



## Article

# Mechanical, Structural, and Environmental Properties of Building Cements from Valorized Sewage Sludges

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**Abstract:** Building materials can enable the recycling of sewage sludge from tannery wastewater treatment by infiltration/percolation over coal and clay waste. The process avoids energy-intensive operations and yields a stable and environmentally friendly product. The sludge under study is mainly composed of SiO<sub>2</sub>, CaO, Al<sub>2</sub>O<sub>3</sub>, and Fe<sub>2</sub>O<sub>3</sub>, which is convenient to replace the mortar in cement. Different mortars were made by substituting a variable amount of sludge, from 0 to 30%, into the standard cement. The microstructure and mechanical properties of the mortar specimens were characterized after curing for 7 days and 28 days. The best properties were obtained with 15% sludge. Above 15%, the strength decreases at an early stage, as confirmed by SEM and XRD analysis, with more voids and ettringites at larger sludge content. The leaching tests of the mortar confirm that the cumulative values of heavy metals are far below the Deutsch regulatory limits (NEN 7043), justifying retention of the metals in the matrix. Radiological assessment of the sludge mortars also confirms their safety with the values of naturally occurring radioactive materials, surface radon exhalation and annual effective dose far below the required limits. The study suggests that 15% sludge can be used to sustainably replace cement and meet building safety requirement standards.

**Keywords:** sewage sludge; sludge mortar; recycling; mechanical properties; leaching; radioactivity



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## 1. Introduction

Industrial activity generates several types of environmentally hazardous waste [1–3]. Industrial by-products and wastes are widely used in the cement and concrete industry whose aluminosilicate wastes are used as supplementary cementitious materials (MCS) [4]. In addition, they can be used as raw materials to elaborate materials activated by alkalis such as geopolymers [5]. Conventional wastewater treatment plants involve coagulation, flocculation, sedimentation, filtration, and disinfection and they generate large amounts of residue or waste called sewage sludge, which is a serious management and environmental issue [6]. A recent study estimates that the industry in Morocco generates more than 1.6 million tons of solid waste per year [7]. Part of these residues come mainly from coal-fired thermal power plants (TAQA Morocco thermal power plant) of which solid waste is produced significantly as fly ash (90%) and bottom ash (10%) [7]. Storage of the unused residues raises two main problems: (i) the management and development of landfills and (ii) the contamination of water and soil by toxic pollutants such as heavy

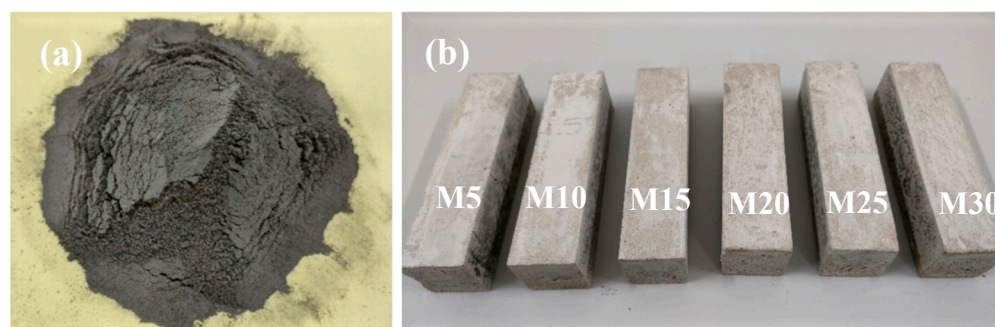
metals [8,9]. The present environmental concern requires careful consideration to be treated in an environmentally acceptable and sustainable manner. For this, a previous study focused on the valorization of coal wastes as adsorbents in tannery wastewater treatment by infiltration/percolation; as a result of this treatment, sewage sludge gets generated [10]. The addition of dried or calcined sewage sludge to the manufacture of mortar can become an effective alternative to existing management methods. The stabilization of sewage sludge with cement and sand additives improves the durability of the end product as compared to standard solutions [11].

This study focuses on the recovery of solid waste from the treatment of tannery wastewater by infiltration/percolation using coal fly ash and bottom ash generated in the Jorf-Lasfar thermal power plant (Morocco), combined with bentonite clay and coastal sand. Tannery sludge contains high concentrations of heavy metals such as Cr, As, Ni, Zn, Fe, and Cd, due to the use of chromium, inert solids, dyes, organics, and other chemicals during the process [12]. Therefore, it is necessary to find alternative solutions to manage these sludges and reuse them effectively, like in the construction industry. Meanwhile, these wastes when used as cementitious binders can constitute one of the most effective means of stabilizing numerous pollutants by immobilization during the hydration process. Note that the present sewage sludge is mineralogically similar to clay and Portland cement, containing mainly major oxides such as  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{CaO}$ , and  $\text{Fe}_2\text{O}_3$ . Several studies addressed the production of building materials from sewage sludges, including lightweight aggregate, bricks, tiles, and other construction products [13–15]. Our approach offers an alternative method for sludge recycling and environmental resources saving [16]. In this context, this study proposes the reuse of the sludge generated after treatment of tannery wastewater in building materials and encourages further study of the mechanical properties and environmental implications.

Mortar specimens were prepared with different proportions of sludge using Portland Pozzolona Cement (PPC) as binder. Their suitability as an engineering material was assessed based on mechanical properties. The environmental assessment was carried out in terms of metal release into the surrounding environment by leaching tests. Radiation risk due to natural radioactivity in sludge mortars was also assessed with the radon-222 activity (Rn-222), which is considered as secondary and as indoor radiological parameters. This offers a strategy for managing and reusing sewage sludge.

## 2. Materials

The mortars were prepared from a mixture of sewage sludge, cement, sand, and water. Sewage sludge is generated after tannery wastewater treatment using coal fly ash (30 wt%), bottom ash (30 wt%), bentonite clay (20 wt%), and sand (20 wt%) as adsorbents in infiltration/percolation system by column. The sludge samples were dried at 105 °C, crushed and then directly used as cement substitute. Figure 1a shows the visual appearance of the sewage sludge used.



**Figure 1.** (a) Sewage sludge, and (b) mortar specimens with varying sludge content.

The cement used in the preparation of the mortars is purchased as CEM II (CPJ45), composed of 68% clinker, 27% limestone, and 5% gypsum [17]. Its chemical composition

and physical characteristics are given in Tables 1 and 2, respectively. The sand samples were taken from Essaouira Beach (Morocco) sieved at 200  $\mu\text{m}$  to obtain a homogeneous particle size, and to homogenize the fraction of the pasty mixture. Its chemical composition is also given in Table 1.

**Table 1.** Chemical composition of cement and sand.

	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	K <sub>2</sub> O	CaO	SO <sub>3</sub>	TiO <sub>2</sub>	MgO	LOI *
Cement	17.77	6.00	3.00	1.23	63.00	3.35	-	2.65	11.90
Sand	42.53	2.76	1.65	0.44	25.01	-	0.29	1.79	4.60

\* Loss on ignition.

**Table 2.** Physical characteristics of the cement.

Initial setting time	180 min
Final setting time	210 min
Specific gravity	3.15 t/m <sup>3</sup>
Blaine fineness	304 m <sup>2</sup> /Kg
Compressive strength	7 days under 30 MPa—28 days under 40 MPa

The evaluation of the mechanical strength, durability and microstructure of cement sludge-based binder systems requires careful preparation of the mortar specimens with different sludge contents according to European standard NF EN 196-1 [18]. The water/cement ratio directly affects the strength and durability of the concrete. The typical water:cement ratio is 0.5 for different grades of concrete mix. The prisms are molded for the first 24 h and then the mortar prisms are cured at room temperature (20 °C). With a cement/sand ratio of 1:3, different mortars are prepared by partially replacing the cement with sewage sludge in 5, 10, 15, 20, 25, and 30% proportions. The corresponding mixtures are designated M5, M10, M15, M20, M25, and M30, respectively, and the control mortar is designated M0. Details of the cement sludge mortar mixtures are given in Table 3.

**Table 3.** Proportions of cement sludge mortars; the quantities of sand and water are set constants for all mortars at 1350 g and 225 g, respectively.

	M0	M5	M10	M15	M20	M25	M30
Cement (g)	450.0	427.5	405	382.5	360.0	337.5	315.0
Sludge (g)	0.00	22.5	45.0	67.5	90.0	112.5	135.0
Sludge %	0	5	10	15	20	25	30

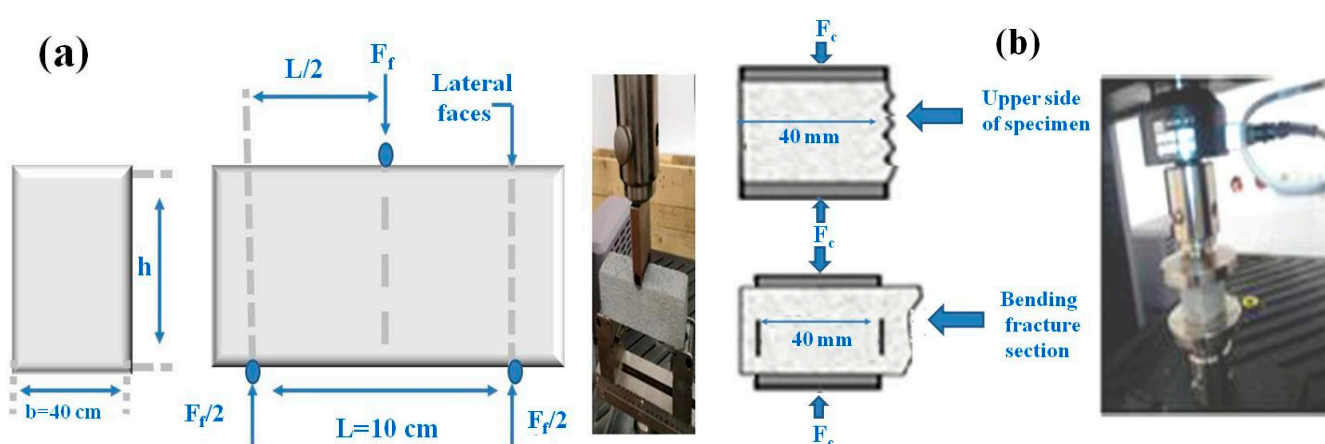
The components of each mortar are mixed for 4 min at room temperature according to the European standards method [19]. The mixture is placed in prismatic molds of  $4 \times 4 \times 16 \text{ cm}^3$  and freshly compacted using a table vibrator [20]. The compacted specimens are removed from the mold after 24 h and subsequently cured for two periods of 7 and 28 days (Figure 1b).

### 3. Methods

The sludge was analyzed before usage by wavelength dispersive X-ray fluorescence (WDXRF) spectroscopy (Axios-type). The particle size was determined by laser granulometry using water as a dispersing agent in MASTERSIZER 2000 MALVERN device. The metals (Cr, As, Pb, Cd, Ni, Cu, and Zn), whether contained in the sludge or leached from the mortars, were analyzed by ICP-AES.

The structure and microstructure of the mortars were studied by X-ray diffraction (XRD) and scanning electron microscopy (SEM). XRD was recorded using An X'Pert PRO Panalytical type diffractometer at diffraction angle of  $2\theta$  (degree) in the range of  $2^\circ$ – $70^\circ$  by steps of  $0.013^\circ$ .

SEM images of ground specimens were taken using a QUATTRO S-FEG-Thermo Fisher science microscope. The flexural and compressive strengths were determined using the MTS Criterion Electromechanical Press (Model 43) based on the three-point bending (TPB). Experiments were carried out in triplicate and the average values obtained were compared to those recorded for the reference sample. The breaking load was determined by applying a progressive force  $F_f$  on the upper face of the specimen at a constant speed of 2 mm/min until the specimen broke, as shown in Figure 2a [19]. The breaking strength was then determined. The flexural rupture strength  $R_f$  is given as:  $R_f = \frac{3F_f \cdot L}{2b \cdot h^2}$ , in which  $F_f$  is the applied force at the fracture point;  $L$  is the distance between the supports (mm), and  $b$  and  $h$  are the width and the height of the sample, respectively, as shown in Figure 2. After the flexural tests, the two half-prisms were used to measure the compressive strength  $R_c$  (Figure 2b) according to the formula:  $R_c = \frac{3F_f}{b \cdot h}$  [20]. A progressive force was applied on the transverse section of  $4 \times 4 \text{ cm}^2$  of the sample with the same speed until breaking, yielding the mechanical resistance to compression.



**Figure 2.** Flexural (a) and compressive strength tests (b).

Leaching experiments are used to assess the potential risk for a mortar to release toxic species into the environment during use, depending on climatic conditions [21]. They were carried out by the batch leaching using the Netherlands tank-leaching test according to NEN 7345 standard [22]: A monolithic block of 4 cm edge obtained by dry cutting  $4 \times 4 \times 16 \text{ cm}^3$  tubes is brought in contact with a fixed volume of distilled water in closed enclosures. The water/surface ratio of  $\ell/s = 5 \text{ mL/cm}^2$  is kept constant, and the sample is immersed in the water at ambient temperature ( $23 \pm 1^\circ \text{C}$ ) in the dark without stirring. The leachates are renewed for selected sample water contact times and the solution is changed every 0.25, 1.0, 2.25, 4.0, 9.0, 16.0, 36.0, and 64.0 days. A blank test was also carried out (i.e., without a solid sample). At the end, the leachates are filtered using a  $0.45 \mu\text{m}$  filter and then analyzed by ICP-AES. The leach ability  $E_i$  ( $\text{mg/m}^2$ ) of each metallic pollutant at the  $i^{\text{th}}$  extraction is given by Equation (1) [23]:

$$E_i = (C_i - C_0) \cdot V \cdot 10^{-3} / S \quad (1)$$

where  $C_i$  is the concentration of the metal at the  $i^{\text{th}}$  extraction ( $\text{mg/L}$ );  $C_0$  is the metal concentration in the blank ( $\text{mg/L}$ );  $V$  = volume of extractant (L); and  $S$  is the surface area of the sample ( $\text{m}^2$ ). After 8 extractions, the total leaching is the sum of all processed tests ( $E = \sum E_{1 \rightarrow 8}$ ) [24], and the cumulative values are compared to the leaching limits according to NEN7345 [22]. If the cumulative leaching values do not exceed the U1 values of NEN7345, the embedded sludge mortars can be used as building material without any restrictions.

For more information on the potential radiological hazard of the mortars, the average activity of Rn-222 in sewage sludge, and in mortar samples, an experimental set up was designed. An amount of 50 g of crushed sample sieved at  $100 \mu\text{m}$  were placed in cylindrical

“cans”. The solid-state nuclear track detectors (SSNTD) LR115 type II non strippable ( $2 \times 2 \text{ cm}^2$ ) was fixed on the top inside of the “can” based on alpha dosimetry [25]. The badge films are type LR115 nuclear track badges produced by KODAK and consist of a  $100 \mu\text{m}$  thick polyester substrate coated with a  $12 \mu\text{m}$  thick red colored cellulose nitrate ( $\text{C}_6\text{H}_8\text{N}_2\text{O}_9$ ) layer. It can record energy particles between 1.4 and 4.7 MeV with an incidence angle up to  $50^\circ$ . The Alpha particles traverse the detector leaving various holes with diameters depending on the incident energy. After two months of irradiation, the LR115 was chemically treated in 2.5 M NaOH at  $60^\circ\text{C}$  for time periods of 100 min. An optical microscope was used to read the developed films. After four weeks of irradiation, the LR115 was treated by 2.5 N NaOH at  $60^\circ\text{C}$  for 100 min. The radon activity  $A_V^{RN}$  ( $\text{Bq m}^{-3}$ ) was estimated according to Equation (2):

$$A_V^{RN} = \frac{D_{LR}}{\varepsilon_{LR}(\theta_C, E_\alpha)} \quad (2)$$

where  $D_{LR}$  is track density, and  $\varepsilon_{LR}(\theta_C, E_\alpha)$  is the detection efficiency. The surface radon exhalation rate  $E_S$  (in  $\text{Bq m}^{-2} \text{ h}^{-1}$ ) is expressed by Equation (3), and the mass radon exhalation rate  $E_M$  (in  $\text{Bq kg}^{-1} \text{ h}^{-1}$ ) is expressed by Equation (4), which quantifies the diffusion of radon gas in the pores of the material [26,27]:

$$E_S = \frac{A_v \cdot V \cdot \lambda_{Rn}}{A \left( t + \frac{1}{\lambda_{Rn}} \right) (e^{-\lambda_{Rn} \cdot t} - 1)} \quad (3)$$

$$E_M = \frac{A_v \cdot V \cdot \lambda_{Rn}}{M \left[ t + \frac{1}{\lambda_{Rn}} \right] (e^{-\lambda_{Rn} \cdot t} - 1)} \quad (4)$$

where  $\lambda_{Rn}$  is the radon disintegration constant ( $\text{h}^{-1}$ );  $A$  the surface area of the can ( $\text{m}^2$ );  $M$  the mass of the sample ( $\text{kg}$ );  $V$  the effective volume ( $\text{m}^3$ ); and  $t$  is the exposure time ( $\text{h}$ ).

The annual effective dose rate  $E$  ( $\text{mSv y}^{-1}$ ) is estimated by converting the absorbed dose in air  $D$  ( $\text{mGy h}^{-1}$ ) to an effective dose of  $0.7 \text{ nSv h}^{-1} \text{ Bq}^{-1} \text{ m}^{-3}$  with an occupancy factor (0.8 inside, and 0.2 outside) taking into account the equilibrium factor between Rn-222 and its descendants (0.4 inside and 0.6 outside). The total annual effective doses  $E$  ( $E_{in} + E_{out}$ ) are determined according to Equation (5) as described in several works [28,29]:

$$E = E_{inside} + E_{outside} \quad (5)$$

where:  $E_{in} = D (\text{mGy h}^{-1}) \times 8760 (\text{h y}^{-1}) \times 0.8 \times 0.4 \times 9.0 (\text{Sv Gy}^{-1}) \times 10^{-6}$  and  $E_{out} = D (\text{mGy h}^{-1}) \times 8760 (\text{h y}^{-1}) \times 0.2 \times 0.6 \times 9.0 (\text{Sv Gy}^{-1}) \times 10^{-6}$ .

## 4. Results and Discussion

### 4.1. Characteristics of Sewage Sludge

The inorganic crystalline composition of the sewage sludge was established by X-ray diffraction (Figure 3a). The main minerals obtained are calcite, quartz, mullite, dolomite and montmorillonite. The different oxides content in the sludge can vary with the quality of the raw wastewater, the nature of the coagulants, the treatment technology, and the final quality of the recycled water. Note that the calcite, mullite, and dolomite peaks constitute the major portion of the sludge, followed by  $\text{SiO}_2$ -quartz and montmorillonite and other minerals in smaller percentages. Some of them are also related to the treatment applied and the concentration of metals in the wastewater. The particle size distribution of grains is also an important physical parameter, which strongly influences their reactivity. The particle size distribution of the sludge is given in Figure 3b, measured by a laser particle size analyzer leading a particle diameter  $D_{50}$  of  $55 \mu\text{m}$  and  $D_{90}$  lower than  $239 \mu\text{m}$ . The difference between both distribution sizes was probably due to the presence of agglomerates in the initial sludge powder.



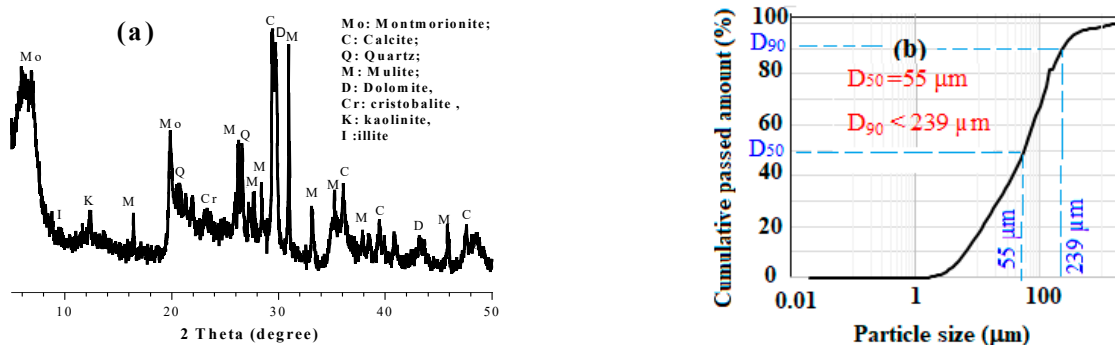


Figure 3. (a) X-ray diffraction and (b) particle size distribution of recycled sewage sludge.

Chemical composition of the sewage sludge was determined as CaO (22.47%), SiO<sub>2</sub> (47.9%), Al<sub>2</sub>O<sub>3</sub> (18.2%), Fe<sub>2</sub>O<sub>3</sub> (5.66%), Na<sub>2</sub>O (0.87%), MgO (1.65%), K<sub>2</sub>O (0.16%), and SO<sub>3</sub> (0.50%). The sludge consists mainly of CaO, SiO<sub>2</sub>, and Al<sub>2</sub>O<sub>3</sub> components, which is far from that of Portland cement due to the high SiO<sub>2</sub> content, while the CaO content is greater than 10% which can play the role of activator or binder of the pozzolanic reaction [30]. Therefore, these sludges can partially substitute Portland cement by contributing to the pozzolanic activity. In addition to the organic matter (LOI = 9.3%), the metal content analyzed is: Cr (35.95 ppm), Zn (31.34 ppm), Cu (10.27 ppm), Ni (9.28 ppm), As (8.21 ppm), and Co (5.54 ppm). In consequence, management of this sewage sludge is required to reduce the environmental risk.

#### 4.2. Mechanical Properties

Compressive strength is the most important factor in evaluating the quality of building materials [31]. The effect of percentage replacement of cement with sewage sludge on the compressive strength of mortar mixes at 7 and 28 days is shown in Figure 4a. Despite a non-significant decrease in the compressive strength of the mortars containing 5%, 10%, and 15% of sewage sludge, they were comparable to the reference mortar at all hardened ages. Beyond 20% sludge substitution, a decrease in compressive strength is recorded, and it is more marked for a higher substitution rate. The addition of 25% sludge reduces the compressive strength by 50% for 28 days curing and by 68% for 7 days, compared to the blank specimen. At 28 days the compressive strength decreases less rapidly than at 7 days, due to the aging. So, the difference between the compressive strength of the sludge mortars and the reference has reduced over time, owing to the relatively slow pozzolanic reactivity of the sludge [32].

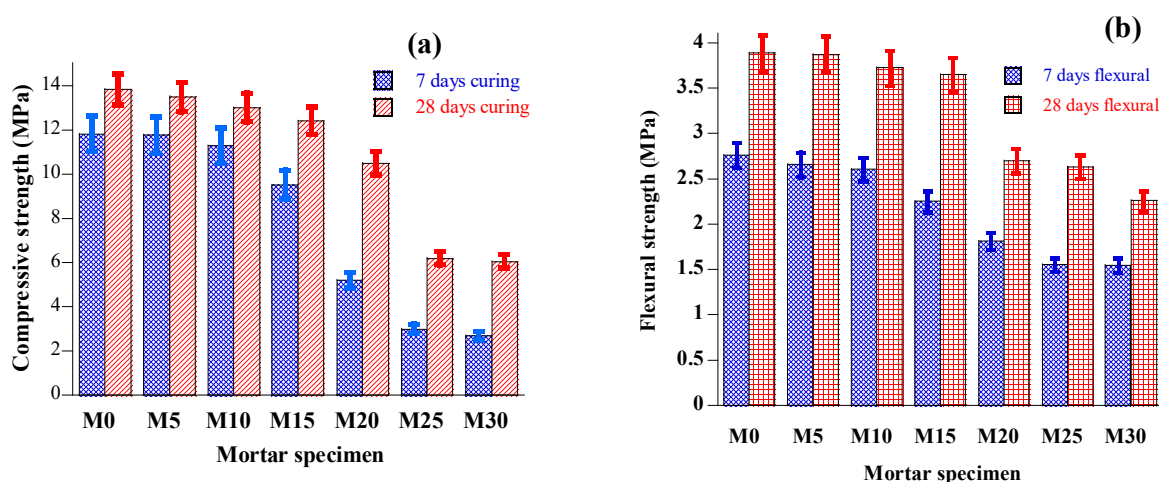


Figure 4. (a) Compressive and (b) flexural strengths of cement–sludge mortars at 7 days and 28 days aging.

The decrease in strength can be explained by excess sludge in the material leading to slow hydration which prevents the formation of portlandite and hydrated calcium silicate C-S-H [14]. This is related to the low amount of CaO in the mud compared to cement. Given the compression behavior of the mortars tested, the maximum amount of sewage sludge that can be applied to a reinforced mortar is 15%.

The flexural strength of the prepared materials is also an essential parameter to warrant the mechanical performance of construction materials (Figure 4b). After a 7- and 28- day curing period, the highest flexural strength is obtained for the 5% mortar while it is close to that of the control mortar for the 10% and 15% samples. Beyond 20%, the flexural strength drops. This can be explained by the microscopic increase in porosity and heterogeneity of the mortar, which can be confirmed by SEM analysis.

#### 4.3. Microstructure Analysis of the Mortars

Figure 5 shows the X-ray diffraction (XRD) patterns of mortar-derived powders after 28 days of curing. In addition to the major phases present in the reference mortar (calcite, quartz and ettringite), portlandite, gaylussite, albite and aragonite are detected in the mortars modified by sewage sludge. Note that the formation of portlandite is the result of the reaction between siliceous and calcic minerals, as confirmed by the decrease in the intensity of the quartz peak with addition of the sludge. Other new phases may originate from anhydrous cement grains. The drop in strength for the 30% sludge mortar can be linked to the appearance of the new microcline intermediate  $\text{KAlSi}_3\text{O}_8$  phase. X-ray diffraction shows that the intensity of the main lines for each phase varies considerably with the amount of sewage sludge incorporated, this relates to the hydration of the mortar since the sewage sludge cannot form significant amounts of Ettringite. Thus, the substitution of cement by large quantities of sludge slows down the hydration reaction and reduces pozzolanic action, justifying the loss in performance of the mortars in the strength tests.

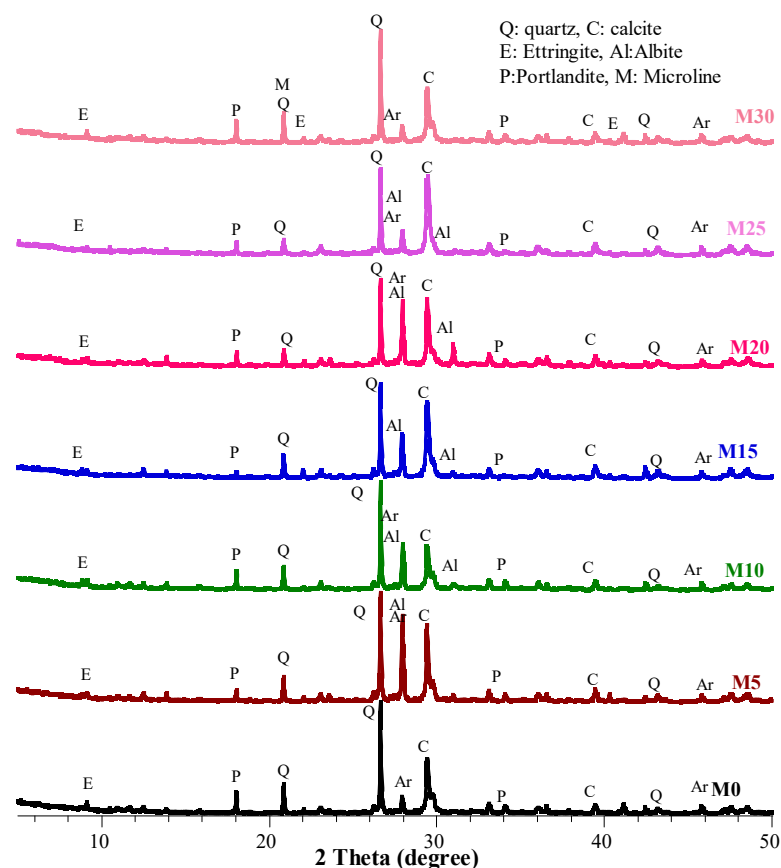
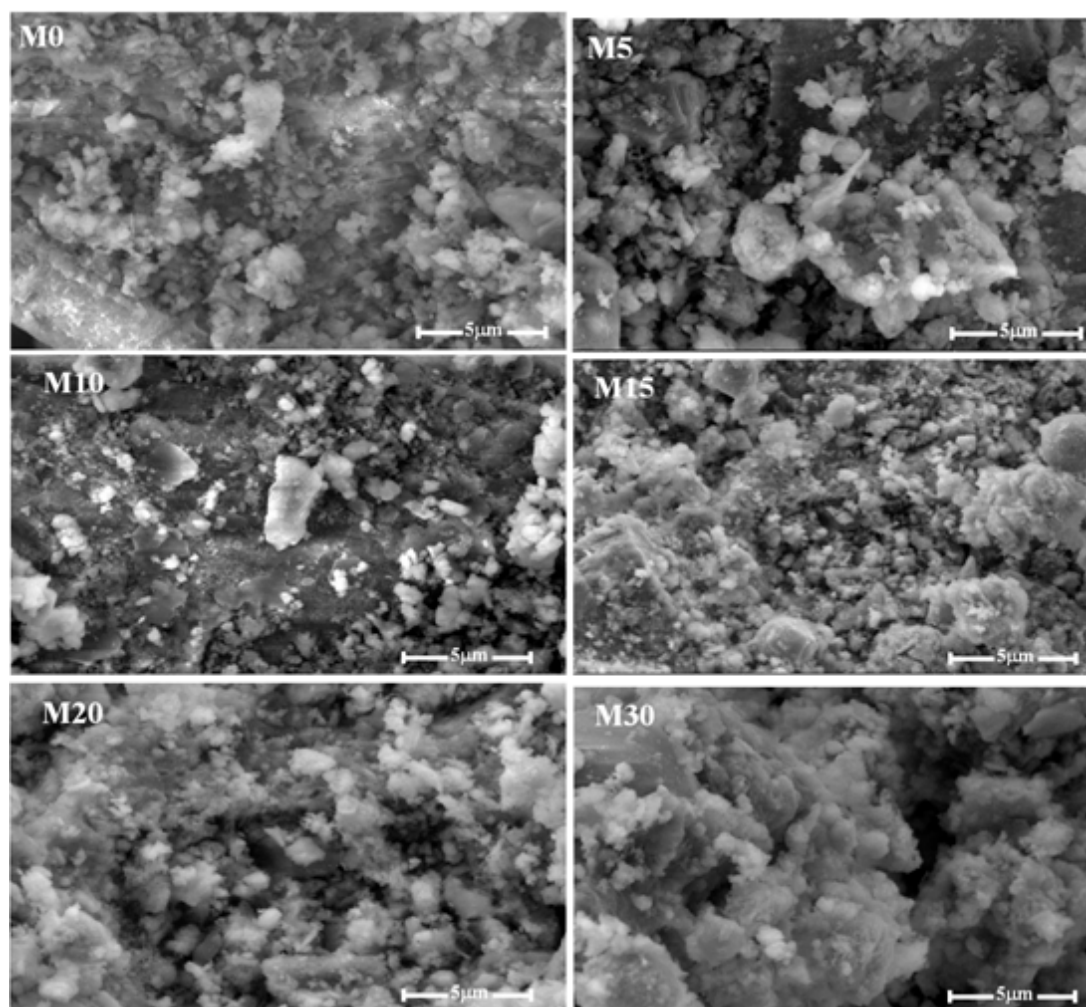


Figure 5. XRD diagrams of powders derived from sludge mortars.

The use of sludge as an alternative additive to cement is mainly attributed to its hydraulic and partially pozzolanic nature. The presence of hydraulic phases such as free lime, anhydrite, aluminates and calcium silicates in the sludge, coupled with the great fineness of their particles, leads to the development of hydration in the cementitious system. Replacing part of the cement with sludge leads to an acceleration of hydration and contributes to the formation of ettringite, portlandite, and hydrated calcium carboaluminate, the formation of this last phase of which is favored by the addition of sludge, which also stabilizes ettringite and prevents its transformation into monosulfoaluminate. Replacing 20% of the cement with sludge reduces the strength of the mortar, even if the minimum pozzolanic activity index required by the standard is reached. However, when the sludge has a high content of hydraulic phases and, particularly, of free lime, combined with a high fineness, the effect of dilution of the cement by the sludge can be compensated for at a higher age.

Scanning electron microscopy (SEM) of mortar with different amounts of sludge is presented in Figure 6. It reveals that the control mortar has the most homogeneous and compact structure. The microstructure of specimens containing less than 15% mud shows a similar dense topography. Beyond 20% sludge substitution, there are more pores larger in size. Therefore, the microstructure becomes more porous with increasing the percentage of sewage sludge, but it can also increase the number of adsorption sites for most toxic chemical species [33]. However, this exaggerated porosity results in a loss of compressive strength of the specimens as observed above.



**Figure 6.** SEM images of prepared mortars compared to control mortar, at the same scale.



#### 4.4. Environmental Assessment of Sludge-Amended Mortar

Based on the mineralogy of the sewage sludge, the environmental impact of incorporating sewage sludge in construction materials is of great concern. It must meet environmental requirements related to the presence of heavy metals in the raw sludge. Heavy metals are of particular concern in sludge that could penetrate the bricks and cause secondary pollution into the environment; they can also leach from the cementitious materials and affect the surrounding environment [34]. Leaching from the mortar should not exceed the maximum authorized limit, even under extreme conditions [35]. For this reason, leaching tests on sewage sludge and sludge-amended products are required.

Cumulative releases of heavy metals from the prepared sludge mortar specimens and the limits set in the NEN 7345 standard are shown in Table 4. Note that the leaching values for all the heavy metals are significantly below the regulatory U1 limits. It shows that these metals have a good chemical affinity with the silica contained in the cement. Note that the solidification of cementitious materials by pressing and subsequent drying or heat treatment are the most effective methods for the immobilization of heavy metals [36]. Despite their low heavy metal content, they could also be integrated into the structure and could react with the components of the hydrated products to form precipitates during the hydration of the cement [37]. In addition, the dense and hardened structure of the sludge-amended mortars offers limited permeability that prevents heavy metal leaching. Hence, the use of recycled sludge from the treatment of wastewater from tanneries is possible without environmental constraints in the development of mortars. To this end, the storage of heavy metals from wastewater treatment sludges in construction materials is an advantageous approach, which considerably reduces the environmental pollution by heavy metals.

**Table 4.** Cumulative release of heavy metals from mortar specimens into deionized water (in mg/m<sup>2</sup>) after leaching tests of 64 days.

Mortars	M0	M5	M10	M15	M20	M25	M30	Leaching Limits NEN 7345 U1	U2
As	<LD *	<LD	<LD	<LD	<LD	<LD	<LD	40	300
Cr	<LD	0.012	0.198	0.24	0.38	0.46	0.48	150	950
Cu	0.002	0.0038	0.0034	0.004	0.0046	0.0054	0.0054	50	350
Ni	0.00562	0.02	0.034	0.032	0.0134	0.014	0.0138	50	350
Zn	0.0078	0.058	0.054	0.78	0.64	0.56	0.5	200	1500

\* Limit of detection.

#### 4.5. Radiological Assessment of Sludge-Amended Mortar

Evaluation of radiological activity in sludge and mortars is recommended to ensure that they do not contain radiation-emitting substances. The radiological characteristics measured in all the samples are given in Table 5. For sewage sludge, its radon activity  $A_r$  is 624 mBq m<sup>-3</sup> h, higher than in amended sludge mortars, ranging from 168 to 418 mBq m<sup>-3</sup>, and below the recommended action level [38]. According to the safety recommendations of UNSCEAR 2000, none of the sludge-containing mortars presents a risk for health. In all mortars, the radiological results also indicate that the mortars exhibit low surface and mass exhalation compared to those of sewage sludge. In mortars specimens, the  $E_s$  values varied between 132 and 341 mBq m<sup>-2</sup> h<sup>-1</sup> and  $E_M$  from 7 to 19 mBq kg<sup>-1</sup> h<sup>-1</sup>, much lower than those of the sludge and the world average (57.6 Bq m<sup>-2</sup> h<sup>-1</sup>) [38]. Obviously, these activities increase with the sludge content in the mortar. This is due to the porosity of the materials because the radon diffusion is faster in porous materials [39], and therefore the rate of radiation emanation increases [40]. This correlates well with the powdery morphology in the SEM images.

**Table 5.** Radon activity, radon exhalation, and annual effective dose of sludge and mixtures.

Samples	$A_V$ (mBq m <sup>-3</sup> )	$E_S$ (mBq m <sup>-1</sup> h <sup>-1</sup> )	$E_M$ (mBq kg <sup>-1</sup> h <sup>-1</sup> )	Annual Dose $E$ (mSv/Year)
sludge	624	489	28	16
M0	168	132	7	4
M5	182	143	8	5
M10	250	196	11	6
M15	312	244	14	8
M20	354	301	17	9
M25	418	328	19	10
M30	418	328	19	11

The effective annual dose of radon gas varies from 4–11 mSv/year in mortars, lower than in the sludge (16 mSv/year). The authorized limit for humans is set at 3–10 mSv/year [40], so only mortars with a low concentration of mud (<20%) are acceptable in this context (Table 5). Therefore, the studied sludge-amended mortars do not have an excessive contribution to the radioactivity in buildings and can be used in construction materials without additional risk for public health.

## 5. Conclusions

Sludge from the wastewater infiltration process of tanneries can be used as a fine aggregate substitute to cement in mortars. Different mortar samples containing up to 30% sludge were prepared. Mechanical, mineralogical, microstructural, environmental, and radiological characterization of the samples reveals their suitability as building materials. The compressive and flexural strengths of mortars mixed with 5%, 10%, and 15% sewage sludge are comparable at all cured ages to those of the reference mortar. Beyond 20% sludge substitution, a considerable decrease in resistance is recorded, but it remains conformed to the resistance requirements of construction materials. The maximum amount of sludge that can be added to the mortars is 20%. After 64 curing days, the concentration of Cr, As, Cu, Ni, and Zn in the leachates was much lower than the required limit values by NEN 7345 Dutch standard. This demonstrates the effectiveness of the process for the management of heavy metals contained in the sludges from tannery wastewaters. The exhalation of radon on the surface and in the mass of the mortars is below the annual effective dose of <sup>222</sup>Rn as per the UNSCEAR requirement. In consequence, sludge from tannery wastewater treatment can effectively be used as a sustainable building material, with acceptable environmental and radiological risks.

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