Chemical-Physical Characterization of Stava Tailings Subjected to an Innovative Aging Technique

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Chemical-Physical Characterization of Stava Tailings Subjected to an Innovative Aging Technique

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Abstract **-** Tailing dams are realized to store the waste products resulting from the mining extraction processes. These complex geotechnical structures should be designed taking into account long-term stability and long-term properties of the deposited materials. Depending on the interactions between source mineralogy and local conditions, tailing wastes can undergo aging processes with chemical and physical modifications. Recently, in many countries tailing wastes are re-used as feedstock for cement and concrete, backfill or landscaping material, so if any, the long-term chemical and physical modifications could affect the hydro-mechanic response of tailings, resulting in relevant environmental and economic consequences. An increased interlocking of particles and oxidation, sometimes making previously safely held contaminants available and mobile, are recognized as common aging processes. Among the long-term aging processes, the natural ionizing radiation due to ultraviolet rays or cosmic rays can be considered. Moving from these reasons, this paper presents an innovative accelerated aging technique to simulate the natural ionizing radiation from the sun. Tailing fluorite ore samples collected form the collapsed Stava dams (Italy) were characterized in dry and wet conditions, before and after the gamma rays treatment. Stava silty tailings showed some physical modifications in terms of specific surface, size particle distribution and inner porosity of the particles, while they revealed a certain chemical stability.

Keywords**:** tailing dams; aging; gamma ray irradiation; Stava dam; tailings.

1. Introduction

Chemical and physical modification of long-term tailings properties could affect their mechanical behaviour, with consequences for engineering applications, like their use as aggregate in road construction, backfill, feedstock for cement and concrete paving, or landscaping material. While some previous experimental results obtained on standard soils showed improvements with time, i.e. in terms of shear strength due to the cementation of particles ([6]), or mineralogical variations ([9]), other investigations showed no significant aging effects ([2]). The current study is aimed to investigate the effects of ionizing radiation of the sun on the Stava silty tailings and its influence on their physical and chemical properties. An innovative accelerated aging procedure has been used on silty tailings by means of exposure of tailing samples to gamma rays ^{60}Co source to simulate the aging process in wet conditions for tailings deposited within the storage facility, or in dry conditions if tailings are used for bricks. Chemical and physical characterization of the samples was performed before and after the induced aging treatment.

2. Testing material

Tailings studied in the current research were collected from the Stava dams collapsed in 1985. The chemical-physical characterization, as well as the induced aging procedure, was performed on the silt fraction passing through a sieve n°200. The liquid limit, plastic limit, specific gravity and permeability were respectively 27.4%, 18.0%, 2.82 and 10^{-7}m/s ([1]). X-ray diffraction analysis showed that the silt fraction was basically constituted of quartz, with a significant amount of calcite and fluorite and kaolinite ([3]; [5]). A sample 1000g weight (STAVA_1), was oven dried at 120°C for 24 hours and then its physical and chemical properties were characterized (Tab. 1). A second sample (STAVA_2), 500g weight was obtained from S_1 and then treated by means of gamma irradiation in the form of loose, dry material and then physical-chemical characterized. Finally, a third sample (STAVA_3), 500g weight, was obtained from sample STAVA_1, irradiated in wet conditions by adding a deionized water to get a slurry, and then its physical and chemical properties were characterized.

3. Gamma-ray treatment

The gamma ray irradiation was performed at the laboratory of Centro de Desenvolvimento de Tecnologia Nuclear (Belo Horizonte, Brazil), in a panoramic multipurpose gamma irradiator, category II, from MDS Nordion, model IR-214 BG-127, equipped with a ^{60}Co gamma source, dry storage (maximum activity of 2200 TBq or 60000 Ci), as shown in Fig. 1.

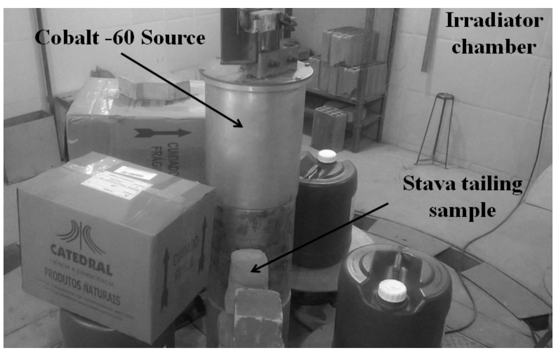


Fig. 1: Irradiator chamber and equipment.

Table 1: Gamma irradiation conditions.

Irradiation time, t_γ	7 days
Exposure dose	1000 kGy
Distance of the samples from the gamma source, d	0 cm
Gamma source (diameter d=20 cm, height h=100 cm)	^{60}Co
Photon energy, E_γ	1.17 ÷ 1.33 MeV
Frequency of gamma photon, f_γ	$2.83\times10^{20} \div 3.22\times10^{20}$ Hz
Temperature, T	Irradiation chamber temp. (60°C)

The imposed photonic energy (E_γ) and the source activity ($A_\gamma = 30000 \cdot 3.7\cdot10^{10}\ s^{-1}$) give the power (P_γ) emitted by gamma ray source. Knowledge of the lateral surface of the cylindrical ^{60}Co source ($S_{S.R.}$), allowed evaluating the gamma ray intensity (I_γ) that results 136 times higher than ultraviolet intensity ([8]):

$$I_\gamma = \frac{P_\gamma}{S_{S.R.}} = \frac{A_\gamma\ E_\gamma}{\pi\ d\ h} = \frac{30000\ \cdot\ 3.7\ 10^{10}\ 1.2\ \cdot\ 10^6}{\pi\ \cdot 20\ \cdot\ 100} = 3.39\ 10^{-2}\frac{W}{cm^2} \quad (1)$$

The imposed irradiation time (t_γ = 7 days) and the estimated gamma ray intensity allowed obtaining the energy density (ρ_γ):

$$\rho_\gamma = I_\gamma \cdot t_\gamma = 3{,}39 \cdot 10^{-2} \cdot 7 \cdot 24 = 5{,}7\frac{Wh}{cm^2} \quad (2)$$

An equivalent energy density $\rho_{\text{sun}}^{\text{UV}} = \rho_\gamma$ needs UV-sun exposition time ($t_{\text{sun}}^{\text{UV}}$) equal to:

$$t_{sun}^{UV} = \rho_{sun}^{UV}/I_{sun}^{UV} = 5.7/250\ \ 10^{-6} = 22400\ h \cong 2.6\ years \quad (3)$$

where $I_{\text{sun}}^{\text{UV}} = 250 \cdot 10^{-6}\ W/cm^2$ is the UV rays intensity (sun radiation on the Earth surface). Ionizing gamma ray activity into the matter is 50÷500 ions/cm and on the safe side 50 ions/cm are assumed. Ionizing UV rays activity is 1÷2 ions/cm so, the UV-sun exposition time should be multiplied by a factor no less than 25, so that the high frequency gamma radiation used in this research simulates at least seventy years of continuous exposure of tailing samples to ultraviolet rays.

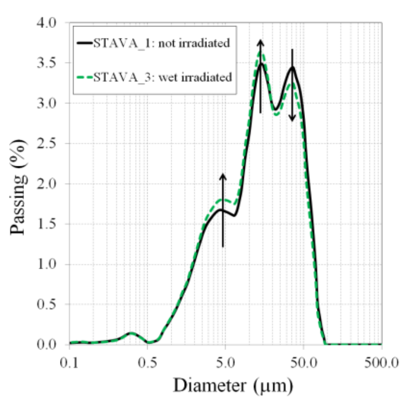
4. Experimental results

Physical and chemical tests carried out on Stava tailings and equipment details are summarized in Table 2. From the physical point of view, modifications of the grain size distribution were observed after irradiation both in dry and wet conditions, with an increase of the silty fraction (about 3-4% in volume), as shown in Fig. 2a. It seems that larger particles are broken by the irradiation, increasing the frequency of finer particles. This outcome was confirmed by a reduction of the particle area and average diameter of grains as observed by processing the digital SEM pictures, or by carrying out adsorption tests that allowed observing an increase of the specific surface area and particle porosity of grain after the gamma ray exposure (Table 3). Details are given in [4].

Table 2: Chemical-physical characterizations of Stava silty tailings and equipment details.

Property	Technique	Equipment
Grain size distribution	Laser diffraction	Cilas, model 1190
Electronic microscopy	SEM	Carl Zeiss, model VP
Energy dispersive X-ray spectroscopy	EDS	Bruker, model XFlash 4.0
Particle morphology	Image analysis	Software Quantikov*
Particle density	Helium pycnometry	Quantachrome, model Ultrapycnometer
Specific surface and pore diameter	N_2 adsorption	Quantachrome, model NOVA 2000
Chemical analysis	Infrared spectroscopy	ABB Bomem, model MB102
	X-rays fluorescence	-
Mineral composition	X-rays diffractometry	Rigaku, model D\MAX\ULIMA**

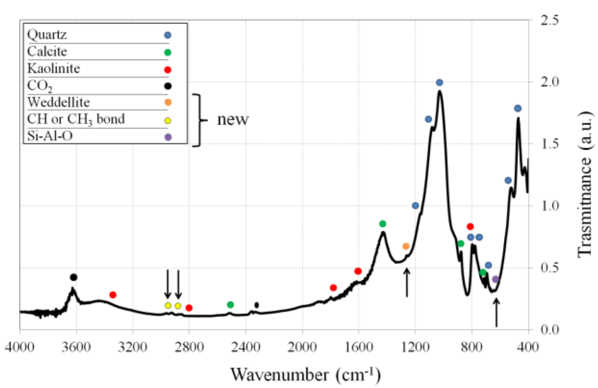
* [7]. ** XRD spectra collected with Cu-Kα radiation and data collected within the range of 20°-80°.



a)

Table 3: Results of image analysis of SEM images, N_2 adsorption and helium pycnometry of Stava silty tailings before and after the gamma-ray treatment.

	Technique	STAVA_1	STAVA _2	STAVA _3
Area (μm^2)	SEM	245.77	198.74	199.42
Diameter (µm)	SEM	15.64	14.86	14.35
Roundness	SEM	0.72	0.71	0.68
Specific surface (m^2/g)	N_2 adsorption	5.35	5.65	5.72
Particle porosity (%)	N_2 adsorption	3.83	4.32	4.43
True density (g/cm^3)	Helium pycnometry	2.897	2.906	2.943
Average pore diam. (Å)	N_2 adsorption	102.741	109.872	106.362



b)


c)

Fig. 2: a) Comparison between grain size distribution of not irradiated sample (solid line, STAVA_1) and wet irradiated sample (dotted line, STAVA_3).

Fig. 2: b) Fourier transform infrared spectroscopy spectra for dry irradiated sample (STAVA_2) and evidence of the new peaks.

Fig. 2: c). Quantachrome Apparatus for specific surface and pore diameter determination.

Chemical tests showed that the Stava tailing samples exhibit a certain stability. Fluorescence, X-ray diffraction and EDS analyses showed little changes of concentration of the constituent elements before and after the treatments. FTIR spectra, both for dry irradiated sample STAVA_2 (Fig. 2b) and wet irradiated sample STAVA_3 showed new peaks if compared to spectra obtained before the gamma ray treatment on sample STAVA_1. The new peaks were due to the presence of a new element (weddellite) and chemical bonds, respectively CH or CH_3 and Si-Al-O ([4]). In this context, the formation of Weddellite could be due to the reaction of calcite with CO_2 and H_2O catalysed by the gamma irradiation, while CH and CH_3 to the reaction of the products of the radiolysis of water molecules with CO_2. Finally, the formation of new Si-Al-O bonds could be attributed to a weak loss of "crystallinity" of the kaolinite mineral of the Stava tailings during the irradiation process.

5. Conclusion

A new accelerated aging procedure was proposed and tested to simulate in the short time, the long-term UV ray exposure of tailings. This technique was applied to the silty fraction of Stava tailings, which have been characterized before and after the induced aging. The variations in their chemical and physical properties observed in these tests were not found to be significantly affected the hydro-mechanical behaviour. In order to improve the correspondence to the deposited tailings, further irradiations tests could be carried out by using processing water, as well as mixtures made of silty and sandy tailings in different percentages to better simulate the heterogeneity of the deposited material inside the basin. Finally, it seems that the tailing behaviour is directly related to its chemical composition and the processes it undergoes during ore beneficiation. Since in this research the accelerated aging procedure was applied on fluorite ore tailings, it should be further validated by testing other tailings, such as iron ore tailings.

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