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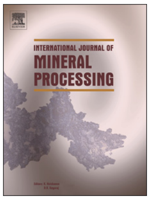

# Sulphate induced changes in the reactivity of cemented tailings backfill

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ABSTRACT

The reactivity of cemented paste tailings (CPT) that contains sulphide mineral-bearing tailings is a key parameter that influences its environmental performance and durability. This reactivity can be influenced by several factors, such as the initial sulphate content of the CPT. In this paper, the effect of the initial sulphate content of the CPT on its reactivity is experimentally investigated by conducting oxygen consumption (OC) tests on CPT specimens. Microstructural testing is also conducted on CPT specimens to better understand the mechanisms responsible for the changes in the reactivity of CPT. These specimens are prepared by mixing defined amounts of pyritic tailings (45 wt%), varying proportions of Portland cement type I or Portland cement partially replaced with different types and amounts of mineral admixtures, and mixing water with various sulphate contents (0, 5000, 15,000 and 25,000 ppm). The samples are cured for 150 days at room temperature. The results show that regardless of the type of binder, the reactivity of the CPT specimens increases with increasing contents of sulphate except for a sulphate content of 5000 ppm. Also, partial substitution of Portland cement type I with mineral admixtures, such as granulated blast furnace slag or fly ash, reduces the chemical reactivity. Regardless of the initial sulphate content, increasing the cement content and/or replacing cement with mineral admixtures leads to the reduction in the reactivity of the paste.

## 1. Introduction

Mining is an important economic activity in Canada as well as in many parts of the world. In 2015, the mining industry added a value of $67 billion to the gross domestic product (GDP) in Canada (Mining Association of Canada, 2017). On the other hand, the mining industry is considered to be the largest solid waste producer in Canada and many countries worldwide. Among the different types of mine waste, a substantial volume of a byproduct of mining activities are produced every year. This byproduct are fine grain residual materials called tailings. This type of mine waste is often disposed and stored in tailings storage facilities (TSFs) on the upper surface of mine sites. Tailings are commonly disposed and stored in the slurry form which consists of a mixture of fine ground rock, processing water and chemical agents. This way of managing a large volume of tailings has economic, social and environmental consequences in the long term (Gleisner, 2005). Furthermore, the management of mine tailings is one of the main challenges that the mining industry is facing worldwide. This is because tailings management is a more complex process if the tailings contain chemically reactive minerals, such as sulphide minerals (commonly pyrite). These sulphide minerals are chemically unstable in the presence of air (oxygen) and water because they oxidize to form a highly acidic, sulphate-rich solution, which is called acid mine drainage (AMD) (Buckby et al., 2003). The generated acid solution can increase the acidity of the surrounding streams (e.g., groundwater and surface water). Furthermore, AMD has the ability to solubilize and mobilize heavy trace metals contained in the tailings into the surrounding streams and soil. This can seriously affect the quality of the surface water and groundwater as well as the land. Pyrite ($FeS_2$) is the most common sulphide mineral found in mining waste (Lottermoser, 2010; Moncur et al., 2009). The oxidation of pyrite minerals is exothermic and a very complex process, which involves several reactions and intermediate steps. In general, the oxidation can be expressed in the following equation form (Bigham and Nordstrom, 2000):

$$FeS_{2(aq)} + 3.75O_{2(g)} + 3.5H_2O_{(l)} \leftrightarrow Fe(OH)_{3(s)} + 2H_2SO_{4(aq)} \quad (1)$$

Commonly, sulphide-rich tailings are stored under water in tailings disposal impoundments and/or covered with inert materials such as soil. The principle behind these disposal methods is to isolate sulphide bearing tailings from coming into direct contact with oxygen and/or water (triggering agents). As a result, the formation of AMD will be prevented or minimized to acceptable levels. However, the implementation of these traditional disposal methods has some environmental and economic issues (Bowker and Chambers, 2015). Along with more

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**Table 1**
Primary physical properties of tailings used in this study.

| Tailings | $G_s$ | $D_{10}$ (μm) | $D_{30}$ (μm) | $D_{50}$ (μm) | $D_{60}$ (μm) | $C_u$ | $C_c$ |
|---|---|---|---|---|---|---|---|
| ST | 2.7 | 1.9 | 9.0 | 22.5 | 31.5 | 16.6 | 1.3 |

stringent environmental legislations, these issues have driven miners and researchers to search for alternative disposal and storage techniques that are cost-effective, environmental and sustainable. As a result, several methods have been proposed and practiced globally to manage reactive mine wastes (e.g., sulphidic tailings). Among these alternative disposal methods, cemented paste tailings (CPT) or cemented paste backfill is a good means for managing sulphidic tailings in a more environmentally friendly manner. CPT has several advantages over traditional and alternative methods, such as reducing surface storage, improving mine production and reducing environmental impacts. The many advantages of CPT means that it has been widely and increasingly used in many underground mines around the world, such as in North America, Australia, China, and Turkey.

CPT can be simply described as a cementitious material that mainly consists of three ingredients, namely, thickened tailings (dominant material; solid percentage commonly between 75% to 85%), binder (commonly 3–7% by dry mass of tailings) which can be Portland cement solely or the partial use of mineral admixtures in lieu, and mixing water which can be either fresh or processing water. The fresh CPT mixture is transported by gravity and/or pumping through pipeline systems to fill the mined voids (stopes) in underground mines. This fluidic mixture will solidify with time and gain the necessary strength (mainly due to the progression of binder hydration) to stand like a "concrete" structure in underground mines.

Environmental performance and durability are two important aspects in the design of CPT structures. They are strongly influenced by the ability of the sulphide minerals (pyrite) to oxide (chemically react with oxygen) within the CPT system. This chemical reaction could lead to the generation of sulphate ions and acidity, and thus to the degradation of CPT through sulphate attacks and consequently, the formation of AMD (Tariq and Yanful, 2013; Fall and Benzaazoua, 2005). Therefore, chemical reactivity measurements (with respect to oxygen) of the CPT can be used as a good indicator for understanding the environmental performance of CPT that contain sulphate with respect to the potential generation of AMD.

Hence, there have been several direct and indirect methods proposed and conducted to determine the reactivity of mine waste or CPT, such as the sulphate release and oxygen gradient methods, and oxygen consumption (OC) testing (Elberling et al., 1994; Elberling and Nicholson, 1996; Ouellet et al., 2003, 2006). Among these different methods, OC testing is considered to be a direct technique that can be used to determine the reactivity of sulphidic mine waste by measuring the rate of oxygen consumption through the oxidation of sulphide minerals. Also, OC testing overcomes the drawbacks (commonly overestimation) of other methods, such as column leach and humidity cell tests (Schmieder et al., 2012). OC testing has been used in several previous studies to evaluate the oxidation rate of sulphide minerals (e.g., pyrite) and the reactivity of sulphide mineral-bearing tailings based materials and/or when incorporated into cemented paste backfill mixtures (e.g., Aldhafeeri and Fall, 2016; Aldhafeeri et al., 2016; Pokharel, 2008; Ouellet et al., 2003, 2006; Fall et al., 2004).

During the last decade, the majority of studies performed on CPT have mainly focused on the mechanical properties and behavior of CPT (e.g., Ghirian and Fall, 2016; Koohestani et al., 2016; Yılmaz et al., 2014; Fall et al., 2007). Some studies have investigated the reactivity of CPT systems by performing OC testing (e.g., Aldhafeeri and Fall, 2016; Aldhafeeri et al., 2016; Pokharel, 2008; Fall et al., 2004; Ouellet et al., 2003). These studies have examined the influence of a few factors on the reactivity of CPT including sulphide (usually pyrite) content, moisture content and temperature. They concluded that the reactivity of CPT systems increases with an increase in the sulphide content and decreases with increased moisture content. Aldhafeeri et al. (2016) also indicated that the reactivity of CPT is temperature dependent. However, despite these positive findings, there are still other important factors that can significantly affect the reactivity of CPT systems, and thus its environmental performance and durability. One of these key factors is the initial sulphate content of the CPT. CPT frequently contain various amounts of sulphate ions. These can originate from different sources, such as the pre-oxidized sulphidic tailings that are used in CPT mixtures, processing water with residual minerals in thickened tailings, gypsum ($CaSO_4 \cdot 2H_2O$) or anhydrite ($CaSO_4$) that are added in small amounts to the cement clinker to act as a setting regulator, and the mixing water which is either mine processing water or fresh water (Li and Fall, 2016; Fall and Pokharel, 2010). The amount of sulphate ions that are present in paste backfill systems vary from relatively low (<5000 ppm) to very high (≥25,000 ppm) (Fall and Pokharel, 2010). However, no study has investigated the effect of the initial sulphate of CPT on its reactivity. Most of the previous studies on the influence of the initial sulphate content on CPT have only focused on the effects on the mechanical properties (i.e., strength) of paste backfill structures for short- and long-term periods of time. Therefore, the main objective of this research is to experimentally study the effect of the initial sulphate content on the reactivity of mature CPT specimens by conducting OC testing. In addition to the OC tests, microstructural analyses are performed on selected specimens to gain a better understanding of the microstructural changes induced by the initial sulphate content and their impact on the reactivity of CPT.

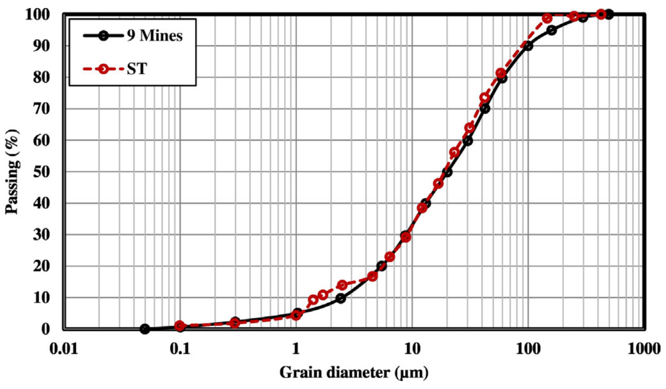

**Fig. 1.** Grain size distribution of ST and average grain size distribution of tailings from 9 mines in eastern Canada.

**Table 2**
Physical properties of pyrite.
(Source: Washington Mills North Grafton, Inc.)

| Bulk density ($g/cm^3$) | Density at 20 °C ($g/cm^3$) | Specific gravity | pH | Melting point (°C) |
|---|---|---|---|---|
| 2.35 | 4.7 | 4.6 | 4.0–6.0 | 1193 |

**Table 3**
Chemical properties of cement and mineral admixtures used.

| Binder | MgO (wt%) | CaO (wt%) | $SiO_2$ (wt%) | $Al_2O_3$ (wt%) | $Fe_2O_3$ (wt%) | $SO_3$ (wt%) | Rel. density |
|---|---|---|---|---|---|---|---|
| PC | 2.65 | 62.82 | 18.03 | 4.53 | 2.7 | 3.82 | 3.1 |
| SA | 10.98 | 41.14 | 34.23 | 9.54 | – | 3.87 | 3.3 |
| FA (Class C) | 5.58 | 21.47 | 38.06 | 19.45 | 5.33 | 2.7 | 2.6 |

Rel.: relative.