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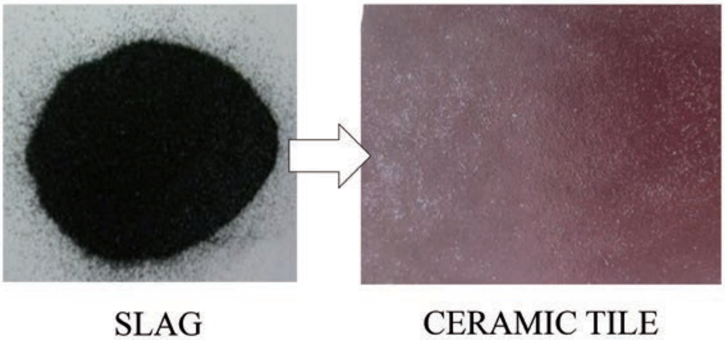
LIFE CYCLE ASSESSMENT OF A SINTERED TILE CONTAINING COPPER SLAG: IS RECYCLING ALWAYS CONVENIENT?

COPPER PRODUCTION CAUSES ENVIRONMENTAL PROBLEMS RELATED TO COPPER SLAG DISPOSAL. WITHIN THIS EXPERIMENTAL WORK, COPPER SLAG HAVE BEEN USED AS SECONDARY RAW MATERIAL FOR THE PRODUCTION OF SINTERED TILES, WHICH POTENTIAL ENVIRONMENTAL IMPACT WAS ASSESSED THROUGH THE LIFE CYCLE ASSESSMENT METHODOLOGY



Copper slag (CS) is a by-product of pyrometallurgical production of copper, generated during the melting of copper concentrates at a temperature of 1,200-1,300 °C [1]. During this process the slag containing iron and silicates are removed from the metal and then cooled. The cooling process transforms the slag in a glassy state and form an aggregate of variable dimensions.

During the last two centuries the worldwide production of copper has increased inexorably [2]. It has been estimated that between 1.6 and 2.2 ton of slag is generated for every ton of copper, creating approximately 24.6 million ton of slag each year, whose disposal causes environmental problems, together with the wastage of metal values they contain [3, 4]. For these reasons during last decades different uses of copper slags were investigated [3]. Particularly CS is mainly used to replace part of the sand or cement in concrete mixture or as abrasive material for removing rust and marine deposits on ship hulls. In spite of its potential applications, the disposal of CS in slag dumps and stockpiles is still practiced [3, 5]. The amount of generated waste have encouraged the community to find alternative solutions also considering the interesting physico-mechanical properties of CS that can be exploited to develop products with interesting final properties.

This study is a part of the WASTE^3 Project, which has investigated the possibility of reuse and valorisation of CS through the production of materials for residential applications, such as enamels, tiles and heating elements [6]. In the first part of the project, of experimental character, the technical feasibility to obtain products containing high amount of CS has been studied. In the second part the environmental impacts of these products and their feasibility from an environmental point of view has been assessed.

In particular, the aim of the present study is to assess the environment damage caused by the production of a sintered ceramic tile containing copper slag and to concurrently compare it with that caused by the traditional procedure, i.e. without any recycled material. Finally, the results were also discussed in comparison with those obtained from a previous study where the copper slag was used to produce frits for ceramic industries. In this way, by using the LCA approach as a decision-support tool, inefficient recycling can be avoided considering that the entire WASTE^3 project aims to identify the best strategy for the copper slag management in view of the increasing interest towards the environmental sustainability.

Methodology

The analysis was conducted with the Life Cycle Assessment methodology (LCA), in order to consider the whole life cycle of the tile, from the raw materials extraction, to the end of life phase thus obtaining a "cradle-to-grave" overview.

Life Cycle Assessment (LCA) is a tool for the systematic evaluation of the environmental aspects of a product or service system through all stages of its life cycle. It is

Tab. 1

Impact categories and Damage categories in the IMPACT 2002+ method [8]

Human toxicity (carcinogens and non carcinogens effects) (kg_{eq} chloroethylene) Respiratory inorganics (kg_{eq} PM 2.5) Ionizing radiation (kg_{eq} carbon-14) Ozone layer depletion (kg_{eq} CFC-11) Photochemical oxidation (respiratory organics) (kg_{eq} ethylene)	Human Health (DALY)
Aquatic ecotoxicity (kg_{eq} triethylene) Terrestrial ecotoxicity (kg_{eq} triethylene) Terrestrial acidification/nitrification (kg_{eq} SO_2) Aquatic acidification (kg_{eq} SO_2) Aquatic eutrophication (kg_{eq} PO_4) Land occupation (mq_{eq} organic arable land x yr)	Ecosystem Quality ($PDF \cdot m^2 \cdot yr$)
Global warming (kg_{eq} CO_2)	Climate Change (kg_{eq} CO_2)
Non-renewable energy (MJ primary non-renewable) Mineral extraction (MJ Surplus)	Resources (MJ)

standardized within the ISO 14040 series [7]. Following the Life Cycle Assessment methodology, after defining the goal and scope of the analysis for each case study, it was created an inventory for each product, containing inputs and outputs in terms of resources and energy consumptions, and environmental releases.

The environmental analysis was conducted using the SimaPro 7.3.3 and SimaPro 8.0.2 software, and IMPACT 2002+ as evaluation method. IMPACT 2002+ is the impact assessment method developed by the Swiss Federal Institute of Technology - Lausanne (EPFL), that proposes a combined midpoint/endpoint approach, linking all types of life cycle inventory results (elementary flows and other interventions) via fourteen impact categories to four damage categories, as shown in the Tab. 1 [8].

Copper slag were considered not as a waste but as a co-product of copper, which exits the system from which it has been generated to produce a secondary raw material, since copper slag have also a market value. This is a common approach in recent LCA studies [9] and it allows to attribute to the slag a part of the environmental impact due to copper production, and to consider this impact in the processes where the secondary raw material is used. This attribution has been carried out with an economic allocation, considering the quantity of copper slag produced for every ton of copper, and their prices.

As result of this economic allocation, in the considered processes the 0.85% of environmental damage due to the production of primary copper was attributed to copper slag.

Inventory

Copper slag used in the experimental part of the project are produced by a foundry located in Hamburg, where copper cathodes, rods and profiles are produced from raw materials coming from mines located primarily in countries outside Europe. The slag produced is then conferred to a subsidiary society, also located in Hamburg, where they are selected to be marketed in the form of crushed stone or granular material based on iron silicate, for different uses (embankment construction, anti-freeze layer in road construction, concrete additive, mineral abrasive).

The experimental phase of the project was carried by the University of Trento, and the final product obtained is composed by 96% of CS and 4% of sodium silicate.

The study has considered all processes involved from raw material extraction to disposal of the tiles, including the use phase. The functional unit of the analyzed system is 1 m^2 of tiles.

It was assumed that the process takes place in a ceramic plant for the production of ceramic tiles, in the province of Modena (Italy), where the raw materials are transported by road, stored in silos, mixed through mechanized systems (according to the formulation required) and then introduced into the milling plant. Further steps include pressing and sintering. The sintered tiles are then finished by cutting, to assure its geometrical dimensions. The modeled processes are described in the flow chart illustrated in Fig. 1.

To estimate the emissions of particulate and gaseous pollutants arising from the different stages of the manufacturing process, primary data were used when available, together with data from literature and data from regulations in air quality control, which limits were taken as emission levels [10-12].

Manufacturing process

It was considered that raw materials are shipped to the manufacturing plant by truck and stored in silos, then mechanically conveyed to the weighting area. The storage and handling of dry bulk materials have high potential of particulates and hazardous emis-

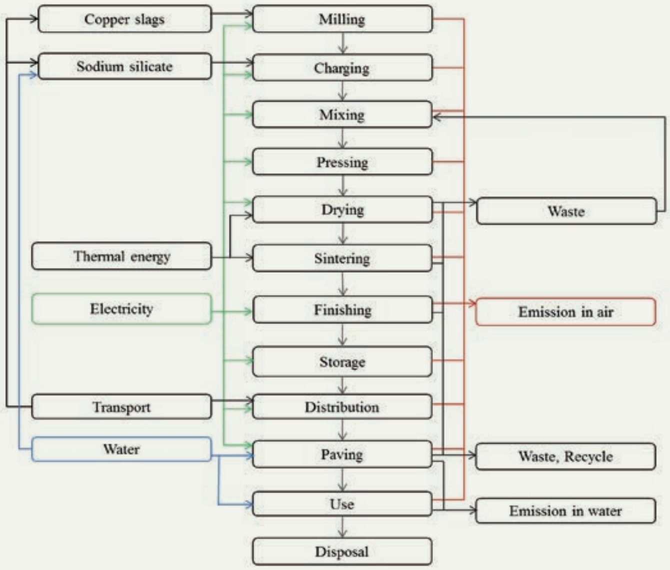


Fig. 1

Flow chart of the glazed tile manufacturing process

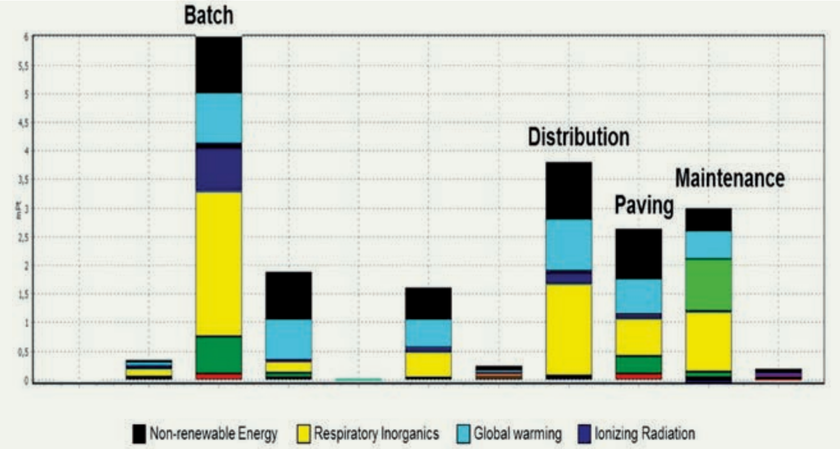


Fig. 2
Evaluation for impact categories of 1 m^2 of tile

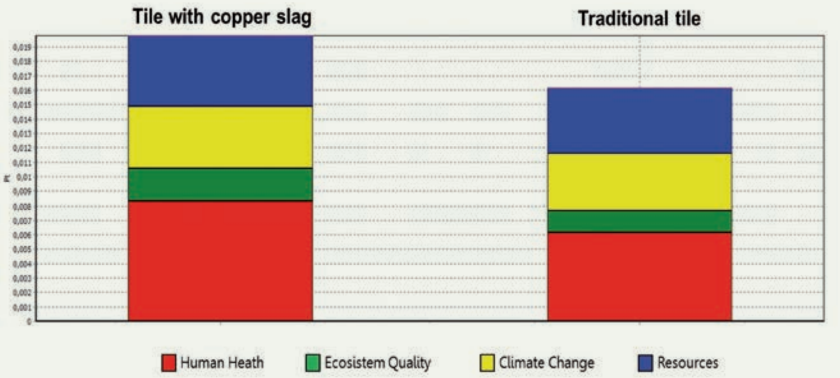


Fig. 3
Comparison by damage categories between CS tile and traditional tile

sions to air, consequently in this area filter systems can be used in order to reduce dust emissions below 10 mg/Nm^3 [11].

Subsequently the raw materials were mixed in a mill and then transferred to the pressing machine for the shaping. For this last step a hydraulic pressing machine with a power of 98 kW was used. A pressure of 15 MPa was applied in order to guarantee the maximum compaction of the raw materials and a production rate of 389 m^2/h of tiles. The obtained green bodies with a moisture content between 5 and 7 wt% are dried at 160 °C in order to reduce the water at 0.5 wt%.

Energy consumption at this stage is related to electricity for the plant operation as well as thermal energy used for combustion processes. The plant has a power of 43 kW, and the natural gas used is about 0,4 Nm^3 for each m^2 of produced tiles.

Half of the waste produced during this step, are recovered from the broken tiles to be reused as secondary raw material within the production cycle.

The green bodies are then sintered at 1,125 °C for 2 h in a roller kilns where the tiles advance on motor-driven rollers at a constant speed and pass though zones at different temperatures.

During this process a series of reactions take place that change the microstructure of the green body, creating the final tile with the required properties. Indeed, firing is the stage of the production cycle during which properties, shape and aesthetic and functional characteristics of the ceramic products become permanent.

The heat needed to sinter the material is supplied by natural gas-air burners located in the kiln walls and transmitted by convection. The gas required is 1,7 Nm^3 for 1 m^2 of produced tiles, while the electrical power of the kiln is 54 kW.

It was considered that the tiles broken during this phase are recovered by an external company, to be reused as secondary raw material.

The next stage is sorting the tiles and detect faulty pieces, through an instrument based on a series of pre-determined parameters which can recognize the defects that are created during the manufacturing processes. Tiles are divided into first, second and third choice, on the basis of the level of perfection achieved, to be allocated in different markets.

Then tiles are stored in cardboard boxes, which are tied with polypropylene strips. Afterwards the boxes are placed on wooden pallets and packed with a plastic film. Processes take place through mechanized systems and fork lift, so electricity and fuel consumption are taken into account. Materials used are recovered externally.

Use and end of life phase

Distribution process takes into account a supply scenario where 50% of the tiles are allocated to the European market, 20% to the North American market, 20% to the South American one, and 10% is shipped to Asia. Transport by truck and by ship were considered, taking into account average distances.

The phase of usage includes the processes of construction and maintenance of the floor tiles, whose life time is estimated at 40 years. Tiles are placed as a floor covering, with the use of adhesive and plaster. This process also considers the first cleaning operation, to remove traces of adhesive and plaster.

After the demolition of the floor, it is assumed that the tiles are allocated in a special landfill.

Tab. 2
Raw materials of traditional tile

Component	wt%
Clay	32
Feldspar	40
Sand	27.6
Fluidizer	0.4

Impact assessment

The analysis was carried out considering 1 m^2 of tile as a functional unit.

The results show that the damage is due mainly to the batch mixing process (30.44%) due to the high quantity of CS contained in the batch, as shown in Fig. 2.

The most affected damage category is the one related to Human Health (54.69%), because of the emissions of harmful substances as arsenic, sulphur dioxide and particulates which take place during copper production process.

Copper production is the major source of atmospheric arsenic and copper, about 50% of the global emissions [10], and since copper slags was considered a co-product of this process, it takes charge of part (0.85%) of the produced damage.

Comparative analysis

Another step of the study aims to assess the difference on environmental damage produced by the studied tile and a traditional one. The batch of the traditional tile considered is composed as illustrated in Tab. 2.

Also the comparative environmental assessment has been carried out with the evaluation method IMPACT 2002+ and considering as a functional unit 1 m^2 of tile. The results are shown in Fig. 3.

The total damage produced by the CS tile is 18.30% higher than the total damage produced by the traditional one. Therefore, the production of ceramic tiles using copper slag is definitively not convenient from an environmental point of view, if compared with the ceramic tiles obtained using the traditional raw materials.

However, it has been proved in a previous work [13] that the result changes by varying the final object of the work. Indeed, comparing the production of a ceramic frit using 70 wt% of CS with that of a traditional one (assuming the production of 1 ton of both) the LCA analysis demonstrated that the use of the slag as raw material leads to an environmental damage lower than 15.35%. Results are shown in Fig. 4.

Conclusions

The work demonstrated that the use of a secondary raw material is not always advantageous from an environmental point of view. Evaluating secondary raw material as a co-product of industrial processes means that the potential environmental impacts from the primary production process have to be considered.

The policy makers should therefore be aware of the possibility that replacing a virgin raw material with a waste based one not necessarily implies an increase of the environmental sustainability. Each case must be considered on its own and properly studied, with appropriate tools such as LCA methodology.

Acknowledgements

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REFERENCES

[1] T.E. Graedel *et al., Ecological Economics*, 2002, **42**(1), 9.
[2] R.B. Gordon *et al., Resources, conservation and recycling*, 2002, **36**(2), 87.
[3] B. Gorai, R.K. Jana, *Resources, Conservation and Recycling*, 2003, **39**(4), 299.
[4] R. Hischier, Life Cycle Inventories of Metals, Ecoinvent Centre, 2009.
[5] H.W. Kua, *Transactions of the Institute of Measurement and Control*, 2013, **35**(4), 510.
[6] C. Siligardi *et al., Ceramics International*, 2015, submitted for publication.
[7] ISO 14040:2006. Environmental management - Life cycle assessment - Principles and framework.
[8] S. Humbert *et al.*, Impact 2002+: User Guide, Quantis, 2012.
[9] H.W. Kua, *Journal of Industrial Ecology*, 2013, **17**(6), 869.
[10] B.M. Scalet *et al.*, Best Available Techniques (BAT) Reference Document for the Manufacture of Glass. JRC Reference Report. European Commission Joint Research Center, 2013.
[11] Giunta Regionale Direzione Generale Ambiente della Regione Emilia Romagna. Determinazione n° 004606/1999, Indicazioni alle Province per il rilascio delle autorizzazioni alle emissioni in atmosfera. Bologna, 1999.
[12] United States Environmental Protection Agency (EPA) - AP 42, Fifth edition, Compilation of Air Pollutant Emission Factors, Vol. 1: Stationary Point and Area Sources. Cap 11.4 Frit Manufacturing. Research Triangle Park, N.C., 1997.
[13] S. Mohaddes Khorassani *et al.*, Analisi ambientale di smalto ceramico ottenuto da scorie derivanti dalla produzione del rame, VIII Convegno della Rete Italiana LCA, Firenze, 2014.

Analisi del ciclo di vita di una piastrella sinterizzata contenente scorie di rame: riciclare è sempre conveniente?

La produzione di rame è causa di problemi ambientali legati allo smaltimento delle scorie. Nell'ambito di questo studio sperimentale, le scorie di rame sono state utilizzate per la produzione di piastrelle sinterizzate, valutandone il potenziale impatto ambientale attraverso la metodologia LCA.

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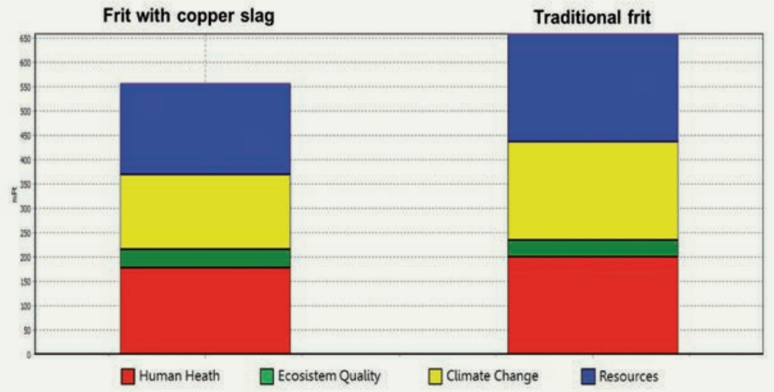

Fig. 4

Comparison by damage categories between CS frit and traditional frit