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GEOME 2
Manuscript received: February 5, 2021
Accepted: May 14, 2021

Geologica Macedonica, Vol. **35**, No. 1, pp. 59–66 (2021)
On print ISSN 0352 – 1206
On line ISSN 1857 – 8586
UDC: 669.3.049-048.583:691
DOI: 10.46763/GEOL21351373059mk

Original scientific paper

APPLICATIONS OF COPPER SLAG IN THE CONSTRUCTION SECTOR

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A b s t r a c t: Copper slags generated during the primary copper smelting have, due to its favorable mineralogical and chemical composition, several possible end uses in the resource demanding construction sector. Copper slags are made via two different processing methods; slow air cooling or water granulation. Air cooling results in final well crystalline and dense product, which is used usually as coarse aggregates in the building sector, however, water granulation results in a sand-like material with a high quantity of amorphous phase. The present paper gives and overview of the recent reports and good practices in the utilization of these slags, including also some drawbacks and potential issues in a certain type of copper slag, either air cooled or granulated and could serve as a guideline for selecting the most promising applications in the construction sector.

Key words: copper slags; construction sector; recycling; circular economy

INTRODUCTION

The pyrometallurgical production of copper inevitably results in a generation of 2.2 tonnes of slag per tonne of the produced copper. Yearly, a-proximate 24.6 million tonnes of copper slag is generated worldwide. This slag is composed generally of Fe_2O_3: 35–60 %, SiO_2: 25–40%, CaO: 2–10 %, Al_2O_3: 3–15 % (Shi et al., 2008) and is due to its composition named also as iron silicate or fayalitic slag.

The copper slags are usually generated via two different processing methods; air cooling and water granulation. The air cooling method consists of slow cooling of slag to ambient temperature with final well crystalline and dense product, which is used usually as coarse aggregates in the building sector. The second method, water granulation, is a form of rapid cooling with water quenching resulting in a sand-like material with a high quantity of amorphous phase. It is mostly used as abrasive but also could be used, if well crystalline, as fine aggregate in concrete and also in geotechnical and road pavement applications. If it is grinded, it can be used as a substitution to cement due to its pozzolanic characteristics and as a raw materials in glass-ceramics and ceramics industry (Dhir et al., 2017).

The construction sector is the biggest consumer of natural resources and the Waste Framework Directive 2008/98/ES (*EUR-Lex-32008L009 8-EN-EUR-Lex*, n.d.) has prescribed a new waste management hierarchy, where priority is given to reuse and recycling, which encouraged construction industry to use the alternative sources for construction materials in order to substitute virgin materials and to reduce the environmental impact in terms of lowering greenhouse gas emissions, energy consumption, pollution and waste disposal.

The European copper sector generates approximately 5 M tonnes of copper slag annually. The Whisper Project (https://whisper-project-eitrabwmaterials.eu/), funded by the EIT RawMaterials, is focused on the development of a more efficient pyrometallurgical system resulting in the copper slag produced with the air-atomization technology. Such slag could have, due to the air-atomization system modifications, tailored properties for a wider range of commercial applications. Furthermore and most important, it will lead to a minimization of the environmental footprint, reduction of energy and water consumption, reduction of waste waters produced while economic benefits will increase. To further explore new possibilities with slag produced via air-atomization technology, the envisioned applications in construction sector are focused on the natural aggregate replacement in concrete, substitution of cement, inorganic polymers, geotechnical fills.

The main outcome of this review is an overview of the recent reports and good practices in the utilization of the copper slag, including also some observed drawbacks and potential issues in a certain type of the copper slag, either air cooled or granulated. This review also could serve as a guideline for selecting the most promising applications in the construction sector and to envision some other applications with high added values. Nevertheless, copper slags in the cited articles are from different copper smelters and therefore could have minor variations in chemical and mineralogical composition due to the applied cooling or granulation process and this could affect their performance in the presented applications below.

COPPER SLAG PROPERTIES

Copper slags are generally solidified by fast cooling with water, forming an amorphous granulated slag. However, the slow solidification by air cooling would result in a dense, hard crystalline product. Nevertheless, the chemical composition, mineralogical composition and the respective microstructure of copper slag varies with the type of copper smelting procedure and source of copper concentrate. All parameters are key determining factors for copper and its potential reactivity when used as a supplementary cementitious composites, road pavements, geotechnical applications and ceramics.

In the following text, properties of water granulated slags and air (slow) cooled slags are presented. Granulated copper slag is composed of regularly shaped, angular particles, mostly between 4.75 mm and 0.075 mm (Gorai et al., 2003), with smooth surface, hard, dense and with low water absorption, low loss on ignition, low chloride and sulphate contents. The specific gravity is dependent on the iron content and varies from 2.8 to 3.8 g/cm^3. The water granulated copper slag is composed mainly of vitreous material (up to 80 wt.%) with some dendritic fayalite crystallites. Magnetite is an accessory phase (<5 vol%) that occurs as dispersed subeuhedral crystals, with an edge length up to 5 µm, accompanied by needle-like crystals of fayalite in some slower cooling crystalline domains of the slag (Figure 1) (Fernández-Caliani et al., 2012).

At the complete review of available data on granulated copper slag, the usual quantities of fayalite are from 4 % to 15 vol% (Dhir et al., 2017). The rapid cooling during water quenching does not allow time for crystallization to occur, which leads to the segregation of Al_2O_3, SiO_2, CaO and K_2O, and the presence of a much greater proportion of amorphous phases. Due to large content of amorphous phase, heavy and potential toxic elements are locked up in the structure and, therefore, copper slags are usually not classified as hazardous material, make it suitable for recycling and utilization in the construction sector and also in other industries, such as cement, glass and ceramic industry. The amorphous fraction affects the material reactivity and as such has important implications on the potential suitability of copper slags usage as a supplementary cementitious component.

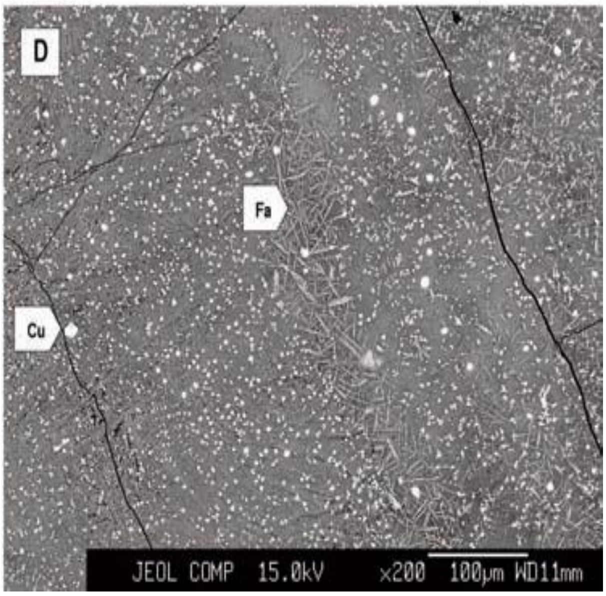

Fig. 1.: Granulated copper slag slag fayalite (Fa) and copper dropes (Cu) (Fernández-Caliani et al., 2012). \

The air (slow) cooled samples are exhibiting textured microstructure with highly elongated, skeletal crystals of fayalite, showing subparallel, radiating and branching growths of millimeter-scale. Magnetite occurs as tiny crystals (< 10 µm in size), often adopting a cruciform morphology, and infilling the hollows and gaps of the skeletal crystals of fayalite, with observed copper bearing particles (Fernández-Caliani et al., 2012) (Fig. **2**).

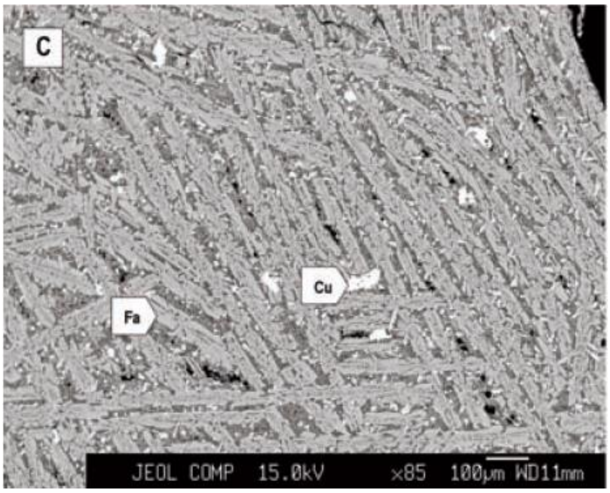

Fig. 2. Air cooled copper slag with fayalite (Fa) and copper drops (Cu) (Fernández-Caliani et al., 2012).

Review of recent research on air (slow) cooled copper slag samples showed that these have from 15-49% of amorphous phase with fayalite as the most abundant crystalline mineral phase, ranging from 45% to 57% (Dhir et al., 2017).

As water granulation is process consuming huge quantities of water, possible technological solutions are sought to achieve similar granulation results. The new air-atomization process, which is being tested through the Whisper project, will offer potential adjustable cooling rates with possibilities to make a copper slag with tailored properties for selected application in the construction sector as its utilization is dependent on its chemical and mineralogical composition as well as on microstructural properties.

APPLICATIONS IN INDUSTRY OF CONSTRUCTION MATERIALS

Addition to PC clinker

The chemical characteristics of water cooled copper slag are favorable for its utilization as a kiln feed, acting as a partial substitute for silica and iron sources in the raw mixture for Portland cement clinker manufacturing (Alp et al., 2008). The addition of copper slag in clinker manufacture slightly improves the raw mix burnability and does not generate any new unwanted phases. Consequently, recovery in cement kilns is currently mostly used co-processing of copper slag, but it is not a high added value application which is currently sought among copper smelters.

APPLICATIONS IN CONSTRUCTION SECTOR

Replacement for cement in cementitious composites

The utilization of copper as a replacement for cement has been studied by several authors (Afshoon & Sharifi, 2020; Mithun & Narasimhan, 2016; Murari et al., 2015; Shi et al., 2008). The Portland cement production is an energy intensive process composed of grinding of raw materials, their calcination at 1500 °C and then additional grinding of cement clinker and gypsum. It requires approximately 4000 MJ per tonne of cement and also causes significant contribution to the emissions of CO_2 (750 kg CO_2/tonne of produced cement in EU). Use of slags, which have pozzolanic properties as supplement to cementitious components would require only grinding/milling, therefore it would result into much lower energy consumption.

Several investigations have been conducted to determine the suitability of glassy copper slag as a partial replacement of cement. The use of glassy copper slag as a mineral admixture in concrete seems feasible and shows similar pozzolanic properties as fly ash from coal combustion (Sharma & Khan, 2017; J. Singh & Singh, 2019). Copper slag up to 15% cement replacement, increased the compressive and splitting tensile strength significantly (Tixier et al., 1997) and showed better mechanical and durability properties as compared to the normal concrete batches (De Rojas et al., 2008).

The recent study (Najimi et al., 2011) focused also on the application of copper slag for the effective reduction of the deteriorative sulfate expansions. In this regard, the partial replacement of cement with copper slag up to 15% led to more than 50% decrease in the sulfate expansion and no occurrence of ettringite destructive crystals (Najimi et al., 2011). Alternative cementitious material or inorganic polymers are getting lots of attention because they could substitute the ordinary Portland cement in selected applications. It was shown that slags with fayalitc composition could be activated by addition of alkali hydroxide or alkali silicate to form a solid with mechanical properties comparable to geopolymers, i.e. compressive strengths reaching up to 110 MPa (Kriskova et al., 2014; Onisei et al., 2012; Phair & Van Deventer, 2001; Pontikes et al., 2013). The amount of amorphous phase in the copper slag content strongly influences the final inorganic polymers properties due to higher availability of Al ions and formation of linkages in inorganic polymers, resulting in good mechanical properties (Pontikes et al., 2013). Research presented that dissolution tests with 10 M NaOH solution for 96 h reveal that approx. 54 wt% of Al and 33 wt% of Si of the total content become available for highly amorphous slag (above 80 wt%). On the contrary, the dissolved Al and Si are below 5 wt% when slag is slow cooled with amorphous phase content around 12 wt%.

Aggregate replacement in concrete (fine and coarse aggregate)

The need for more sustainable construction sector is also triggering an intensive search for alternative aggregate materials to replace natural aggregates. This would contribute to decrease in the sector dependence on natural resources. Copper slag has according to data in Table 1 similar physical characteristics as the natural siliceous aggregate (density, hardness). Nevertheless, the microstructure was not evaluated in most of reports, even if it is one of the most important parameters in the evaluation of the potential utilization as an aggregate.

Table 1

Properties of the granulated copper slag (compiled data from several studies retrieved from article (Shi et al., 2008), where various standards have been applied)
Copper slag properties (Shi et al., 2008).

Appearance	Black, glassy, more vesicular when granulated
Particle shape	Irregular, rounded
Density (Mg/m^3)	3.16–3.87
Water absorption (%)	0.15–0.55
Hardness (Mohs)	6–7
Water soluble chloride (ppm)	<50
Soundness (%)	0.8–0.9
Aggregate crushing value (%)	10–21
Aggregate impact value (%)	8.2–16
Abrasion loss (%)	24.1
Conductivity (µs/cm)	500

For utilization of copper slag as aggregate it must have eluates in the limits set in the national regulations.

To fully evaluate the possibility to utilize granulated copper slag as aggregate, tests on the resistivity to alkali silica reaction were performed. The alkali silica reaction (ASR) is a deleterious reaction in cement composites, caused by alkalis present in Portland cement, water and some types of siliceous aggregate, leading to a serious degradation of concrete. During this reaction a hydrophilic gel forms around aggregate grains leading to expansion and resulting in formation of characteristic cracks, building a path for further water penetration in concrete body and acceleration of the degradation. The report (Mavroulidou, 2017) assessing the ASR reaction in water granulated copper with gradation 0.2–2.5 mm performed according to standard ASTM C1260-01 (ASTM C1260 – 01 Standard Test Method for Potential Alkali Reactivity of Aggregates (Mortar-Bar Method), claims only minimal expansions well below standard limit of 0.1%, implying that copper aggregate is resistant to ASR. Nevertheless, to completely evaluate the level and certainly the rate of the ASR reaction, microstructure needs to be observed prior and after test period to assure no hydrophilic gel formation around siliceous aggregates with poor crystalline/amorphous microstructure.

Several researchers already have investigated the possible utilization of various copper slags as fine and coarse aggregates in ordinary concrete and its effects on the different mechanical and long-term properties of mortar and concrete (Al-Jabri et al., 2011; Gorai et al., 2003). Based on these studies, the overall performance of concrete with several percentages of substituted natural aggregate with copper slag was in most cases similar or better than ordinary concrete with only natural sand (Al-Jabri et al., 2011). Despite some benefits of using copper slag as fine and coarse aggregates, some negative effects have been reported, such as delayed setting time especially when only copper slag has been used as fine aggregate (Al-Jabri et al., 2009).

Several works reported that the compressive and tensile strengths of the concrete specimens made with copper slag fine aggregate are almost the same as that of normal concrete or even significantly higher than control mixtures (Fernández-Caliani et al., 2012; Kriskova et al., 2014; Onisei et al., 2012; Phair & Van Deventer, 2001; Pontikes et al., 2013). The evaluation of the effects of copper slag aggregate on the sulfate attack resistance and the depth of carbonation shows no significant influence (Najimi et al., 2011) and slower rate of carbonation by using copper slag.

The utilization of copper slag as an aggregate in the ultra-high performance concrete (UHPC) was also demonstrated and confirmed that the compressive strength after 28 days greater than 150 MPa at the optimum quantity of copper slag substitute 40 wt.%. However, the complete replacement of standard natural sand by copper slag resulted in a decrease in a 28-day compressive strength of about 15–25 %, whereas the flexural strength and fracture energy recorded were of the similar order. It can be concluded from the results that use of copper slag as a fine aggregate in UHPC is technically viable (Ambily et al., 2015).

The applications of copper slag with particle sizes from 2 mm to 10 mm as an aggregate were studied in the pervious concrete pavement (Lori et al., 2019). The used air cooled copper slag had well crystalized microstructure and according to XRD was mainly composed of fayalite (Fe_2SiO_4) and magnetite (Fe_3O_4). The results revealed that using copper slag generally led to an increase in the concrete strength because of the better mechanical and physical properties of the copper slag, namely better aggregate crushing value, good abrasion resistance and higher density compared to the conventionally used dolomite aggregate. However, such applications are viable for well crystallized coarse sized slags, as the vitreous microstructure affects the skid resistance and possess an enormous problem in the concrete pavement surfaces (Gorai et al., 2003).

Based on the above research results, it can thus be concluded that some copper slag might be used as a suitable substitute for natural aggregate and in the crystalline form could be used for pavement surfaces. Nevertheless, as for any other cases where recycled waste materials are suggested for use in concrete, the viability of using copper slag will depend on the local concrete production plants with availability of the material in sufficiently large quantities and in costs lower than for natural aggregates. In the case of amorphous copper slag it is necessary to analyze the durability of slag in alkaline media.

Use of copper slag as an aggregate in hot mix asphalt

The air (slow) cooled copper slag was reported to be used in hot asphalt mixes (Raposeiras et al., 2016b, 2018) (Figure 3). It had even better properties than traditional aggregates (limestone, igneous rocks) if it was initially air cooled and then crushed, possessing high angularity, resistance to wear, high density and hydrophobic properties (Dhir et al., 2017) suitable for applications in asphalt pavements. Nevertheless, the usage of granulated copper slag has not been reported, mostly due to its vitreous nature and issues with cracking, fragileness and sharp surfaces, which are not favorable in pavement surfaces.

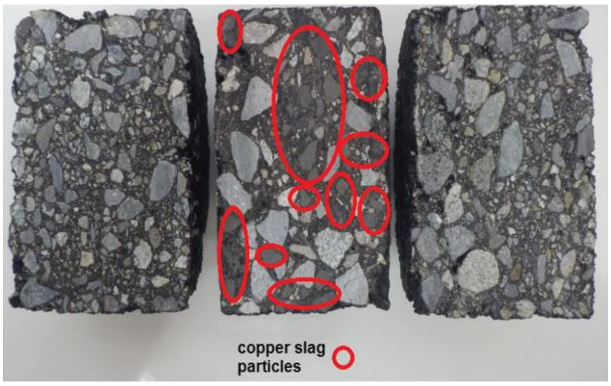

Fig. 3. Asphalt mix with slow air cooled copper slag aggregate (marked with red circles in the composite) (Raposeiras et al., 2016a).

Use of copper slag in geotechnical applications (structural fill material, granular base and embankment materials)

The potential to use copper slag in all forms, granulated, crushed (from slow cooling) and milled has been assessed for geotechnical applications, mainly in earthworks and river embankments.

The granulated copper slag physical properties, particle distribution (with wide grain size distribution), shear strength characteristics and chemical properties satisfy the standard specifications for a structural fill material recommended for use in mechanically stabilized earth (MSE) walls and reinforced soil slopes (RSS)(Das et al., 1983; Prasad & Ramana, 2016).

In order to avoid pollution of ground water at the application of granulated copper slag as a fill material, the leachates were analyzed and confirmed that the toxic compounds were below restricted limits (Reuter et al., 2004). The important factor that should be taken into consideration here is the vitreous nature of granulated slag, where heavy metals (Cu, Cd) are locked into the glassy structure.

On the other hand, copper slag particles in sizes below 100 μm could effectively reduce severely toxic Cr(VI) into Cr(III) in polluted waters

(Kiyak et al., 1999). The reduction reaction is possible due to presence of Fe(II) in the copper slags.

Incorporation into glass, tiles, bricks

The usage of copper slags in the production of tiles was researched on Iranian slags (Marghussian & Maghsoodipoor, 1999). The Iranian granulated copper slag was added to the mixture for unglazed floor tiles in maximum 40% mixed with clay and sand and sintered up to 1075 °C. The processed tiles possessed very low water absorption (2 wt%), hardness of 750 Vh and a very good acid resistance (Marghussian & Maghsoodipoor, 1999). Copper slags were also utilized as additive due to high content of iron in manufacturing of colored glasses and ceramic materials (Yang et al., 2013).

Special applications with high added value

The particular properties of copper slag based on fayalite and magnetite could serve as a potential source material for electromagnetic shielding concrete (Wanasinghe et al., 2020). Electromagnetic interference (EMI) shielding exhibits recent enormous interest due to the increasing abundance, sensitivity of electronics, potential issues with wireless communications and possible health threats due to the exposure to EM fields. Many researchers (Wanasinghe et al., 2020) already investigated EMI shielding cements with addition of steel or carbon fibers (Ameri et al., 2020; A. P. Singh et al., 2013), ferrite (Feng et al., 2020), and also coal fly ash (Guan et al., 2006). As most these materials and composites are expensive, usage of cheap or even waste materials, such as fly ash and copper slag would decrease the current high prices for production of EMI shielding concrete and furthermore, open possibilities for high added value applications of copper slag utilization.

In a recent study (Fan et al., 2018), the addition of 45 wt.% of granulated copper slag to the cement matrix weakens the electromagnetic wave approximately by 60%, as attributed to fayalite and magnetite acting as magnetic dielectric loss absorbents. Therefore, the successful application of granulated copper slag could be viable also for current coppers slags from Atlantic Copper and also the one processed with the air-atomization technology in Whisper project.

Another possible utilization of copper is also for special concrete used in marine applications as usage of copper results in increase of compressive strength, flexural strength and reduction of sorption and chloride ion penetrability values (Geetha & Madhavan, 2017).

CONCLUSIONS

Copper slag is a material obtained during copper smelting via pyrometallurgical route. It is usually classified as inert waste material and therefore could be recycled and utilized in the construction industry. This paper reviews the recent reports on utilization of copper in construction industry. The reviewed applications for copper slag envisioned possible utilizations in construction sector, limitations and potential issues.

Copper can be used as a substitution for cement (partial), natural aggregates (mostly fine), both in mortar and concrete, in asphalt mixtures, as a source for inorganic polymers, embankment, fill, glass, tiles and other specific applications with high added values. However, the final application will mostly dependent on the mineralogical composition and microstructural properties of air-atomized copper slags, their grain shape and resistance to alkaline media in the case of high quantities of amorphous phase. Nevertheless, as for any other cases where recycled waste materials are suggested for use in concrete, the viability of using copper slag will depend on the local construction sector needs and economic situation.

The air-atomization technology tested through Whisper project will offer opportunities for adjusting copper slag properties favourable for more applications in the construction sector. Furthermore, the new process will decrease costs and water consumption, contributing to the overall more sustainable and cost effective copper production.

Acknowledgements: The proposed review article was part of the activities in the frame of Whisper project (https://whisper-project-eitrawmaterials.eu/), funded by the EIT RawMaterials, part of the European Innovation Institute (EIT), funded by the European Union.

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Р е з и м е

ПРИМЕНА НА БАКАРНАТА ЗГУРА ВО ГРАДЕЖНИОТ СЕКТОР

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Клучни зборови: бакарна згура; градежен сектор; рециклирање; циркуларна економија

Бакарната згура создадена при примарното топење на бакарот, поради својот поволен минералошки и хемиски состав, има неколку можни крајни употреби во градежниот сектор кој постојано бара ресурси. Бакарната згура се создава преку два различни методи на обработка: бавно ладење на воздух или гранулација во вода. Ладењето на воздух резултира со финален добро кристализиран и густ производ, кој обично се користи како груб агрегат во градежниот сектор, додека гранулацијата во вода резултира со материјал сличен на песок со големо количество аморфна фаза. Овој труд дава преглед на неодамнешните извештаи и добрите практики во користењето на оваа згура, вклучително и некои недостатоци и потенцијални проблеми со одредени видови бакарна згура, било таа да е воздушно ладена или гранулирана во вода. Трудот може да послужи како упатство за избор на најперспективни примени во градежниот сектор.