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Investigation of Radiation Properties of CO_2 Reacted Portland

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Abstract: In this study we explored the radiation properties of CO_2-reacted Portland which take place in kiln process of cement production. The use of CO_2 can change Portland density via chemical process to obtain $CaCO_3$. When the CO_2 capture rate of Portland cement is zero, the density is 2.3 g/cm^3, while the CO_2 capture rate is 100%, the density is reached to 2.705 g/cm^3. The radiation shielding properties were explored using FLUKA code. To define the radiation shielding properties of the CO_2-reacted Portland, four types of beams (photons, electrons, protons and neutrons) were used. These beams have been used to explain the leptonic and hadronic interactions. The CO_2-reacted Portland radiation length and density have been calculated and presented. The energy depositions of four beams with various beam energies were examined by considering density variation of the cement. It has been found that CO_2-reacted Portland has more efficient radiation shielding than traditional Portland materials. The carbonization of Portland will be carried out during the kiln process, not by the CO_2 diffusion process, which is a very slow process, but by the faster and more convenient spray method.

***Key words**:* CO_2-reacted Portland, Radiation, Radiation length

CO_2 Reaksiyonlu Portland'ın Radyasyon Özelliklerinin İncelenmesi

Öz: Bu çalışmada, çimento üretiminin fırınlama esnasında elde edilebilecek CO_2 reaksiyonlu Portland'ın radyasyon özelliklerini araştırdık. CO_2 ile reaksiyona giren Portland çimentosunun, kimyasal süreç yoluyla $CaCO_3$ üretilmesi neticesinde, yoğunluğu değişir. Portland çimentosunun, CO_2 yakalama oranı sıfır iken yoğunluk 2.3 g/cm^3, CO_2 yakalama oranı %100 iken yoğunluk 2.705 g/cm^3 olmaktadır. Radyasyon koruma özellikleri FLUKA simülasyon programı kullanılarak araştırıldı. CO_2 reaksiyonlu Portland'ın radyasyon kalkanı özelliklerini araştırmak için dört tip ışın demeti (fotonlar, elektronlar, protonlar ve nötronlar) kullanıldı. Bu ışın demetleri, leptonik ve hadronik etkileşimleri açıklamak için kullanıldı. Bu çalışmada CO_2 reaksiyonlu portlandın, radyasyon uzunluğu ve yoğunluğu hesaplanıp sunuldu. Farklı demet enerjilerine sahip dört demetin hedef portland üzerindeki enerji birikimleri, çimentonun yoğunluk değişimi dikkate alınarak incelendi. CO_2 reaksiyonlu Portland'ın geleneksel Portland malzemelerden daha verimli radyasyon kalkanlama özelliğine sahip olduğu bulundu. Portland'ın karbonizasyonu, çok yavaş bir işlem olan CO_2 difüzyon işlemi ile değil daha hızlı ve daha elverişli bir işlem olan püskürtme yöntemiyle fırın işlemi sırasında gerçekleştirilecektir.

***Anahtar kelimeler**:* CO_2 reaksiyonlu Portland, Radyasyon, Radyasyon uzunluğu

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1. Introduction

The main idea in protection against radiation is to know the tolerable doses and to prevent radiation workers and the surrounding people from overdosing [1]. Due to these desirable properties concrete which would created by Portland should have radiation resistive as much as possible. There are many types of concrete used today. These are classified according to their stress-strain, and radiation shielding properties. For this purpose, and they use aggregates. Radiation matter interaction is affected by both radiation and matter properties, such as leptons (electron), photons are interacted with matter via electromagnetic interaction and also hadrons (proton, neutron) interact with hadronic interaction. Energy deposition of matter is defined by Bethe-Bloch formula and it is directly affected by matter's atomic mass number, atomic mass and density, besides logarithmically affected by the incoming beam energy [2,3,4,5,6].

During the production of conventional cement, which requires kiln process, will produce CO_2 gases which effect on environmental greenhouse gas level [7]. Recently, CO_2-reacted (carbon-enriched) concrete studies have gained importance [8,9] in order to reduce carbon dioxide emissions into the atmosphere. The during the carbonization reaction diffusion is not taken into consideration, rather a process which can be applied using heating mechanism while taking into account not only large surface area of conventional Portland but also stirring. The transited phase is portlandite and the process is called as Calcium silicate hydrate (CSH) which will not be investigated present study [8].

In this study, the radiation damage of the CO_2-reacted Portland sample will be examined. A chemical reaction takes place when carbon dioxide is sprayed during the kiln process. Then the cement is being prepared. While producing of CO_2-reacted Portland, the density and radiation absorption and many properties of the cement would change. Radiation damage of the Portland would be calculated using the FLUKA simulation code [10,11]. Moreover comparison of the radiation damage of CO_2-reacted concrete with the conventional Portland has been presented. Radiation properties of proposed Portland has not been studied before. The CO_2-reacted Portland is not an alternative option of aggregated heavy concrete.

2. Material and Method

2.1 The concrete

Concrete has been used since ancient times. The production method of modern concrete has remained effectively unchanged over the past two centuries. Concrete; It consists of a mixture of sand and gravel (aggregate combination), Portland cement, water, chemical additives and other trace components in proportions to meet the structural and performance needs of different applications [12,13,14].

There are many efforts have been done for concrete studies to improve the concrete in terms of stress-strain, and radiation shielding properties etc [12,13,14]. The density of heavy concretes using as a radiation shielding material have more than 2.6 g/cm^3.[12] Heavy concretes are generally used for radiation shielding material such as in particle accelerator buildings, radiation therapy room of hospitals which is used for diagnostics and treatment, nuclear power plant and medical x-ray imaging rooms. Heavy concrete aggregates are made of heavy metal composite aggregates. These aggregates are barite (barium sulfate $BaSO_4$), limonite, magnetite, ferrous minerals etc. Their densities are over than 3.2 g/cm^3 [12,15].

Moreover some concrete types are under investigation such as bio-concrete for self-repairing and carbon enriched concrete which reduce to greenhouse gases. The carbon enriched Portland is CO_2-reacted Portland [16]. The CO_2 reaction with Portland increase the Portland density without including any type of aggregates. The procedure used has been explained in the next subsection.

2.2 Properties of Proposed concrete

The production of CO_2-reacted Portland, would reduce the greenhouse gas impact due to the using of CO_2 which is generated during kilning in the conventional Portland's production process.The density and, mass fractions of the conventional Portland, which is world accepted one, are given as a FLUKA input card format in Table 1.

Table 1. Conventional Portland Mass Fractions [10,11]

MATERIAL	**Z:**	**Name:** PORTLAND **Am:**	**# A:**	**ρ:** 2.3 **dE/dx:**
COMPOUND		**Name:** PORTLAND	**Mix:** Mass	**Elements:** 10..12
	f1: 0.01	M1: HYDROGEN	f2: 0.001	M2: CARBON
	f3: 0.529107	M3: OXYGEN	f4: 0.016	M4: SODIUM
	f5: 0.002	M5: MAGNESIU	f6: 0.033872	M6:ALUMINUM
	f7: 0.337021	M7 SILICON	f8: 0.013	M8: POTASSIU
	f9: 0.044	M9: CALCIUM	f10: 0.014	M10:IRON
	f11:	M11:	f12:	M12:

In the Table 1, the mass fractions of atoms are given with numbers of following “f”. The density of carbon dioxide reacted concrete varies according to the amount of captured carbon dioxide. Density variation occur during the chemical reaction given in Equation 1. In the reaction, production of calcium carbonate ($CaCO_3$), would decrease the volume of the cement and, then density would increase due to crystallization [17]. The $CaCO_3$ has hexagonal crystal structure. Therefore density of the CO_2-reacted Portland can be found as following:

$$\mathrm{Ca(OH)_2 + CO_2 \quad \rightarrow \quad CaCO_3 + H_2O} \tag{1}$$

When we use the same numbers for the atoms in Table 1, for indicates their moles and their mass are would be $n_1,...,n_9$, $m_1,\ldots,m_9$ respectively. To find out how many moles of calcium (n_9) would react, first we should determine mass of Ca (m_9) in the conventional Portland, and then we can find the mole of Ca as given in following equations:

$$\mathrm{m_9 = m_T \times f_9 = 2.3 \times 0.044 = 0.1012gr} \tag{2}$$

$$\mathrm{n_9 = m_9 \div m_{Ca} = 0.1012 \div 40.078 = 2.525\ 10^{-3}mole} \tag{3}$$

- We have assumed that an ultimate hold of CO_2 which is 90% by conventional Portland in orrder to produce CO_2-reacted Portland. The mole of used Ca would be $\mathrm{n_9 = 2.272\ \ 10^{-3}mole}$.
- To equalize the reaction, same amount of mole of CO_2 must be used. Therefore, the mole of CO_2 is also equal to $\mathrm{n_{CO_2} = 2.272\ \ 10^{-3}mole}$. The volume difference of input and output reaction is (for $2.272\ 10^{-3}$ mole of each reactant in the chemical reaction)

$$V_{diff} = \frac{V_{Ca(OH)_2} + V_{CO_2}}{V_{CaCO_3} + V_{H_2O}} = \frac{\frac{m_{Ca(OH)_2}}{d_{Ca(OH)_2}} + \frac{m_{CO_2}}{d_{CO_2}}}{\frac{m_{CaCO_3}}{d_{CaCO_3}} + \frac{m_{H_2O}}{d_{H_2O}}} = \frac{0.07614 + 0.064007}{0.08391 + 0.04093} = 1.1226 \tag{4}$$

- The mass of CO_2 reacted Portland;

$$m_T + m_{CO_2} \quad m'_T = 2.3 + 0.09998 \qquad m'_T = 2.39998\ \mathrm{g} \tag{5}$$

- The volume difference can be used to find the density of CO_2-reacted Portland. The volume of conventional Portland is used as V_1=1 cm^3.

$$d'_T = \frac{m'_T}{V_1/V_{diff}} = 2.694\ \mathrm{g/cm^3} \tag{6}$$

- As water (H_2O) is released as a result of this chemical reaction, it is expected that there will be a decrease in the amount of water needed when using the conventional Portland. 15% of the products by mass was determined as water and 85% as calcium carbonate ($CaCO_3$).

While the density of conventional Portland is 2.3 g/cm^3 before carbon dioxide reacted, this value becomes 2.705 g/cm^3 in the case of 100% CO_2 retention. Since the CO_2 retention is assumed to be 90% in this study, the density of CO_2-reacted Portland is found to be 2.694 g/cm^3. Therefore, CO_2-reacted Portland concrete cannot be an alternative to heavy concrete which has a density of ~3.2 g/cm^3 [18,19].

In order to find the mass fractions of CO_2-reacted Portland; the mass fractions in Table 1 are used with Equaiton 7, (the new mass of CO_2-reacted Portland) will be used.

$$\sum_i \left(f_i \frac{m_T}{m'_T}\right) = \sum_j f'_j - (f'_2 + f'_3) \tag{7}$$

The primes are indicating the new mass fractions of compounds. The some of mass fractions are given as;

$$f'_1 = f_1 \times (m_T/m'_T) = 0.01 \times (2.3/2.39998) = 0.0095834 \tag{7a}$$

$$f'_2 = \left(f_2 \times (m_T/m'_T)\right) + \left(m_{CO_2} \times \left(m_{c\_ratio}\right)/m'_T\right) \tag{7b}$$
$$= \left(0.001 \times (2.3/2.39998)\right) + (0.09998 \times 0.2727/2.39998) = 0.012319$$

$$f'_3 = \left(f_3 \times (m_T/m'_T)\right) + \left(m_{CO_2} \times \left(m_{o\_ratio}\right)/m'_T\right) \tag{7c}$$
$$= \left(0.529107 \times (2.3/2.39998)\right) + (0.09998 \times 0.7272/2.39998)$$
$$= 0.5373593$$

Where m_{c_ratio} =0.2727 and m_{o_ratio} =0.7272 are mass ratio of Carbon and Oxygen to mass of CO_2 respectively. The results of new mass fractions are presented as a FLUKA card in Table 2.

Table 2. CO_2-reacted Portland Mass Fractions [10,11]

MATERIAL	**Z:**	**Name:** PORT2 **Am:**	**#** **A:**	**ρ:** 2.694 **dE/dx:**
COMPOUND		**Name:** PORT2	**Mix:** Mass	**Elements:** 10..12
	f1: 0.0095834	M1: HYDROGEN	f2: 0.012319	M2: CARBON
	f3: 0.5373593	M3: OXYGEN	f4: 0.015333	M4: SODIUM
	f5: 0.0019668	M5: MAGNESIU	f6: 0.032461	M6: ALUMINUM
	f7: 0.322981	M7 SILICON	f8: 0.0112458	M8: POTASSIUM
	f9: 0.042167	M9: CALCIUM	f10: 0.013417	M10: IRON
	f11:	M11:	f12:	M12:

2.3 Radiation length

Radiation length of a material is a characteristic of material, related to the energy loss of particles electromagnetically interacting with it given as in Equation 8 [20];

$$X_0 = 716.4 * \frac{A}{Z(Z+1)\ln\frac{287}{\sqrt{Z}}} \tag{8}$$

where A is atomic mass number and Z is the atom number. For the conventional Portland case radiation length can be found via including atomic mass fractions is calculated as;

$$\frac{1}{X_0} = \sum_{i=1}^{10} \frac{f_i}{X_{0i}} = 0.037367 \qquad X_0 = 26.76138 \text{ cm} \tag{8a}$$

where mass fractions taken from Table 1. For the CO_2-reacted Portland case radiation length can be found using same formula and Table 2.

$$\frac{1}{X_0'} = \sum_{i=1}^{10} \frac{f_i'}{X_{0i}'} = 0.036956 \qquad X_0' = 27.05915\ cm \tag{8b}$$

So radiation length is increased by ~1.113%.

3. Results

In order to obtained energy deposition on the concrete, FLUKA USRBIN card is used [10,11]. Because of symmetry properties, yz dimension of 2D energy deposition plots have not been presented. Because of hadrons are interact with matter via hadronic process but leptons are interact with electromagnetic interaction four beam (photon, electron, proton and neutron) have been chosen.

3.1 Energy deposition using photon beam

The concrete used as the target is 20cm×20cm×30cm in size. This target is used for the 0.03 GeV photon beam energy. When photon beam interacts with target a cascade process is initiated as electromagnetically and loses its energy as electromagnetic shower where secondary particles are produced via pair production which is dominant process while incoming photon energy higher than 10 MeV. When the incoming photon energy is in between 1- 10 MeV Compton and photoelectric effect take place. In the low energy photon beam Rayleigh, Mie or Thomson scattering is one of possible process depending target properties. Two-dimensional energy depositions are given in Figure 1.

As seen in the figure, the energy deposited in the middle point of the target where photon beam hit. At this point, the target has the maximum value of 1 MeV/cm^2/pp energy deposition The energy deposited 1 keV/cm^2/pp on the value of 5 cm on both axis of conventional Portland case. On the other hand, this value decreases to 0.2 keV/cm^2/pp for CO_2-reacted Portland. It has been obtained that CO_2-reacted Portland absorbs radiation at a shorter range, where the cascade is reduced, compared to the conventional Portland case.

Figure 2 shows the 1-dimensional graph of energy deposition according to the length of the target. As seen in Figure 2 an exponential decrease in energy deposition up to the end of the target is observed in both the conventional Portland and CO_2-reacted Portland cases. In the case of CO_2-reacted Portland, lower radiation output from the target is obtained.

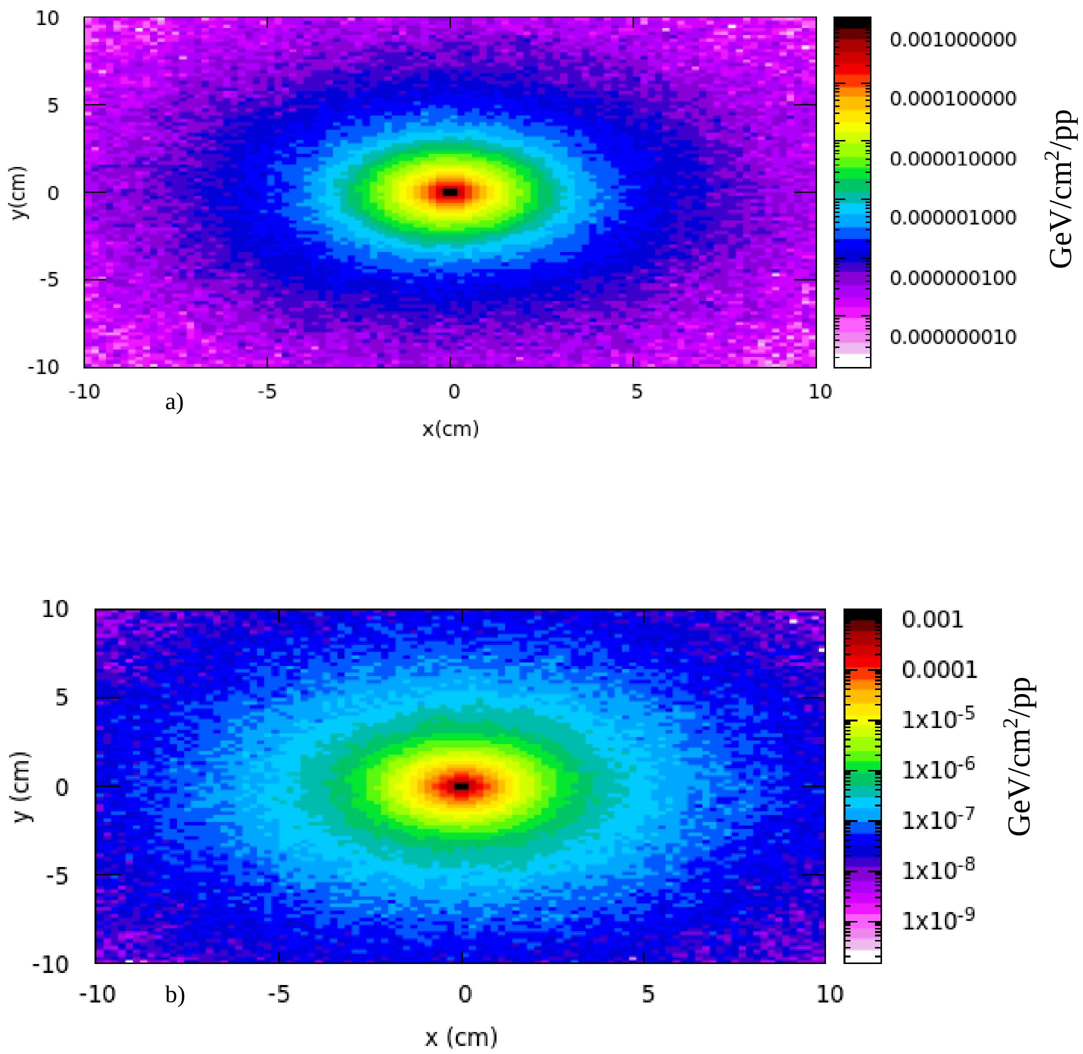


Figure 1. a) Conventional Portland, b) CO_2-reacted Portland(Port2), energy deposition in the xy axis obtained by photon beam

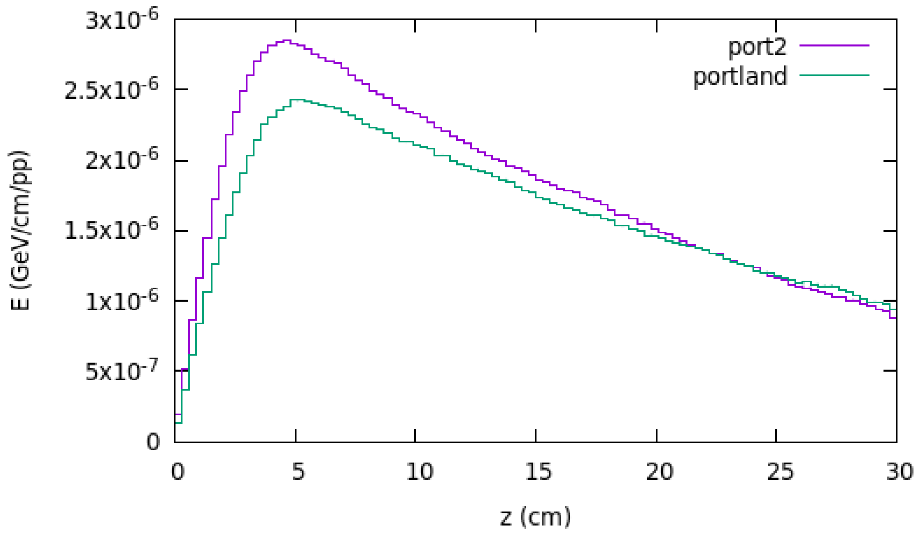


Figure 2. Energy deposition in the z-axis obtained by photon beam

3.2 Energy deposition using electron beam

The interaction of charged particles with matter takes place by taking the Coulomb interaction into account, unlike the interaction of photons. The interaction triggers a leptonic interaction. In this situation bremsstrahlung is the dominant process to produce secondary particles as photons. This photon continues to interact with target same as explained before until their energy is absorbed.

The target used is 20cm×20cm×30cm in size. This target is irradiated with 0.05 GeV energy electron beam. Two-dimensional xy energy deposition is given in Fig. 3. As seen in the figure, the energy deposited in the middle point of the target where electron beam hit. At this point, the target has the maximum value of 1 MeV/cm^2/pp energy deposition. The energy deposited 1 keV/cm^2/pp on the value of 5 cm on both axis of Portland case. On the other hand, this value decreases to 0.2 keV/cm^2/pp for CO_2-reacted Portland. It has been observed that CO_2-reacted Portland absorbs radiation at a shorter range compared to conventional Portland.

In Figure 4 Longitudinal energy deposition on the target is seen. It is understood from the figure that the produced electromagnetic shower has a peak value which is also shown from Figure 5 which is 1D graph of Figure 4 (energy deposition according to the length of the target). As seen in Figure 5, energy deposition has been obtained, in the case of CO_2-reacted Portland is14 keV/cm^2/pp at 4 cm thickness and 10 keV/cm^2/pp at 6 cm thickness in respectively. The Bragg peak is obtained at 4 and 5 cm of the target respectively which means that, less target thickness is required for CO_2-reacted Portland case in the case of using shielding material which can be seen from the Figure.

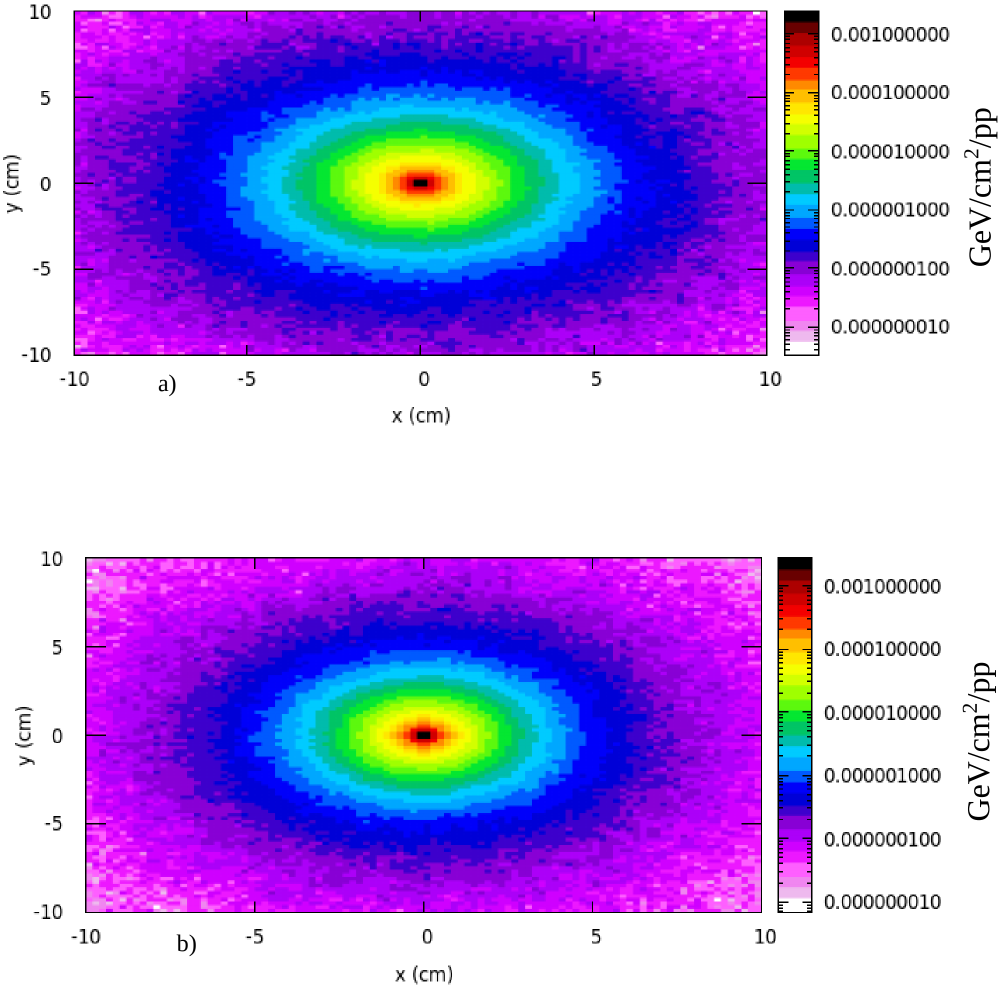
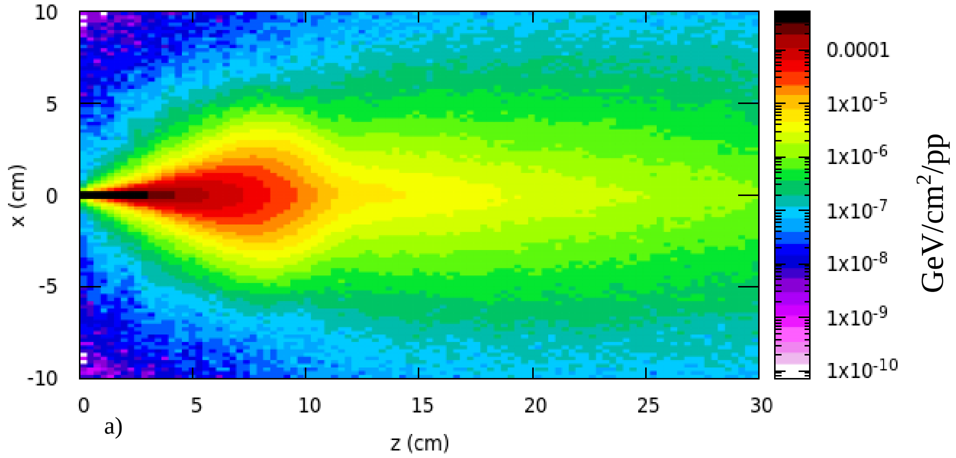


Figure 3. a)Conventional Portland, b)CO_2-reacted Portland (Port2), energy deposition in the xy axis obtained by electron beam



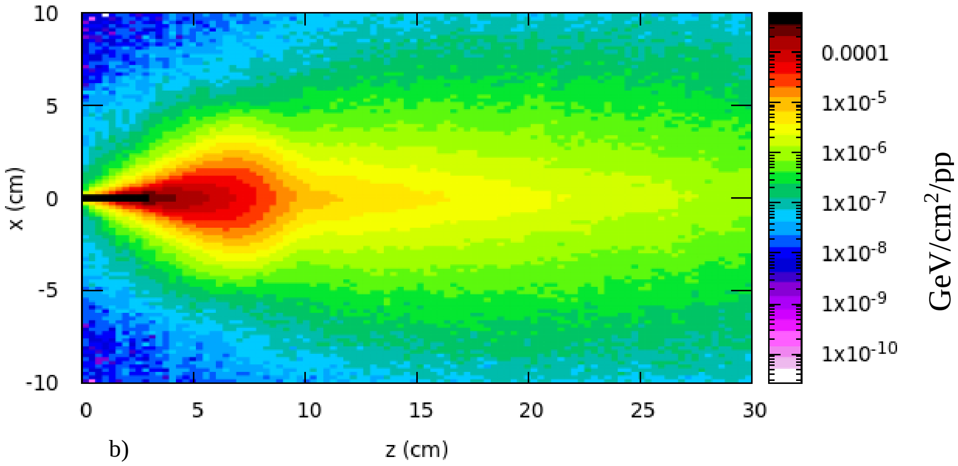


Figure 4. a)Conventional Portland, b)CO_2-reacted Portland (Port2), energy deposition in the zx axis obtained by electron beam

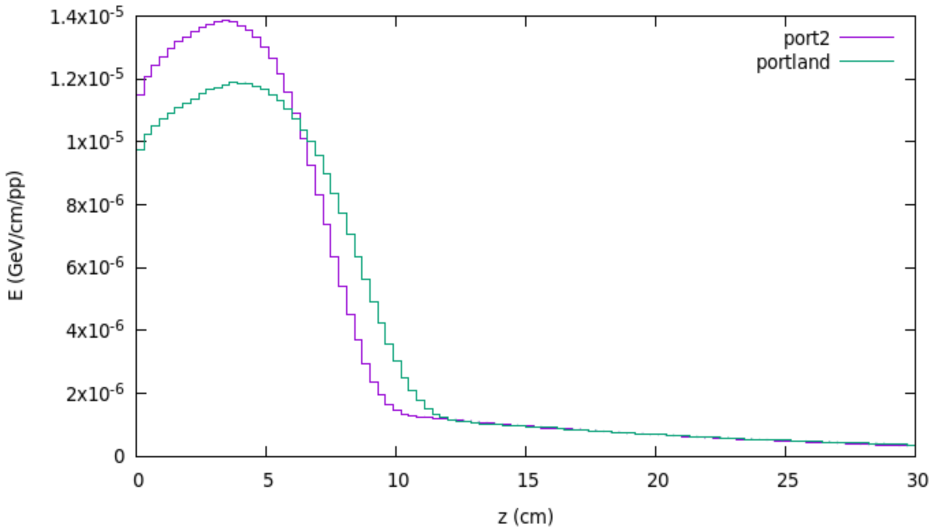


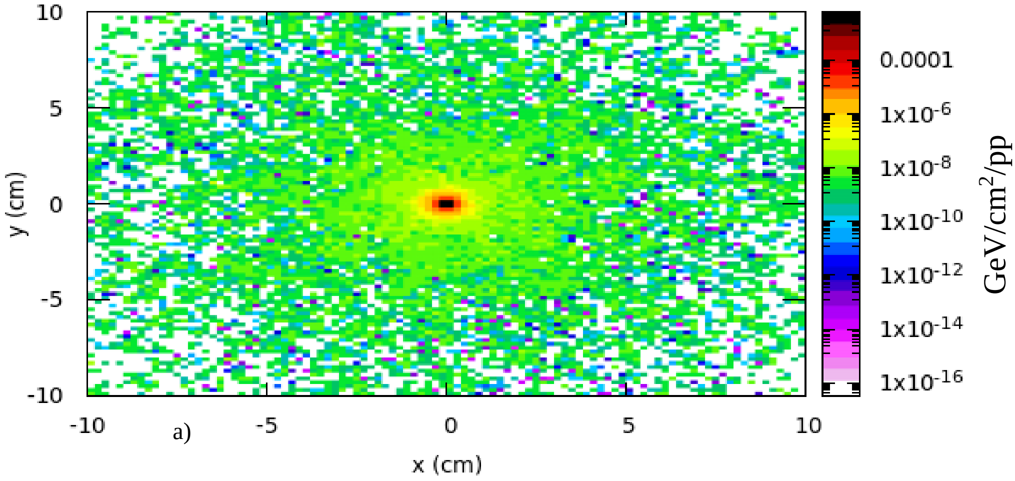
Figure 5. Energy deposition in the z-axis obtained by electron beam

3.3 Energy deposition using proton beam

The target used is 20cm×20cm×50cm in size. This target was used for the 0.05 GeV value of the proton beam energy. In this interaction hadronic shower is developed and since protons as heavy (comparing electron) and charged particle they will lose its energy in short range.

Two-dimensional energy depositions are given in Figure 6. As seen in the figure, the energy deposited in the middle point of the target where proton beam hit. At this point, the target has the maximum value of 1 MeV/cm^2/pp energy deposition. The energy deposited 0.02 keV/cm^2/pp on the value of 5 cm on both axis of Portland case. On the other hand, this value decreases to 0.01 keV/cm^2/pp for CO_2-reacted Portland on the 5 cm of both axis. It has been observed that CO_2-reacted Portland absorbs radiation at a shorter range compared to conventional Portland.

Figure 7 shows the 1-dimensional graph of energy storage according to the length of the target. As it can be seen in the Figure 7, energy deposition reaches to ~0.01 eV/cm^2/pp conventional Portland and CO_2-reacted Portland case respectively. The Bragg peak is obtained at around 1 cm of target.



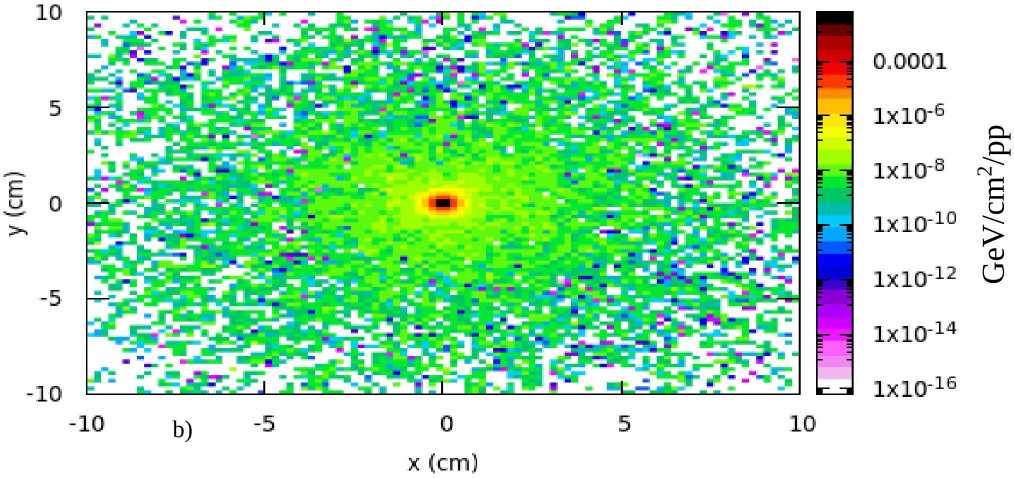


Figure 6. a)Conventional Portland, b)CO_2-reacted Portland (Port2), energy deposition in the xy axis obtained by proton beam

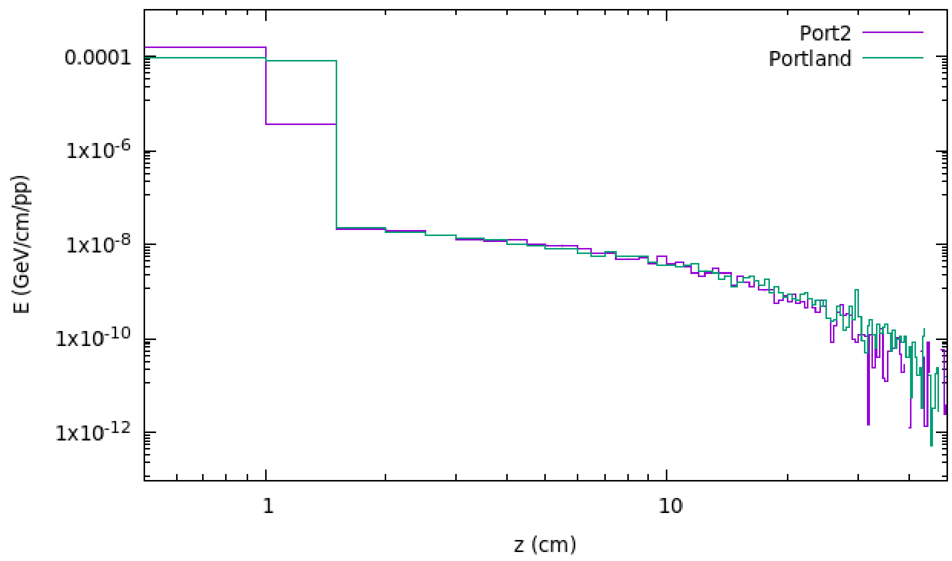


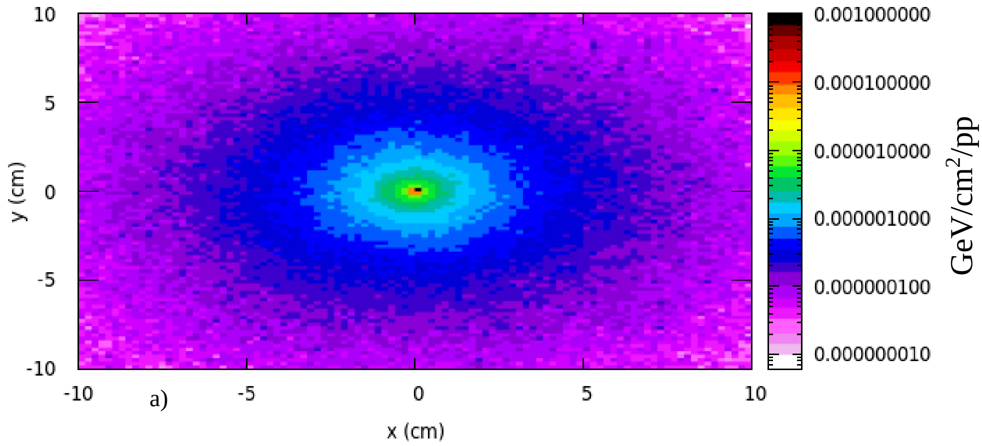
Figure 7. Energy deposition in the z-axis obtained by proton beam

3.4 Energy deposition using neutron beam

Neutrons are classified according to their energies. While slow (thermal) neutrons have ~0.025 eV, medium energy neutrons have 0.5-10 KeV, fast neutrons have 10 KeV-10 MeV and relativistic neutrons have more than 10 MeV energy. This target was used for 0.015 GeV of neutron beam energy which is in relativistic neutron beam regime. Thermal neutrons interact with target mostly radiative capture process by target nuclei. However, when the energy of neutrons increased to ~1 MeV than inelastic scattering take place in the nuclear reaction.

The target used is 20cm×20cm×50cm in size both conventional Portland and CO_2-reacted Portland case respectively. Two-dimensional energy deposition is given in Figure 8. As seen in the figure, the energy deposited in the middle point of the target where neutron beam hit. At this point, the target has the maximum value of 10 MeV/cm^2/pp. The energy deposited 0.01 keV/cm^2/pp on the value of 5 cm on both axis of conventional Portland case. On the other hand, this value decreases to 0.05 keV/cm^2/pp for CO_2-reacted Portland. It has been observed that CO_2-reacted Portland absorbs radiation at a shorter range compared to conventional Portland at the same thickness.

Figure 9 shows the 1D graph of energy storage according to the length of the target. As seen in Figure 9 Bragg peak is obtained on the target about 2 cm which is longer than proton beam, shorter than electron and photon beam. The energy deposition after the Bragg peak exponentially decrease till to end of target.



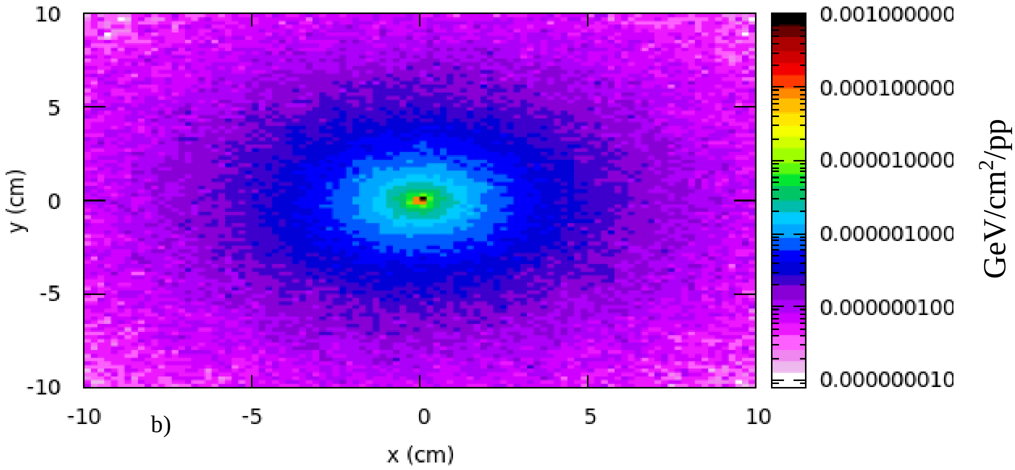


Figure 8. a)Conventional Portland, b)CO_2-reacted Portland (Port2), energy deposition in the xy axis obtained by neutron beam

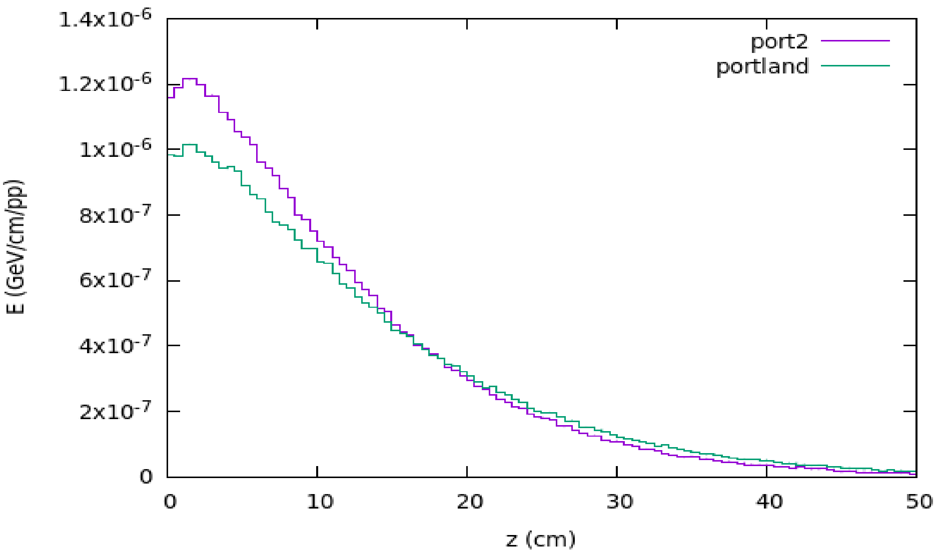


Figure 9. Energy deposition in the z-axis obtained by neutron beam

4. Conclusion and Comments

Radiation shielding properties of CO_2-reacted Portland is examined. It has been found that it is possible to obtained better radiation shielding properties than conventional one with producing less greenhouse gaseous effect than conventional Portland.

- Energy deposition on CO_2-reacted Portland using 30 MeV photon beam energy deposition decrease dramatically in 30 cm, because of photons are chargeless and massless particle they will interact mostly with target electrons and interaction length is longer than electron and proton beam case.
- Using 50 MeV electron beam, energy deposition will occur short distance than photon beam interaction because of it is a charged and light particle, it can be absorbed short distance (10 cm) than chargeless particle because of electromagnetic interaction. Energy deposition drops to 2 keV at 8 cm in CO_2-reacted Portland, however this value is 12 cm in conventional Portland case.
- Protons are heavy and charged particle and interact with hadronically with matter and 50 MeV proton beam has been absorbed in a 10 cm distance. Portland case has 2 cm to drops energy deposition drop the value in keV, however CO_2-reacted Portland has 1 cm to get the same amount energy deposition.
- In the case of 15 MeV neutron beam, Portland and CO_2-reacted Portland have absorbed the beam in 20 cm. For the neutron beam the heavy concrete should be used, because neutrons are chargeless and more penetrable than proton and electron beams.
- It has been found that the produced concrete density is 2.694 g/cm^3 and it is radiation length is increased by 1.113% comparing with conventional concrete.

Author Statement

Vildan Çinğili: Research, Analyzing, Original draft writing
Hüsnü Aksakal: Research, Verification, Examination, Checking, Correction and Arrangement

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As the authors of this study, we declare that we do not have any support and thank you statement.

Conflict of Interest

As the authors of this study, we have no conflict of interest to declare.

Ethics Committee Approval and Informed Consent

As the authors of this study, we declare that we do not have any ethics committee approval and/or informed consent statement.

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