BJRS
BRAZILIAN JOURNAL OF RADIATION SCIENCES

doi.org/10.15392/2319-0612.2025.2942
Braz. J. Radiat. Sci., Rio de Janeiro
2025, 13(4) | 01-17 | e2942
Editor: Prof. Dr. Bernardo Maranhão Dantas
Editor: Prof. Dr. Alfredo Lopes Ferreira Filho
Submitted: 2025-05-20
Accepted: 2025-08-06

Radionuclides Leaching Assessment and External Exposure on Building Materials Containing NORM Residue

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Abstract: Brazil's titanium dioxide production generates approximately 30,000 tons of waste annually, known as unreacted ore waste (UOW), which is currently disposed of in industrial landfills. This residue is enriched with naturally occurring radionuclides from the uranium and thorium decay series and is classified as Naturally Occurring Radioactive Material (NORM). The reuse of NORM residues in building materials is only permissible if the activity concentrations of radionuclides in the final product do not pose any additional exposure risk to individuals. Radiation exposure from such materials may be classified as external—due to direct gamma irradiation—or internal—resulting from inhalation of radon (^{222}Rn) and its short-lived decay products. In a previous study, the authors assessed both internal and external **indoor** exposure associated with incorporating up to 23% of UOW into cement and interlocking blocks. This study evaluates the feasibility of using these materials in **outdoor** applications, with particular emphasis on radionuclide leaching potential and external radiological exposure. Cement and interlocking blocks containing varying proportions of unreacted ore waste (0%, 3.5%, and 23%) were subjected to weathering for 60 days. The activity concentrations of ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{228}Ra, and ^{40}K in the leachate were measured using gamma spectrometry and spectrophotometry. No significant increase in radionuclide concentrations were observed in the leachate from blocks containing 3.5% and 23% UOW compared to those without any residue. Gamma radiation exposure remained consistently below 0.1 mSv, and no notable changes were detected in radon concentrations in the vicinity of the experiment. The results indicate that incorporating up to 23% UOW in cement and interlocking blocks is safe and compliant with regulatory limits related to groundwater contamination and human exposure when using these materials in an open air environment.

Keywords: Safe reuse of NORM residue; construction materials; radionuclide leaching; sustainability, circular economy.

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Avaliação da Lixiviação de Radionuclídeos e Exposição Externa em Materiais de Construção Contendo Resíduos NORM

Resumo: A produção de dióxido de titânio no Brasil gera aproximadamente 30.000 toneladas de resíduos anualmente, conhecido como resíduo de minério não reagido (UOW), que atualmente são descartados em aterros industriais. Este resíduo é enriquecido com radionuclídeos naturais da série de decaimento de urânio e tório e é classificado como Material Radioativo de Ocorrência Natural (NORM). A reutilização de resíduos NORM em materiais de construção só é permitida se as concentrações de atividade dos radionuclídeos no produto final não representarem qualquer risco adicional de exposição para os indivíduos. A exposição à radiação de tais materiais pode ser classificada como externa - devido à irradiação gama direta - ou interna - resultante da inalação de radônio (^{222}Rn) e seus produtos de decaimento de curta duração. Em um estudo anterior, os autores avaliaram a exposição *indoor* associada à incorporação de até 23% de UOW em cimento e blocos intertravados. Este estudo avaliou a viabilidade do uso desses materiais em aplicações externas, com ênfase particular no potencial de lixiviação de radionuclídeos e exposição radiológica externa. Blocos de cimento e intertravados contendo proporções variadas de rejeito de minério não reagido (0%, 3,5% e 23%) foram submetidos ao intemperismo por 60 dias. As concentrações de atividade de ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{228}Ra, e ^{40}K no lixiviado foram medidas por espectrometria gama e espectrofotometria. Não foi observado aumento significativo nas concentrações de radionuclídeos no lixiviado de blocos contendo 3,5% e 23% de UOW em comparação com aqueles sem nenhum resíduo. A exposição à radiação gama permaneceu consistentemente abaixo de 0,1 mSv, e nenhuma mudança significativa foi detectada nas concentrações de radônio nas proximidades do experimento. Os resultados indicam que a incorporação de até 23% de UOW em blocos de cimento e de intertravados é segura e está em conformidade com os limites regulatórios relacionados à contaminação das águas subterrâneas e à exposição humana ao usar esses materiais em ambiente ao ar livre.

Palavras-chave: Reutilização segura de resíduos NORM; materiais de construção; lixiviação de radionuclídeos; sustentabilidade, economia circular.

1. INTRODUCTION

The use of NORM (Naturally Occurring Radioactive Material) residues as a component of building materials is only possible if the activity concentration of radionuclides in the final product does not cause any additional risk of exposure to individuals. The policy that has been adopted to stack NORM residues generates, in the long run, high maintenance and safety costs, in addition to causing environmental impact. To minimize the environmental impact of this type of waste disposal, it is necessary to offer alternatives for its safe reuse, aligned with the needs of the Circular Economy and that contribute to the delivery of the Sustainable Development Goals (SDGs) (IAEA, 2013)[1]. A possible alternative would be to use this waste as a component of building materials.

The use of construction materials containing NORM residue can offer significant advantages in terms of sustainability, including reducing energy consumption and decreasing the need for mining natural resources. However, for these materials to be considered safe for reuse, it is important to perform a comprehensive assessment that involves both the environmental safety and durability of the resulting products.

The reuse of NORM residues in building materials is only permissible if the activity concentrations of radionuclides in the final product do not pose any additional exposure risk to individuals. Radiation exposure from such materials may be classified as external — due to direct gamma irradiation — or internal — resulting from inhalation of radon (^{222}Rn) and its short-lived decay products. The study by Van der Sloot et al. (2017)[2] highlights the importance of considering the entire life cycle of building materials containing NORM, from production to disposal. The leaching, defined as the release of constituents of a solid material into the aqueous phase when in contact with water, is also a critical aspect to evaluate. This process can result in direct human exposure to radiation, radon emission, and groundwater

contamination, making it necessary to develop appropriate test methods to predict leaching behavior under different uses and disposal scenarios.

The production process of titanium dioxide; the most commonly used white pigment, usually involves one of two alternative processes, called "chloride" and "sulphate" routes, generating undissolved ilmenite mud. The radionuclides' concentration is in many cases sufficiently high for these residues to be classified as NORM residues (IAEA, 2013)[1]. Several studies are found in the literature that deal with the recovery of ilmenite mud residues in the manufacture of commercial ceramics and sulfur polymer cement (Gázquez et al. 2011; Contreras et al., 2013; Contreras et al., 2014)[3][4][5].

In the Brazilian production of titanium dioxide by the sulfation method, about 60,000 tons of TiO_2 are generated annually, with the consequent production of 30,000 tons of residue known as UOW (unreacted ore waste), which is disposed of in industrial landfills. During the sulfation process, uranium and thorium are soluble in sulfuric acid, and therefore the bulk of these elements are found in the liquor, while radium isotopes, with low solubility, accumulate in the residue. The Brazilian titanium dioxide production is classified as NORM due to the levels of radioactivity present in the residue, 538±103 Bq.kg^{-1} of ^{238}U, 1103±115 Bq.kg^{-1} of ^{226}Ra, 960±166 Bq.kg^{-1} of ^{210}Pb, 400±82 Bq.kg^{-1} of ^{232}Th and 2906±262 Bq.kg^{-1} of ^{226}Ra (Mazzilli et al., 2022)[6].

Within the scope of radiation protection, the Brazilian Regulatory Agency published in 2016 the standard CNEN-NN-4.01 – Requirements of Safety and Radiation Protection for mining and milling facilities (CNEN, 2016)[7]. This standard establishes a regulatory approach for NORM facilities, including storage and management of radioactive waste. However, at present, there is no specific regulation in Brazil concerning the re-use of NORM residues, except for the standard that deals with phosphogypsum residue generated in the phosphate fertilizer industry (CNEN, 2014)[8].

Several papers were published in Brazil concerning the recovery of the residue from the titanium dioxide production as building materials. Ribeiro et al. (2021)[9] investigated the effect of the addition of UOW residue from the Brazilian production of TiO_2 on the properties of coating mortars and evaluated its radiological impact. Mazzilli et al. (2025)[10] evaluated the radiological exposure of using different percentages of UOW residue as a component of interlocked and cement blocks. The authors assessed both internal and external ***indoor*** exposure associated with incorporating UOW residue into cement and interlocking blocks. Their results indicated that incorporating up to 23% of the residue does not elevate indoor radon concentration above the international limit of 200 $Bq.m^{-3}$ and keeps external gamma exposure below 1 mSv. As a complementary study, this paper evaluates the feasibility of using these materials in ***outdoor*** applications, with particular emphasis on potential leaching of radionuclides and external radiological exposure. Cement and interlocking blocks containing varying proportions of unreacted ore waste (0%, 3.5%, and 23%) were subjected to weathering for 60 days. Leaching assessment is important to evaluate the amount of radionuclides that are being released into the environment that may eventually contaminate groundwater. The main purpose of this study is to control and prevent radiological problems in future large-scale applications of these materials in open air environment.

Several steps were taken to ensure the safety and feasibility of using this NORM residue in construction materials. First, the radionuclides concentration was measured in the cement and interlocking blocks with 0%, 3.5% and 23% of UOW residue. The leaching assessment was conducted to evaluate the release of radionuclides under adverse environmental conditions such as exposure to rain and wind. In addition, measurement of radon emission was carried out to understand the risks of inhalation and the gamma exposure was monitored in the vicinity of the experiment, allowing the evaluation of the outdoor exposure.

2. MATERIALS AND METHODS

To perform the leaching assessment, three interlocking structures and three block structures were constructed, each with a different percentage (0%, 3.5% and 23%) of UOW, as illustrated in Figure 1 for the interlocking blocks and Figure 2 for the cement blocks. A retention tank was built under the interlocking blocks and cement blocks for the collection of leached water, which flows through PVC pipes to a water collection station for the characterization of the radionuclides of interest.

Figure 1: Outdoor experiment using interlocked blocks with different percentages of UOW

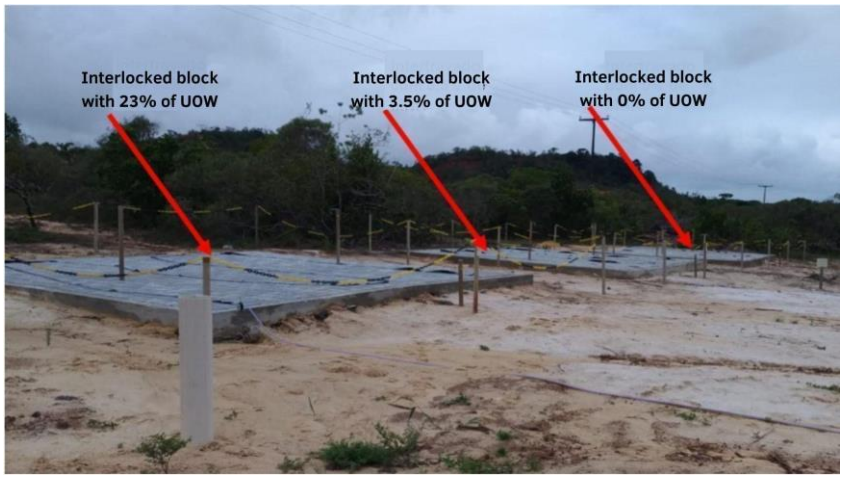


Figure 2: Outdoor experiment using cement blocks and preparation of the rainwater collecting system.

The choice of the experimental site considered the predominant wind direction and the slope of the terrain, with the test with 0% UOW located upstream, not being affected by the other two tests with 3.5% and 23% UOW, respectively. The test without the addition of UOW was performed to determine the background level of radioactivity. The monthly rainfall at the experiment site shows significant seasonal variation, with May being the wettest month, averaging 159 millimeters of precipitation, and January the driest, with an average of just 46 millimeters. In periods without rain, it was possible to irrigate the blocks with a flow rate of up to 2 thousand liters of water, to assess the most critical scenario. Leachate sampling was performed after 30 days and 60 days.

The determination of the activity concentration of natural radionuclides of radiological importance ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{228}Ra and ^{40}K was carried out in the cement and interlocking blocks with 0%, 3.5% and 23% UOW. For the analysis of the samples, gamma spectrometry was used to measure the activity concentration of ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{40}K. The detector used was a Hyper Pure Germanium detector (HPGe) GX2518, with 25% relative efficiency, from CANBERRA. The counting time was 86,000 seconds. The determination of U and Th was carried out using spectrophotometric techniques, employing a SHIMADZU UVMini-1240 spectrophotometer. Uranium determination is based on its extraction using tri-n-butyl phosphate (TBP) in an $Al(NO_3)_3$ medium, which prevents interference from phosphate and fluoride. EDTA is used to complex certain metal ions and remove them prior to uranium extraction, while tartaric acid masks the presence of zirconium. The complex formed between uranium and Arsenazo(III) displays a reddish-violet color, with an optimal absorption measurement at a wavelength of 650 nm. For thorium determination, its extraction was performed using tri-n-octylphosphine oxide (TOPO) in a cyclohexane medium, effectively removing interfering species such as iron(III) and titanium(IV). However, zirconium—an important interferant—remains in the extract and is removed using oxalic and ascorbic acids. To prevent the formation of thorium precipitates or complexes as fluoride, sulfate, or phosphate, the determination is conducted

in a strongly acidic medium. The thorium-Arsenazo(III) complex exhibits maximum absorption around 665 nm.

The radon concentration and the gamma exposure were monitored close to the experiment to evaluate the exposure of individuals. Radon measurements were conducted using a passive method with solid-state nuclear track detectors (CR 39 - Columbia Resin #39) and the external gamma exposure by using Thermoluminescent Dosimeters - TLDs). Both sets of measurements were performed over a three-month period.

The environmental monitoring included the measurement of radon and the gamma exposure in the air at 11 points, six points in the interlocking block system, two in each material tested (point 7 and 8 for 0% of UOW, 9 and 10 for 3.5% of UOW and 11 and 12 for 23% of UOW, in Figure 3) and three points in the cement block system (points 13, 14 e 15 in Figure 4). Measurements of radon and gamma exposure in the air were also performed in the soil near the experiments to monitor the background (points 3 and 4 for Test B.2 and points 5 and 6 for Test B.3).

Figure 3: Collecting points for the determination of radon concentration and gamma exposure in the interlocked block experiment

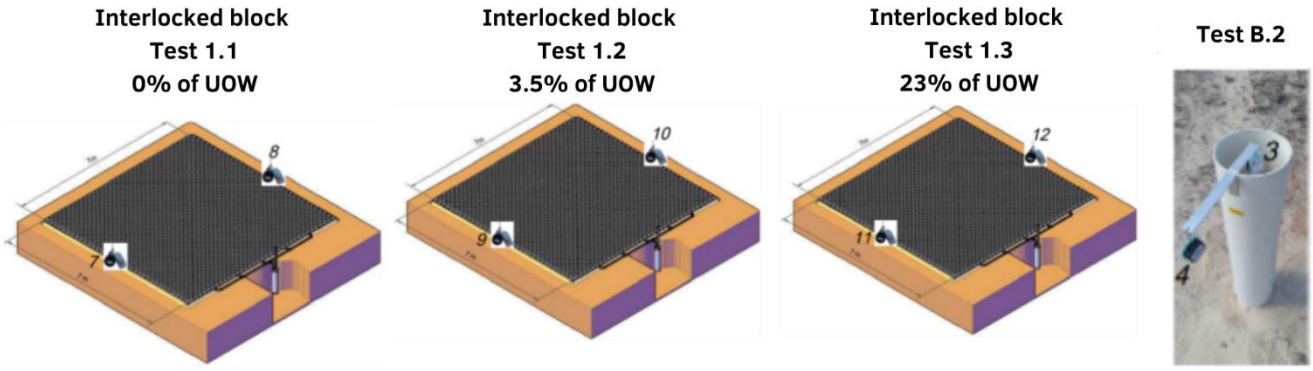
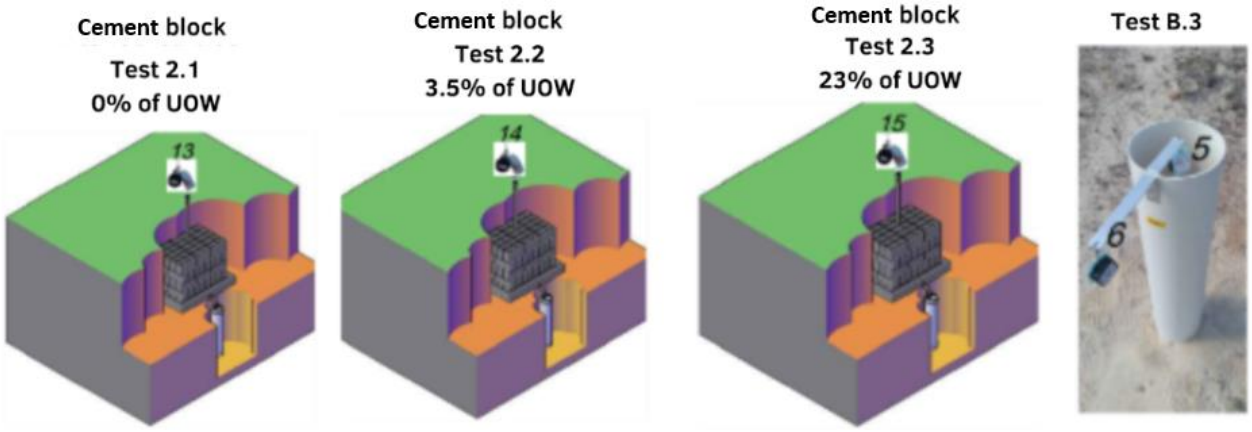


Figure 4: Collecting points for the determination of radon concentration and gamma exposure in the cement block experiment



3. RESULTS AND DISCUSSIONS

The results of the activity concentration of ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{228}Ra and ^{40}K in cement blocks and interlocking blocks with different proportions (0%, 3.5% and 23% of the residue) are presented in Table 1 and Figure 5. The interlocked and cement blocks with no addition of residue presented activity concentration of ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K of the same order of magnitude of the world-wide average concentrations in the earth's crust reported by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000) [11]. The addition of the residue will proportionally increase these values, especially for ^{226}Ra, ^{210}Pb, ^{232}Th and ^{228}Ra. Potassium-40 seems to be evenly distributed among all components of the blocks studied.

Table 1: Concentration of radionuclides (Bq.kg^{-1}) in the cement block and interlocked block with 0%, 3.5% and 23% of UOW.

SAMPLE	U-238	Ra-226	Pb-210	Th-232	Ra-228	K-40
Interlocked block 0%UOW	<43.5	<6.5	<13.5	22	37	578
Interlocked block 3.5%UOW	58	27	31	49	120	845
Interlocked block 23%UOW	51	100	41	78	255	893
Cement block 0%UOW	<43.5	7	17	18	39	714
Cement block 3.5%UOW	<43.5	25	22	41	116	920
Cement block 23%UOW	58	129	70	57	381	895
Earth's crust [11]	35	35	-	30	-	400

Figure 5: Concentration of radionuclides in the cement and interlocked blocks and average earth's crust values from UNSCEAR [11] (Bq.kg^{-1})

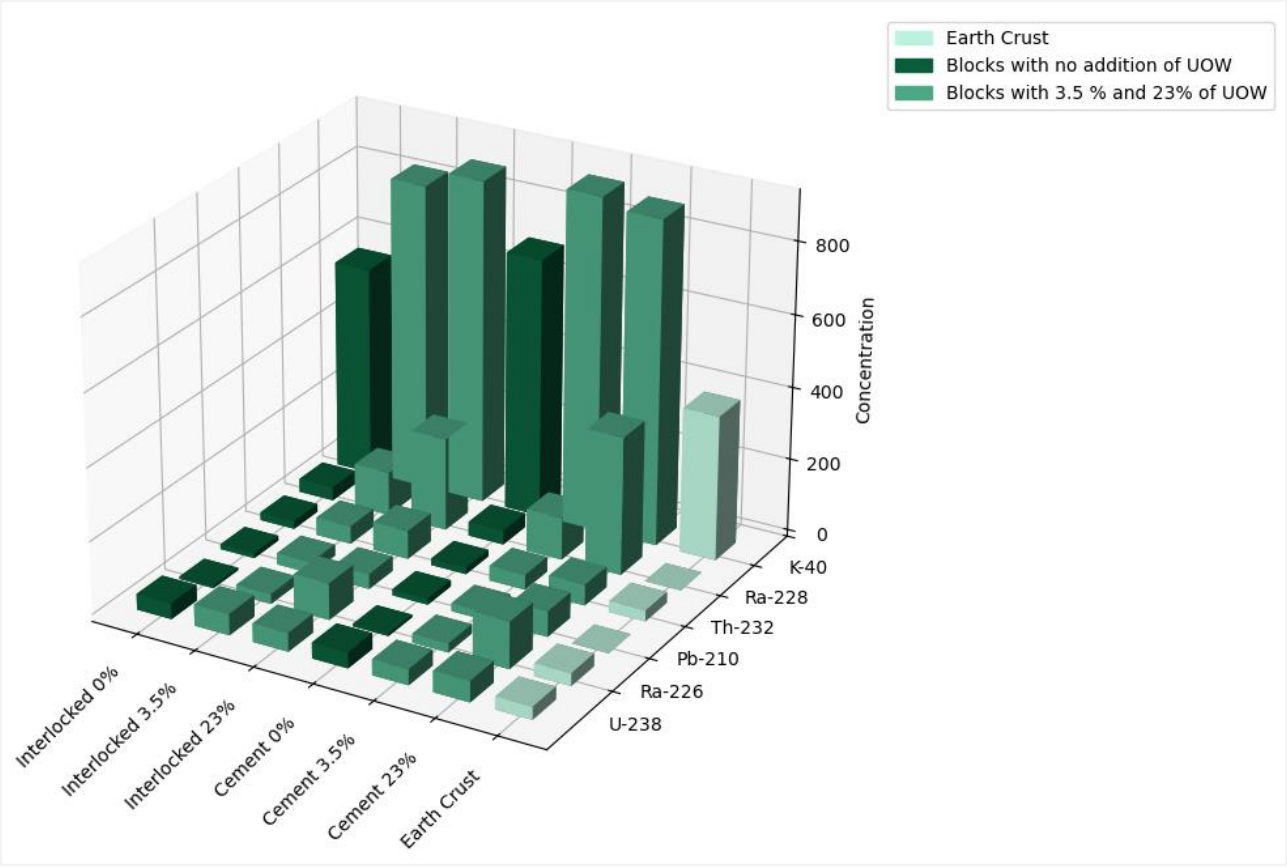


Table 2 and Table 3 present the results of radionuclide concentration in leaching water samples from the interlocked and cement block experiments, collected after 30 and

60 days of exposure. No significant difference was observed in the concentration of the radionuclides in the leaching water collected after 30 and 60 days of experiment (Figure 6 and Figure 7). The results obtained for the concentration of radionuclides (^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{226}Ra and ^{40}K) in the leachate of cement and interlocking blocks samples without the addition of the residue can be considered as reference values. No significant increases were also observed in the concentration of radionuclides in the experiments carried out without the addition of residue and with the addition of 3.5% and 23% of residue. It can be concluded that the leaching of radionuclides present in cement and interlocked blocks by the action of weathering does not pose a threat to the environment and does not affect the quality of groundwater.

Table 2: Activity concentration of radionuclides in the lixiviation water collected after 30 and 60 days of exposure (Bq.L^{-1}) in the interlocked block experiment

Radionuclide	Interlocked block 0%UOW*	Interlocked block 3.5% UOW	Interlocked block 23% UOW	Interlocked block 0% UOW*	Interlocked block 3.5% UOW	Interlocked block 23% UOW
	30 DAYS OF LIXIVIATION			60 DAYS OF LIXIVIATION		
U-238	0.06	0.06	0.06	0.06	0.06	0.06
Ra-226	0.93	0.93	0.93	0.93	0.93	0.93
Pb-210	4.6	3.7	3.0	3.5	4.1	3.2
Th-232	0.01	0.02	0.03	0.01	0.01	0.01
Ra-228	1.08	1.08	1.53	1.08	1.08	1.08
K-40	9.4	9.8	10.7	3.0	4.5	4.8

Figure 6: Concentration of radionuclides in the lixiviation water from interlocked blocks collected after 30 and 60 days of exposure (Bq.L^{-1})

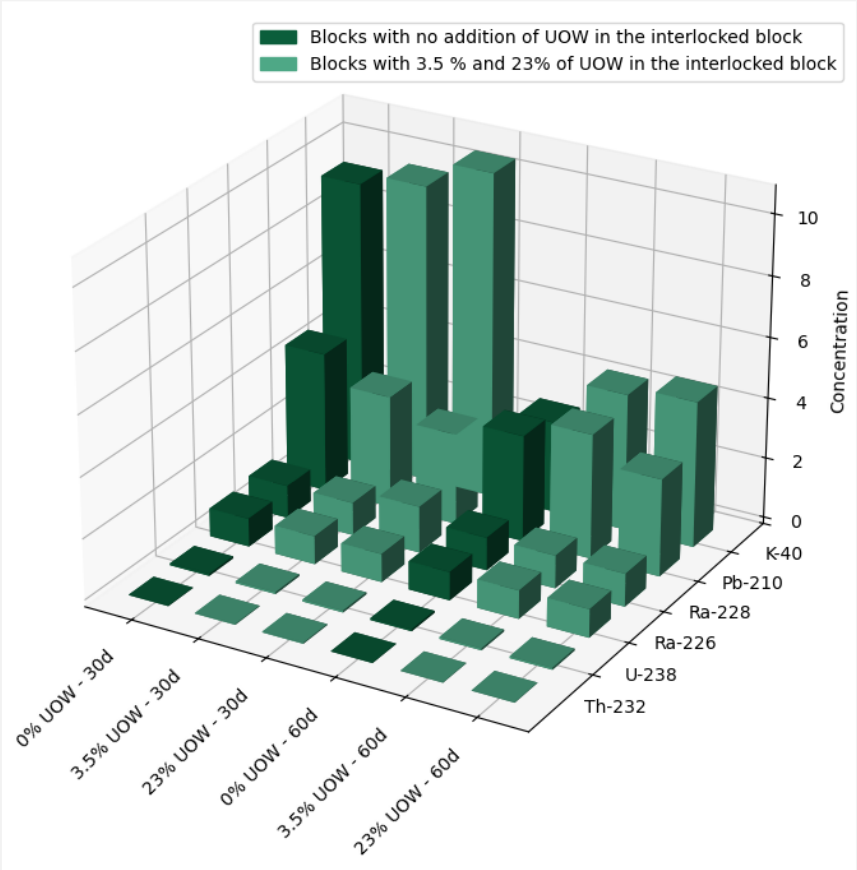


Table 3: Activity concentration of radionuclides in the lixiviation water collected after 30 and 60 days of exposure (Bq.L^{-1}) in the cement block experiment

Radionuclide	Cement block 0%UOW	Cement block 3.5% UOW	Cement block 23% UOW	Cement block 0% UOW	Cement block 3.5% UOW	Cement block 23% UOW
	30 DAYS OF LIXIVIATION			60 DAYS OF LIXIVIATION		
U-238	0.06	0.06	0.06	0.08	0.07	0.08
Ra-226	0.01	0.02	0.04	0.01	0.04	0.02
Pb-210	0.14	0.14	0.14	0.14	0.14	0.14
Th-232	0.01	0.01	0.01	0.01	0.01	0.01
Ra-228	0.06	0.07	0.08	0.06	0.06	0.06
K-40	5.7	8.9	4.4	3.0	3.8	5.4

Figure 7: Concentration of radionuclides in the lixiviation water from cement blocks collected after 30 and 60 days of exposure (Bq.L^{-1})

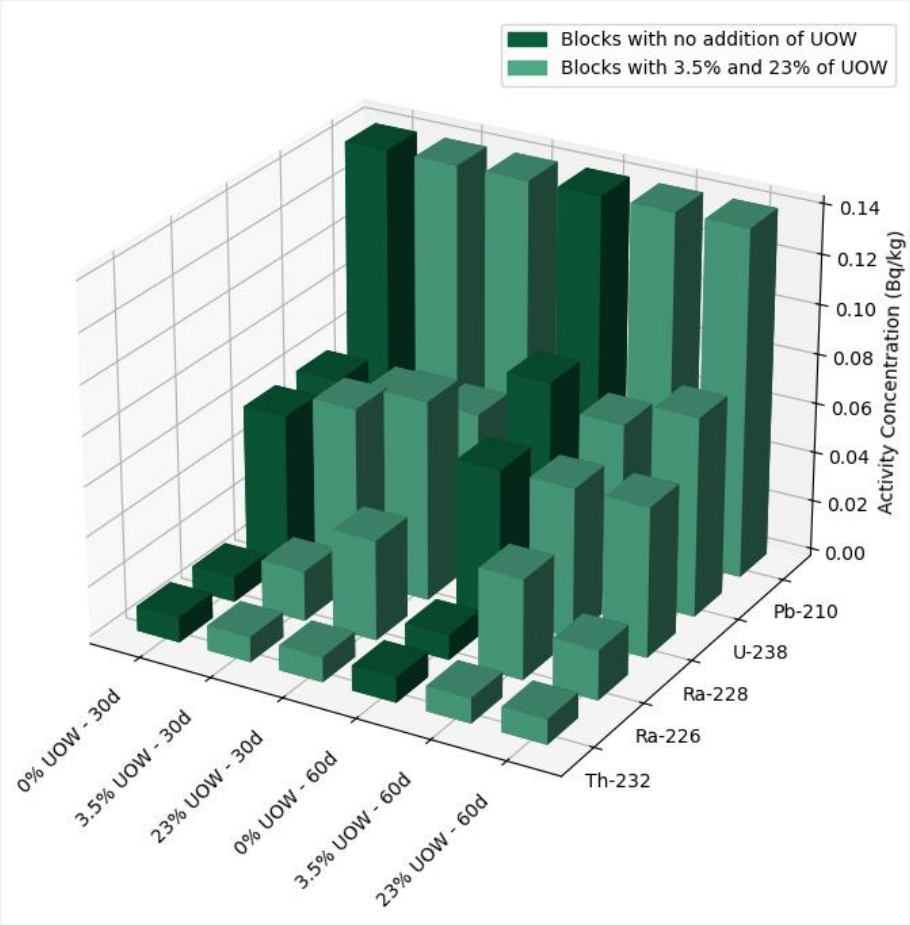


Table 4 presents the results of the activity concentration of the soil samples collected at points B2 and B3 close to the experimental area with the interlocked blocks and the cement blocks, respectively. The results obtained are of the same order of magnitude of the world average concentrations of radium, thorium and potassium in the earth's crust given by UNSCEAR (2000)[11]: 40 Bq.kg^{-1}, 40 Bq.kg^{-1} and 400 Bq.kg^{-1}, respectively.

Table 4: Activity concentration of K-40, U-238, Ra-226, Pb-210, Th-232 and Ra-228 (Bq.kg^{-1}) in soil samples collected close to the interlocked and cement block experiments

Radionuclide	Soil conc. B2	Soil conc. B3	Reference Value (UNSCEAR)
K-40	<30	<30	400
U-238	<43.5	75	35
Ra-226	<6.5	<6.5	35
Pb-210	<13.5	<13.5	-
Th-232	<16	<16	30
Ra-228	17	<11	-

The radon concentration and the gamma exposure were monitored close to the experiment to evaluate the exposure of individuals. The sampling points are depicted in Figure 3 and Figure 4. The gamma exposure was always below 0.1 mSv, indicating that the external exposure is negligible. The internal exposure due to inhalation can be evaluated through the radon concentration in the air. The results obtained for the radon concentration are presented in Table 5.

Table 5: Radon concentration (Bq.m^{-3})

Monitoring Point		Radon Concentration
Interlocked block 0%MNR	Point 7	14 ± 8
	Point 8	20 ± 6
Interlocked block 3.5%MNR	Point 9	12 ± 8
	Point 10	21 ± 8
Interlocked block 23%MNR	Point 11	13 ± 8
	Point 12	20 ± 8
Cement block 0%UOW	Point 13	27 ± 8
Cement block 3.5%UOW	Point 14	42 ± 8
Cement block 23%UOW	Point 15	59 ± 10
Background	Point 5	39 ± 10
	Point 6	20 ± 8
Background	Point 1	38 ± 10
	Point 2	18 ± 8

Measurements taken near experiments with blocks containing 0% UOW (points 7, 8, and 13) served as a baseline (blank) for comparison with data collected from experiments using blocks with 3.5% and 23% UOW (points 9, 10, 11, 12, 14, and 15). The baseline Rn concentration (mean 20.3 $Bq.m^{-3}$) was of the same order of magnitude as the background level (mean 28.7 $Bq.m^{-3}$). The lack of significant variation in Rn concentration between experiments with blocks without residue and those containing 3.5% and 23% residue confirms that no additional exposures occurred due to the use of the residue. In conclusion, the results indicate that incorporating up to 23% UOW in cement and interlocking blocks is safe and compliant with regulatory limits related to groundwater contamination and human exposure when using these materials in open air environment.

4. CONCLUSIONS

This study explored the feasibility of using the residue from the titanium dioxide production as a component of cement and interlocking blocks, in outdoor applications, emphasizing radionuclide leaching assessments and external radiation exposure. The findings from this case study provide valuable insights for developing national standards and guidelines that promote the safe reuse and management of this residue in construction materials, aligned with the sustainable principles and the Circular Economy.

ACKNOWLEDGMENT

This work was supported by Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq): Barbara Mazzilli- grant number 300835/95-7, Fernanda Yuri Muta- grant number 145651/2024-0 and Lara Vitoria Santos Couto-grant number 154057/2024 -0.

FUNDING

This work was supported by Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq): Barbara Mazzilli - grant number 300835/95-7, Fernanda Yuri Muta - grant number 145651/2024-0 and Lara Vitoria Santos Couto - grant number 154057/2024-0.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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