Abstract

Tailings storage facilities are disposal systems for storing the waste products of the mining industry consisting of a slurry mixture made of soil, rock and water that remain after the mineral values have been removed from the patent ore. Tailings dams are supposed to last forever, so after their deposition, tailings can experience aging processes with physical and chemical changes depending on the interactions between local conditions and source mineralogy. The consequences of these aging processes are increased interlocking of particles and oxidation processes, sometimes making previously safely held contaminants available and mobile. Among the long-term aging processes, the natural ionizing radiation (from radioactive isotopes of the soils, cosmic rays, and also ultraviolet rays from the sun) can be considered, as proposed in the current research. Furthermore, in many countries, tailings are beginning to be re-used as backfill, landscaping material or feedstock for cement and concrete. So if any, the long-term physical and chemical modifications could affect the hydraulic and mechanical behaviour of tailings with relevant economic consequences. For these reasons, wet and dry silty samples of tailings spilled out after the failure of the Stava tailings dam (Trentino Alto Adige, Italy) were exposed to gamma rays, as an accelerated aging technique to simulate the natural ionizing radiation, and then characterized. The modifications on physical and chemical properties were observed and, despite certain chemical stability, some physical changes were observed, particularly in terms of size particle distribution, inner porosity of the particles and specific surface.

Keywords: aging, tailings, irradiation, gamma rays.

1. Introduction

Tailing dams are complex geotechnical structures that should be designed taking into account long-term stability and long-term properties of materials (DME, 1999; Szymanski, 1999; ONTARIO, 2000; Silva et al., 2003; Xendis et al., 2004; Bjelkevik, 2005; Jantzer and Knutsson, 2010; ANCOLD, 2011; ICOLD 2013). Long-term physical, chemical and geotechnical properties of tailings need to be deeply understood in order to approach engineering problems related to this complex material, i.e. stability analyses of tailing embankments, use of tailings as backfill, landscaping material, aggregate in road construction, or feedstock for cement and concrete paving, especially in Brazil, USA, Canada, China, India and Australia (Esposito et al., 2014).

Some studies concerning simulation of long-term processes on natural soils and tailings wastes can be found in literature, sometimes with contrasting results. Troncoso et al. (1988) studied the cyclic resistance on aged copper tailing samples by performing cyclic triaxial tests. They estimated that cementation between particles can increase the cyclic liquefaction resistance of the impoundment more than 250% over 30 years. The results of cyclic triaxial tests on the 30,5 and 1-year old quartz tailings samples are shown in Fig. 1a in terms of cyclic stress ratio (defined as the ratio of amplitude of cyclic axial stress to twice the initial confining stress) required to produce 5% double amplitude axial strain.
Larrahondo and Burns (2014) performed some triaxial tests aimed to simulate the increased interlocking of particles (cementation), in order to evaluate the shear strength of sandy tailing wastes, showing an increasing of the friction angle at the critical state.

![Graph showing the effect of aging on tailings properties](image)

Craw et al. (1999) and Devasahayam (2007) performed chemical tests on tailings samples after drying, wetting and oxidation for 4 years, obtaining some chemical and mineralogical variations. Others laboratory studies on sandy soils show improvements with time due to the aging process (Mitchell and Solymar, 1984; Schmertmann, 1987; Mesri et al., 1990; Baxter and Mitchell, 2004) by showing increase of shear modulus $G$ (Daramola, 1980, Schmertmann, 1991, Pender et al., 1992), and also at small strains, $G_0$ (Afifi and Woods, 1971; Anderson and Stokoe, 1978; Baxter and Mitchell, 2004 as shown in Fig. 1b), liquefaction resistance (Seed, 1979; Ishihara, 1985) and penetration resistance, as reported by Joshi et al. (1995, Fig. 1c). Despite an increasing number of examples of property improvements, there are field studies (Baxter, 1999; Jefferies et al., 1988; Charlie et al., 1992; Jefferies and Rogers, 1993) and laboratory tests (Human, 1992; Miller, 1994; Baxter; Mitchell, 2004) on sandy samples in which no significant aging effects, such as an increase of penetration resistance, were shown.

In the deposit, tailing materials are subjected to physical and chemical alterations due to weathering and other factors. Several minerals contain potassium in their composition, which is composed of 0.01% of $^{40}$K, a natural occurring isotope that emits beta rays and gamma rays. On the surface of tailings deposits or in the tailings bricks faces used for constructions, the minerals are exposed to cosmic rays and ultraviolet rays. The energy of these radiations is absorbed by the minerals by displacing electrons from their original positions in the crystal structure of minerals. In the long-term, these radiations may trigger alterations of the materials due to their ionizing character. Aimed to study the effects of ionizing radiation on tailings and its influence on physical and chemical properties, induced aging tests were performed by means of exposure to gamma rays, and are reported in this paper. Silty tailings samples from Stava (Italy) fluorite extraction were chosen. Dry and wet samples were exposed to gamma rays from a $^{60}$Co source up to a dose of 1000kGy in order to simulate the aging process in two extreme cases: dry conditions if tailings are used for bricks, wet conditions for tailings deposited within the storage facility.

2. Material properties and testing techniques

Tailings used in this research were collected in 2005 from the lower portion of the Stava upper dam, after its failure in 1985. They were deposited within two tailings storage facilities raised with the upstream method, built one above the other on a natural slope near village of Stava (Italy) and aimed to store the slurry tailings coming from the separation flotation process of the Prestavel fluorite mine (Sarsby, 2013). The lower embankment failed due to overtopping caused by the failure of the upper one, triggering a flowslide consisting of more than 20000m$^3$. The mixture of water debris and sediments travelled down the Stava valley to speeds of 90km/h obliterating everything in its path for a stretch of 4.2km, until it reached the municipality of Tesero and then it flowed into the Avisio river, causing 269 deaths and damages valued at 133 million euros (Lucchi, 2011). Factors such as high phreatic level, poor drainage, and the unconsolidated state of the tailings have caused the static liquefaction that led to the collapse of the Stava tailings dam.
2.1 Material characterization

After collection in 2005 and until running the tests, Stava tailing were kept air dried at a constant temperature equal to 22°C. Only the silty fraction of the Stava tailings passing through sieve ASTM n°200 was tested. This choice was done in order to simplify the experimental program even if it is well recognized that, because of the adopted deposition technique, tailing basins are characterized by a great heterogeneity of materials in terms of grain size distribution. Geotechnical characteristics of the Stava tailings are summarized in Tab. 1. A sample of 1kg of silt, labelled Sample 1, was oven dried at 120°C for 24 hours and then characterized as shown in Tab. 2.

| Property          | Specific weight, $G_s$ (g/cm$^3$) | Permeability, $k$ (m/s) | Liquid limit, $w_L$ (%) | Plastic limit, $w_P$ (%) | Plasticity index, PI |
|-------------------|-----------------------------------|-------------------------|-------------------------|-------------------------|----------------------|
| Finer fraction    | 2.828                             | $2.10^{-7} + 4.10^{-9}$ | 27.4                    | 18.0                    | 9.4                  |
| Coarser fraction  | 2.721                             | $1.10^{-8} + 9.10^{-9}$ | 27.4                    | 18.0                    | 9.4                  |

Table 1 Geotechnical index properties of Stava tailings (Carrera, 2008).

From Sample 1, a second sample, 500g weight, labeled Sample 2, was obtained and then treated by means gamma irradiation in the form of loose, dry material. A third sample, 500g weight, named Sample 3, was also obtained from Sample 1 and irradiated in wet conditions by means of adding a quantity of deionized water corresponding to the liquid limit in order to obtain a slurry.

![Gamma irradiator, location of the tailing sample; (b) Dry sample before and after the treatment by gamma irradiation.](image-url)
Aging simulation of the tailings from Stava fluorite extraction by exposure to gamma rays

### Table 3

| Parameter                  | Value                        |
|----------------------------|------------------------------|
| Irradiation time           | 7 days                       |
| Exposure dose              | 1000kGy                      |
| Distance to the gamma source | 0cm                          |
| Gamma source               | $^{60}$Co                    |
| Photon energy              | $1.17 \pm 1.33\text{MeV}$    |
| Frequency of gamma photon  | $2.83\times10^{20} \pm 3.22\times10^{20}\text{Hz}$ |
| Temperature                | Irradiation chamber temperature (about 60°C) |

Before the treatment, Sample 2 and Sample 3 were both sealed within plastic containers and then covered with aluminum paper and masking paper. The irradiations were 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, with maximum activity of 2200TBq or 60000Ci. The samples were attached on the wall of the cylindrical gamma source as shown in Fig. 2. Table 3 summarizes the irradiation conditions. After the irradiation, Samples 2 and 3 were characterized according to Table 2. Use of gamma rays permits acceleration of the aging process. In one week, the high frequency gamma radiation used in this research (intensity $3.39\times10^{-2}\text{W/cm}^2$) simulates about seventy years of continuous exposure of tailings samples to ultraviolet rays (intensity $250\times10^{-2}\text{W/cm}^2$).

### 3. Results

Mineral constituents were quantified carrying out X-rays diffractometry by using incident X-rays through a not irradiated sample and irradiated samples, in dry and wet conditions. Diffractograms present practically the same peaks with the same intensities, meaning the three samples have the same composition, with fluorspar (26%) and quartz (35%) as the main constituents, and calcite (9%), muscovite (3%), rutile (0.3%) and dolomite (0.5%) as the minor constituents. Little variations of mineral constituents before and after the treatment suggest no modifications occurred after irradiation.

The chemical composition was then investigated by means of X-rays fluorescence on a not irradiated sample and irradiated samples, again in dry and wet conditions. The X-ray fluorescence is a technique for quantitative identification of chemical elements by using incident X-rays through a solid sample (Beckhoff et al., 2006). The elementary composition is practically the same for all samples, giving high percentages of calcium (34%), aluminium (27%), fluorine (26%) with traces of potassium (3%), iron (1.3%), zinc (0.5), magnesium (0.4) and other elements, i.e. sodium and manganese.

The same results were observed from analyses performed by scanning electron microscope (SEM), by using the Energy Dispersive Spectroscopy technique (EDS), which showed the same spectra for all samples.

Figure 3a, 3b and 3c show the Fourier Transform Infrared spectroscopy (FTIR) spectra of the three samples. This technique is a powerful tool for qualitative and quantitative identification of either organic or inorganic chemicals, impurities and percent crystallinity by using infrared radiation through solid, liquid or gaseous samples. Before the spectra acquisition, the samples were oven dried at 120°C for 24 hours, dispersed in potassium bromide (KBr) and pressed in a form of thin disks. Potassium bromide was used in order to disperse the tailings powder to decrease the absorbance to infrared radiation. The mass of tailings and KBr, as well as the compaction pressure, was the same in all disks, so that they had the same dimensions. By comparing the spectra of Samples 2 and 3 with Sample 1, it is possible to see small differences. Sample 2 shows a small band at 1262cm$^{-1}$, which was attributed to the formation of hydrated calcium oxalate, weddellite (Frost et al., 2003), probably due to the reaction of calcite and residual water. Samples 2 and 3 also show bands at 2923cm$^{-1}$ and 2966cm$^{-1}$, related to C-H bonds and at 662cm$^{-1}$, related to Si-Al-O bonds.

Figures 3d and 3e show the particle size distributions (Stojanovic and Markovic, 2012) of Samples 2 and 3 compared with Sample 1. Considering Sample 1 (before irradiation) and Sample 2 (after dry irradiation), there is a 3.2% of volume decrease of particles with sizes between 19μm and 100μm. The same amount is related to a volume increase of particles with sizes between 0.5μm and 10μm. In the case of Sample 3 (after wet irradiation), there is a 3.0% volume decrease between 19 and 100μm and the same amount of volume increase between 0.5μm and 10μm. By comparing Samples 2 and 3, the differences in particle size distributions are very small (0.3 to 0.4% in volume).
Figure 3
Fourier transform infrared spectroscopy spectra for not irradiated sample (a), dry irradiated sample (b) and wet irradiated sample (c). Comparison between grain size distribution of not irradiated-dry irradiated sample (d), and not irradiated-wet irradiated sample (e).

Images obtained by scanning electron microscopy are shown in Fig. 4 with 450 X magnification. Particles of Sample 1 appear well defined, while, after irradiation, particles appear smaller and show a slight trend to form clusters.

Table 4 shows the results of image analysis. All samples show the same roundness (Koivuranta et al., 2013) of about 0.7.

Figure 4
(a) SEM images for not irradiated sample; (b) SEM images for dry irradiated sample; (c) SEM images for wet irradiated sample.
Irradiated samples show an average diameter (14.6μm) smaller than the not irradiated one (15.6μm). In terms of surface, tailings particles of the not irradiated sample are bigger than the irradiated ones. These results are in good agreement with laser particle analysis. By analysing area, average diameter and roundness of Sample 2 and Sample 3, it is possible to observe there are no meaningful differences, if the ageing process is carried out on dry or wet tailings samples.

The results of specific surface, particle density and pore diameter, obtained by helium pycnometry and N₂ adsorption techniques, are shown in Table 4. It is possible to observe an increase of the specific surface, particle porosity and average pore diameter (inside of the particles) of the irradiated samples. The true density is practically the same for all samples.

4. Discussion

The tailing samples tested in this study are composed of fluorite, quartz, calcite, kaolinite and small amounts of rutile and dolomite. Small changes were observed in the FTIR spectra of irradiated samples, which can probably be attributed to reactions in consequence of the ionization character of gamma rays. A small fraction of larger particles (about 3% in volume) were disintegrated into small particles. The consequences were the decrease of the mean particle size and increase of the specific surface of the irradiated samples. The inner porosity of the irradiated particles also increased, maybe due to an increase of the inner pore sizes. Dry and wet irradiation produced practically the same results.

The obtained results allow to assert that the tailings analysed in the current research have shown a certain physical and chemical stability if exposed to solar radiation for about 70 years. Depending on their ionizing energy, cosmic rays and ultraviolet rays from the sun have a penetration depth equal to about 1cm. Based on results of the current physical and chemical characterization, the effects on mechanical and hydraulic of tailings are not relevant.

5. Conclusions

Indirect aging tests were performed on silty samples of Stava tailings dams, by means of gamma radiations bombardment carried out at Centro de Desenvolvimento de Tecnologia Nuclear in Belo Horizonte (Brazil). Tailings were treated and characterized both in dry and wet conditions in order to analyze the influence of UV radiations on the chemical and physical properties of tailings on the surface of tailings deposits or in the tailings brick faces used for buildings. Physical modifications have been studied by means of different characterization tests before and after the ionizing treatment. Changes in terms of particle size distribution were observed after irradiation in dry and wet conditions. In both cases an increase of some silty fractions was observed. Processing of the digital SEM pictures showed a reduction of area and average diameter of grains for Sample 2 and Sample 3. Adsorption tests showed an increase of the specific surface area after the treatment. Contextually, an increase in the particle porosity of grains was obtained. From the chemical point of view, the tailings samples exhibit a certain stability. X-ray diffraction, fluorescence and EDS analysis showed little changes of concentration of the constituent elements before and after the treatments. Finally, for Sample 2 and Sample 3, infrared spectroscopy showed peaks due to the presence of new chemical bonds.

It is worth noting that the wet irradiation was carried out in pure water, while the real environment is more complex. In the site, tailings are mixed with water which contains dissolved salts, heavy metals, contaminants and residual chemicals from the mineralogical processes. This choice was adopted to simplify the experimental program. Hence, in order to improve the affinity with the real environment, further wet irradiations tests can be carried out by using processing water. Additionally, other tests can also be performed by increasing the photon energy emitted by gamma rays, in order to simulate a greater aging period both on dry and wet samples, as well as the mixtures made of sandy and silty tailings in different percentages have to be tested in order to take into account the heterogeneity of in situ tailings.

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