geometric Progression (G-P) approximation, which is used to calculate the exposure build-up factors (EBF) of the prepared samples (1993). Using the interpolation formulas below, one can calculate the G-P fitting parameters and the related EBF [34,35].

$$B(E, \ X) = 1 + \frac{b-1}{K-1}\left(K^X - 1\right) \text{ for } K \neq 1 \tag{8}$$

$$B(E, \ X) = 1 + (b-1)X \text{ for } K = 1 \tag{9}$$

where X is the penetration depth ($X \leq 40$ mfp), b is the EBF value at 1 mfp, $K(E, X)$ is the multiplicative dose factor, and b, c, a, X_K and d are the computed G-P fitting parameters, which depend on the attenuating medium and the source energy. The G-P fitting parameters (b, c, a, X_K, and d) of the prepared samples throughout the 0.015–15 MeV gamma-ray energy range up to 40 mfp may be interpolated using a comparable logarithmic formula, such as Formula (5) [36,37]. The G-P fit requirements for the elements were derived from a research study conducted by the American Nuclear Society [38].

The fast neutron removal cross-section (Σ_R) of this material is the proportion of fast neutrons removed from a beam of neutrons during its penetration and after the first contact with the target material. For neutron energies of 2–12 MeV, the effective removal cross-section is nearly constant [36]. In the case of a combination of components, the removal cross-section may be calculated using the following formula:

$$\Sigma_R = \sum_i w_i (\Sigma_R)_i \tag{10}$$

where w_i and $(\Sigma_R)_i$ are the elemental weight fraction and removal cross-section, respectively. Dividing by the mass density (ρ), the fast neutron mass removal cross-section Σ_R/ρ (cm^2 g^{-1}) can be calculated.

3. Results and Discussion

3.1. Investigation of Mechanical Integrity and Porosity

3.1.1. Cement-Based PVC

The mechanical stability, reflected in the compressive strength of the reference sample of cement without any additives, had the highest value of 24.4 MPa; this is compared with the steady decrease in compressive strength, to 4.5 MPa, with an increasing PVC ratio from 10 to 50 wt.%, as shown in Figure 1. This fall in compressive strength is attributed to the heterogeneity between the two components, and their pores. On the other hand, the porosity showed the opposite trend; a gradual increase in porosity was correlated with an increasing PVC content in cement. The behavior of porosity explains the decreasing compressive strength.
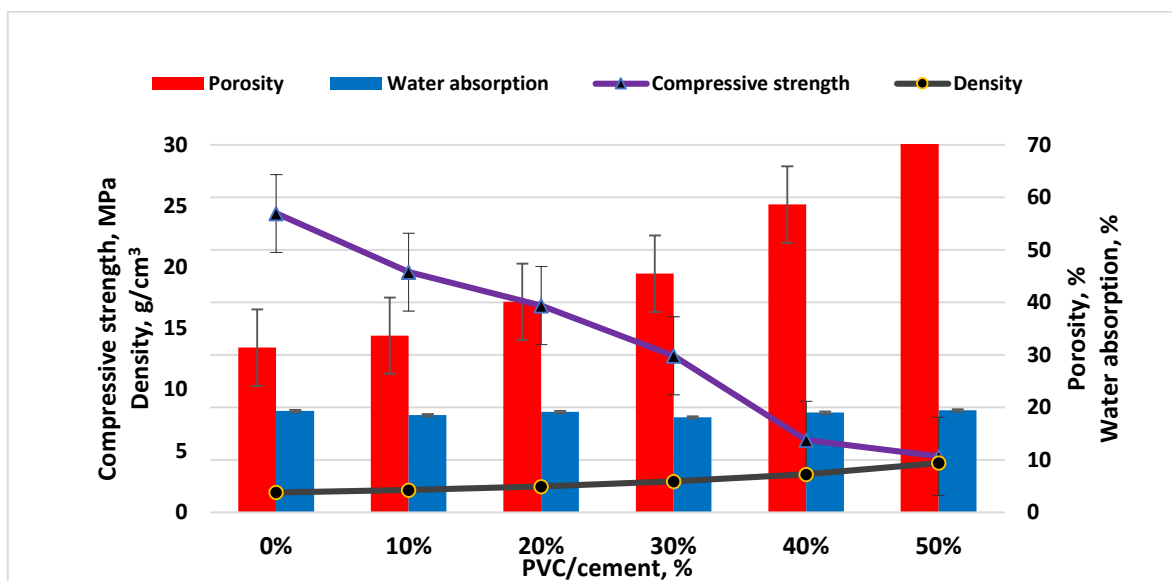

Figure 1. Compressive strength, density, water absorption, and porosity of cement blended with extended ratios of PVC.

However, the bulk density of the hardened cementitious composites was slightly increased by increasing the additive amount of PVC granules, due to their relatively

high density, while the water absorption showed a constant value while increasing the addition of recycled PVC. According to previous research and in agreement with the current experiment, absorption affinity indirectly reflects the porosity due to permeable pore volume and connectivity [39]. As per the US Nuclear Regulatory Commission, the standard value relative to the compressive strength of solidified radioactive waste has to be more than 3.4 MPa [40], and the compressive strength of cement mixed with 50 wt.% PVC has reached more than 4.5 MPa; this value is significantly higher than recommended, and displays the standard parameters required for the immobilization of low and intermediate levels of radioactive waste.

3.1.2. Cement-Based Asphaltene

To examine the effect of increasing the addition of asphaltene into cement paste on the mechanical properties of the produced composite, the compressive strength, bulk density, porosity and water absorption are reported in Figure 2. In this condition, asphaltene will produce a homogeneous mixture with cement compared with PVC. A significant improvement in porosity to 24.24 can be observed with the gradual decrease in water absorption by increasing the amount of asphaltene, due to the potential filling of voids and pores inside the cementitious paste with asphaltene granules. A reasonable decrease in compressive strength was achieved with higher additions of the asphaltene, to reach 8.8 MPa at 50 wt.% asphaltene. It can be concluded from the results presented in Figures 1 and 2 that the heterogeneity and low density of the PVC with cement are higher than in the case of using asphaltene with cement, thus the porosity and water absorption increase while the compressive strength decreases [41,42].
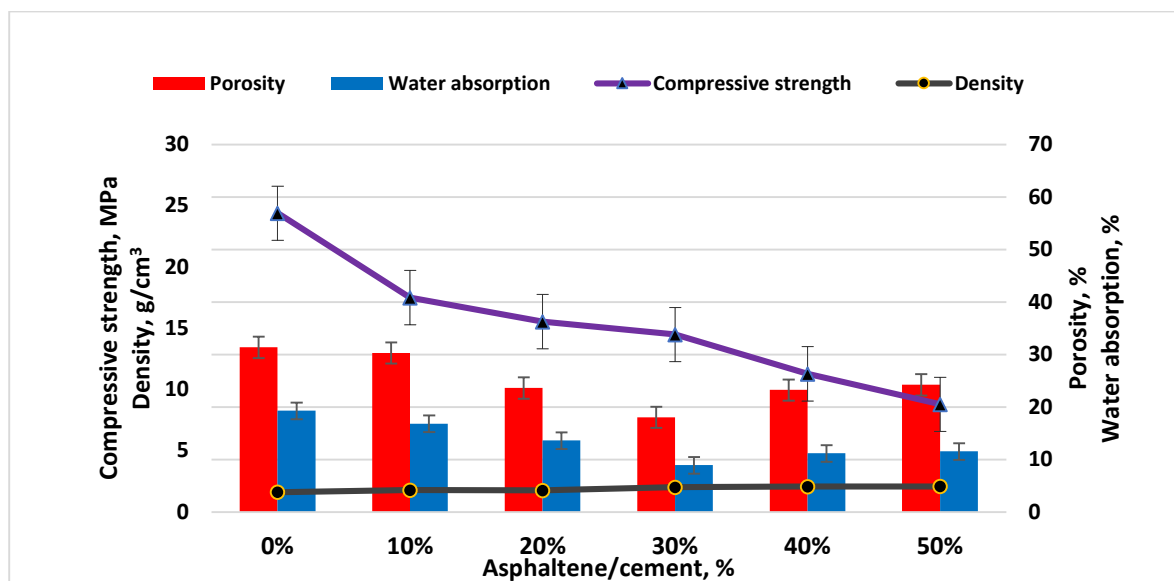

Figure 2. Compressive strength, density, water absorption, and porosity of cement blended with extended ratios of asphaltene.

A slight increase in the bulk density of the cementitious samples including asphaltene was detected when increasing the ratio of mixed asphaltene. Despite the presence of asphaltene not causing improvements in the mechanical integrity, the compressive strength of cement-based asphaltene at 50 wt.% has a value more than twice that required of materials used in radioactive waste stabilization, according to the standard specifications previously mentioned (3.4 MPa) [40], and this value is higher than in the case of PVC with cement (4.5 MPa). It is possible to conclude that asphaltene is more advantageous than PVC when mixed with cement up to 50 wt.%, and that it could be considered as a good additive to Portland cement for use in the solidification/stabilization of radioactive

waste. Table 1 shows a descriptive comparison between the results here obtained and those reported previously in the recent literature.

Table 1. Comparing the mechanical properties of the present materials and others previously studied.

System	Compressive Strength, MPa	Porosity, %	Reference
Standard value	3.4	-	[40]
Cement without additives	32.0–36.5	27.5–30.0	[43,44]
Cement mixed with bitumen (30 wt.%)	7.62	60.14	[8]
Cement mixed with PVC (50 wt.%)	4.5	78.3	Present study
Cement mixed with asphaltene (50 wt.%)	8.8	24.24	Present study

3.2. Radiation Shielding Performance

3.2.1. Gamma-Rays Shielding Properties

The obtained results of the mass attenuation coefficients (MAC) of the prepared samples are shown in Figure 3. A regular trend in the MAC for all the prepared samples is observed. The MAC values are dramatically reduced at low photon energies of less than 0.1 MeV, where photoelectric interactions are prominent. The controlling of the photoelectric effect in such a region is perceived as a dramatic drop in the MAC. Compton scattering eventually became the dominant interaction as the energy range increased from 0.1 to 1 MeV, and the likelihood of interaction was inversely related to the energy. Therefore, the values of the MAC for all samples were also gradually decreasing. The MAC values for all samples increased somewhat in the energy range 1.02–15 MeV, where the pair formation interaction became the most prominent and was directly proportional to the energy. The obtained results indicate that adding asphaltene or PVC to cement samples slightly decreases the mass attenuation coefficient, and in turn increases the half-value layer of the cement, as shown in Figures 3 and 4. This was expected due to the low Z_{eff} of asphaltene and PVC compared with cement.
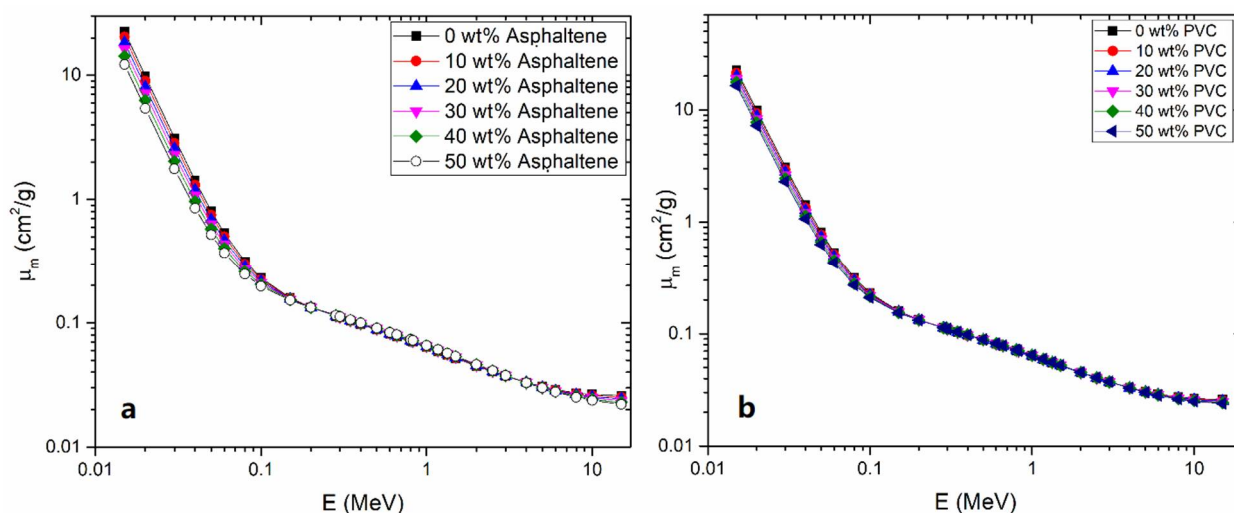

Figure 3. Mass attenuation coefficients of cement samples containing different concentrations of (**a**) asphaltene and (**b**) PVC.
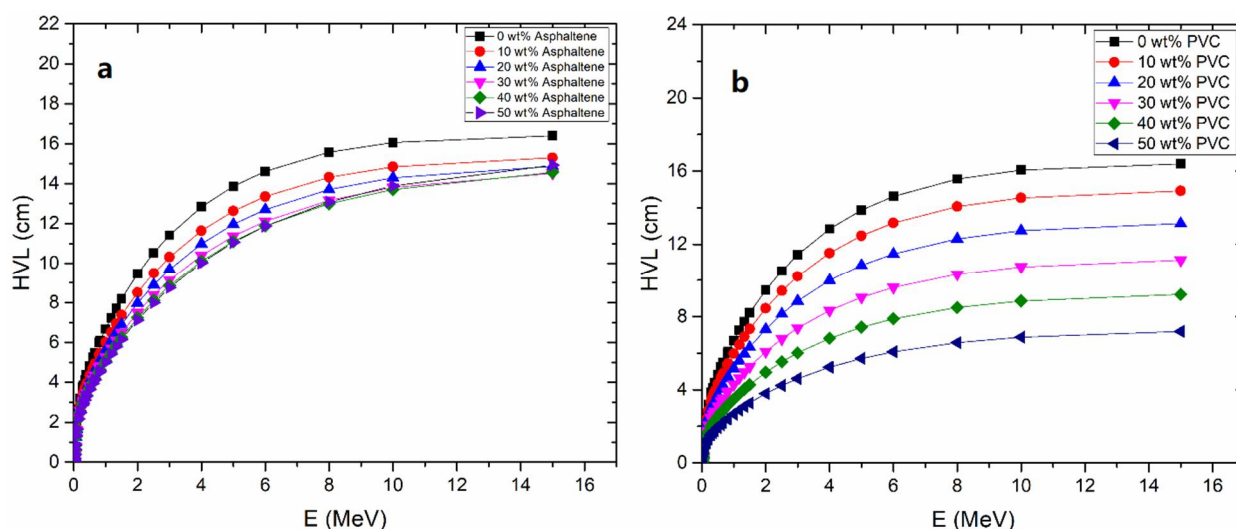
Figure 4. Half value layer (HVL) results of the cement samples containing different concentrations of (**a**) asphaltene and (**b**) PVC.

3.2.2. Radiation Protection Efficiency (RPE)

The prepared samples' radiation protection efficiencies compared with those of known materials, such as barite concrete at 0.662 MeV and 2 cm thickness, are shown in Figure 5. The obtained results show that cement with asphaltene and cement with PVC have larger RPEs than pure cement. Additionally, cement with 50% PVC has the largest RPE (79.4%) compared with pure cement (46.8%) and barite concrete (63.2%). The enhanced RPE of cement with the addition of asphaltene or PVC indicates these composites for use in radioactive waste stabilization. Moreover, the highest RPE of cement with 50% PVC recommends its use for gamma radiation shielding applications, more strenuously than concrete.
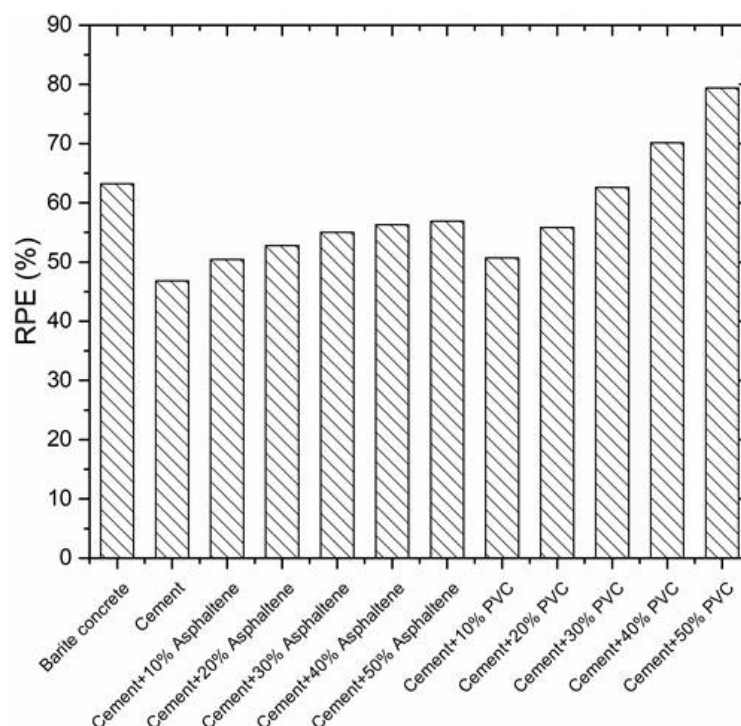

Figure 5. Radiation protection efficiency (RPE) results of the cement samples containing different concentrations of asphaltene and PVC.

3.2.3. Effective Atomic Number (Z_{eff})

Figure 6 shows the predicted effective atomic number Z_{eff} findings for the samples including asphaltene or PVC at energy ranges of 0.015–15 MeV. The Z_{eff} varies depending on the dominant interaction in each energy range, since the attenuation cross-section is proportional to Z^{4-5} for photoelectric attenuation, to Z for a Compton interaction, and to Z^2 for pair creation [45].
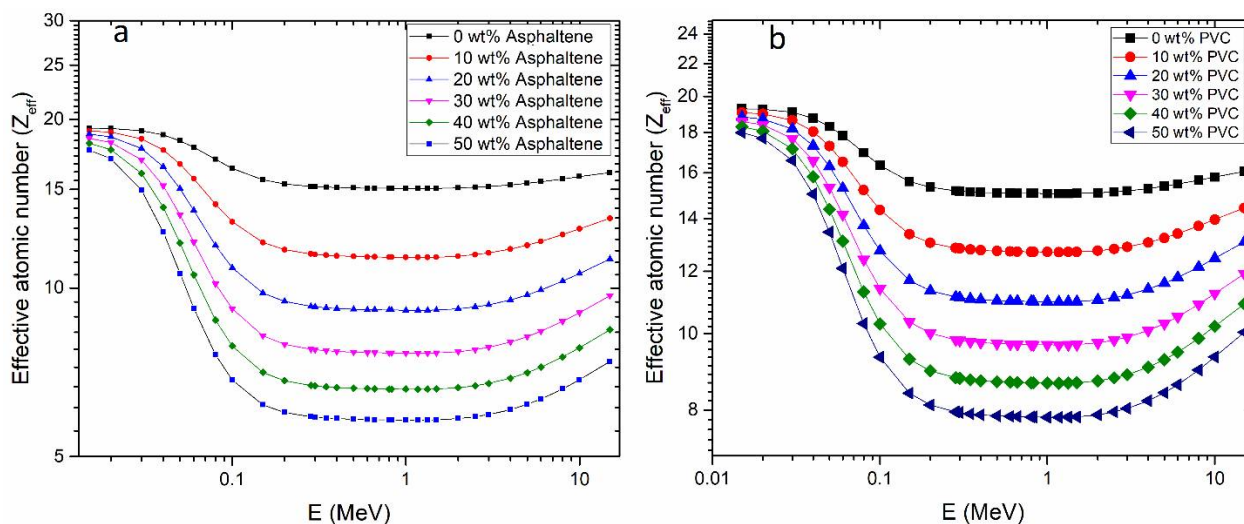

Figure 6. Effective atomic numbers of the cement samples containing different concentrations of (**a**) asphaltene and (**b**) PVC.

3.2.4. Exposure Build-Up Factors (EBF)

The computed gamma-ray energy exposure buildup factors (EBF), as a function of the incoming energy at 1, 2, 5, 10, 20, 30 and 40 MFP, are presented in Figures 7 and 8 for the cement samples with asphaltene and PVC, respectively, based on the acquired G-P fitting parameters (b, c, a, X_k, and d) for each sample.

Using the photon energy, additive concentrations (PVC or asphaltene), and penetration depth dependencies of the EBF, the data may be understood. The lowest EBF values were found in low- and high-energy areas, whereas values in the intermediate energy zone were greater for all asphaltene and PVC concentrations, as shown in Figures 7a–f and 8a–f, respectively. In the low- and high-energy areas, total absorption was most likely caused by the photoelectric effect and pair creation. Multiple Compton scattering, and hence larger EBF values, were seen in the intermediate area [45].

3.2.5. Fast Neutron Removal Cross-Section

Furthermore, the calculated fast neutron removal cross-sections Σ_R of the examined samples are shown in Figure 9a,b. Although the gamma MAC is slightly decreased with the adding of asphaltene or PVC, the neutron removal cross-section was highly increased, reaching 171% in the case of 50 wt.% asphaltene and 304% in the case of 50 wt.% PVC.

This can be attributed to the higher fast neutron removal cross-sections of the constituent elements of both PVC and asphaltene. The effective fast neutrons mass removal cross-sections Σ_R/ρ (cm^2 g^{-1}) of cement and PVC are 0.03069 and 0.06252 cm^2 g^{-1}, respectively [46], while for asphaltene this value is about 0.08820 cm^2 g^{-1}. Finally, based on the obtained radiation shielding results, we have found that adding PVC or asphaltene does not significantly change the gamma shielding qualities, but it does improve the neutron removal cross-section and the mechanical properties of the cement.
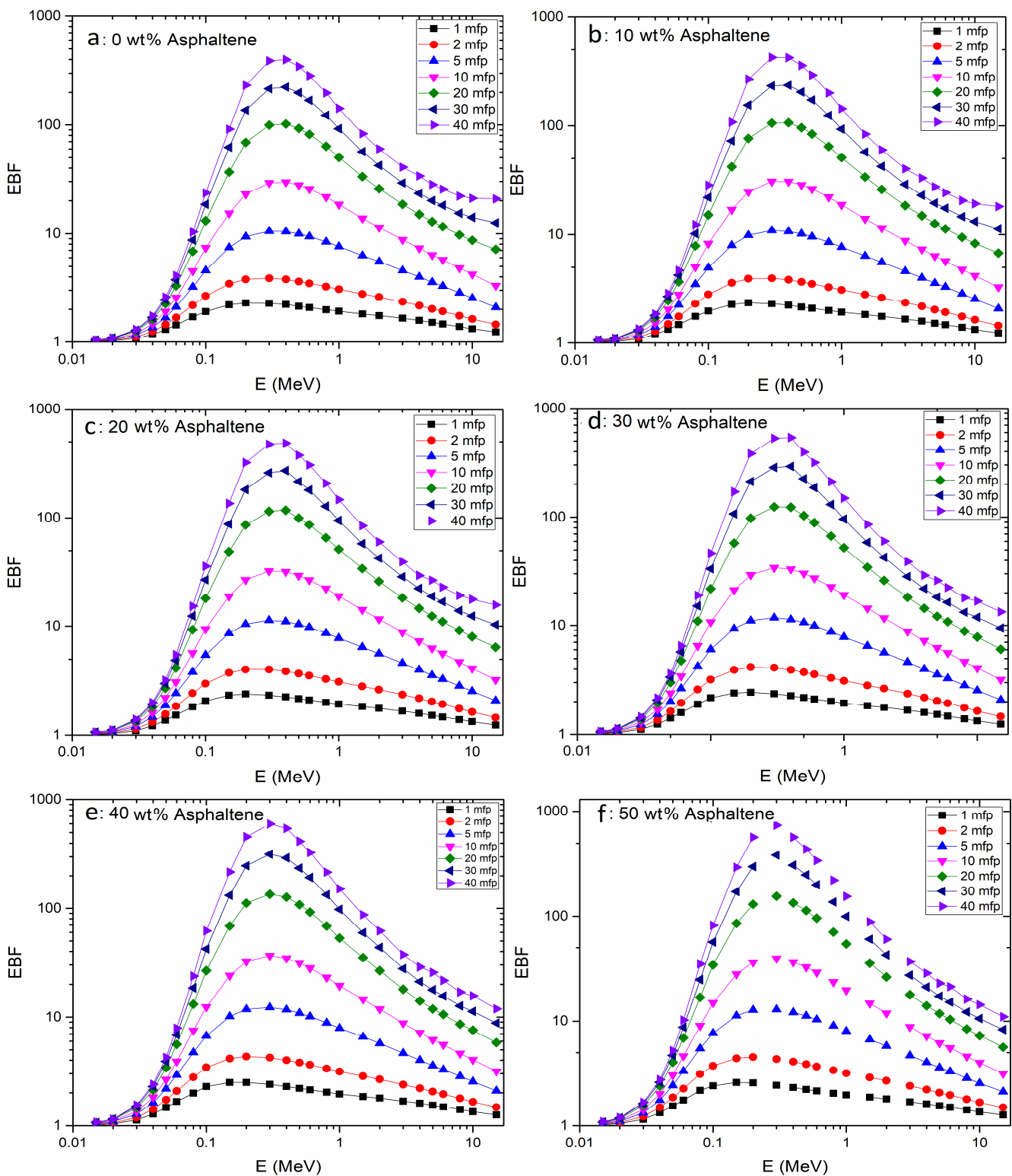
Figure 7. Exposure build-up factors of the cement samples containing different concentrations of asphaltene at photon energies of 0.015–15 MeV up to 40 mfp for (**a**) 0 wt.% asphaltene, (**b**) 10 wt.% asphaltene, (**c**) 20 wt.% asphaltene, (**d**) 30 wt.% asphaltene, (**e**) 40 wt.% asphaltene and (**f**) 50 wt.% asphaltene.
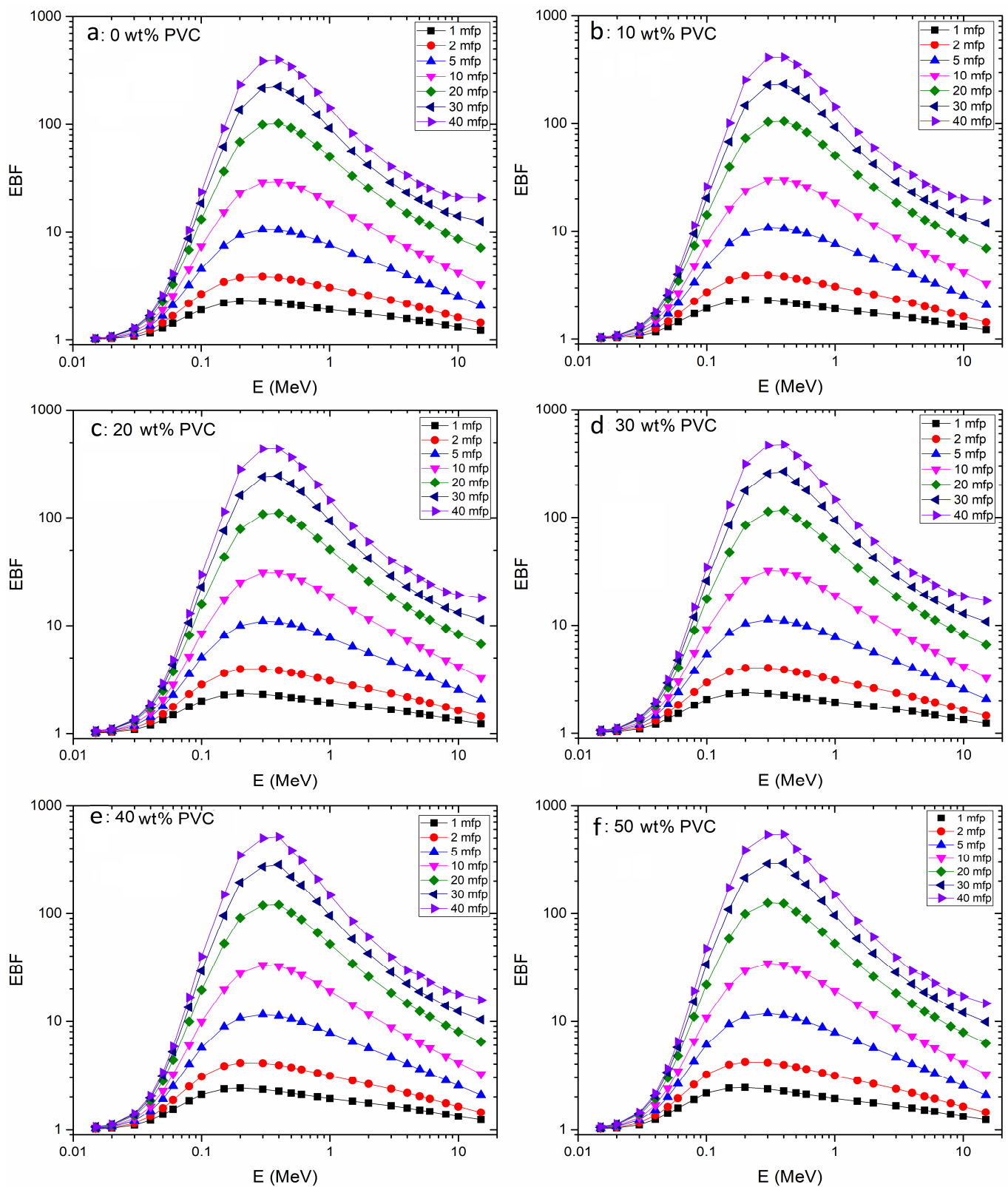
Figure 8. Exposure build-up factors of the cement samples containing different concentrations of PVC at photon energies of 0.015–15 MeV up to 40 mfp for (**a**) 0 wt.% PVC, (**b**) 10 wt.% PVC, (**c**) 20 wt.% PVC, (**d**) 30 wt.% PVC, (**e**) 40 wt.% PVC and (**f**) 50 wt.% PVC.
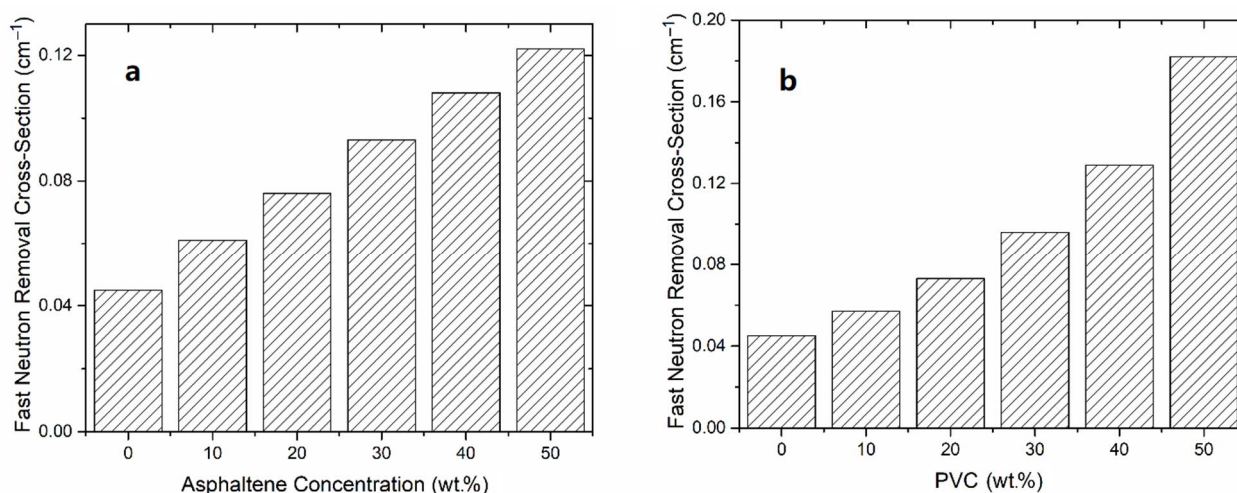
Figure 9. Fast neutron removal cross-sections of the cement samples containing different concentrations of (**a**) asphaltene and (**b**) PVC.

Comparisons between the results obtained here for the prepared composites and those for the well-known ones such as Barite concrete and Portland cement are shown in Table 2. The tabulated results indicate that cement mixed with bitumen has a higher mass attenuation coefficient than the other composites, while the fast neutron removal cross section of cement mixed with PVC or asphaltene showed the highest values.

Table 2. Comparison between similar shielding materials.

System	Rang of μ_m (cm^2/g) $\times 10^{-2}$ (at 0.662 MeV)	Fast Neutron Mass Removal Cross-Sections, Σ_R/ρ (cm^2 g^{-1})	Reference
Barite concrete	6.7–7.8	0.041–0.027	[47,48]
Portland cement	7.76	0.031	[30]
Cement mixed with bitumen (10–50 wt.%)	9.94–14.11	-	[8]
Cement mixed with PVC (10–50 wt.%)	7.78–7.84	0.045–0.182	Present study
Cement mixed with asphaltene (10–50 wt.%)	7.80–8.01	0.045–0.122	Present study

4. Conclusions

In this study, the elimination of two problematic waste products (asphaltene and PVC), in order to produce an innovative product with a reasonable efficiency that can be used in the immobilization of radioactive waste or radiation shielding construction, has been conducted, while the achieving of environmental sustainability, low energy demand, and cost minimization have been considered in parallel.

Asphaltene or PVC were mixed with Portland cement in different proportions up to 50%. The performances of the resulting compounds were evaluated using several parameters, such as compressive strength, porosity, bulk density, water absorption, and attenuation coefficient. The physical and mechanical stabilities of the modified cement composites gradually decreased with the increases in the amounts of both additives, while the attenuation and isolation performance, as well as γ-radiation shielding, were slightly affected. The effective cross-sections for removing neutron mass from cement with PVC or asphaltene were significantly increased. The results of this study show that the new compound prepared by mixing cement with asphaltene or PVC can be used to safely

encapsulate hazardous toxic and radioactive wastes, while providing adequate radiation shielding from gamma-rays for the environment at the same time.

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