Sustainable use of (cleaned) sulphidic mining waste in building ceramics

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European Training Network for the Remediation and Reprocessing of Sulphidic Mining Waste Sites

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Sustainable use of (cleaned) sulphidic mining waste in building ceramics

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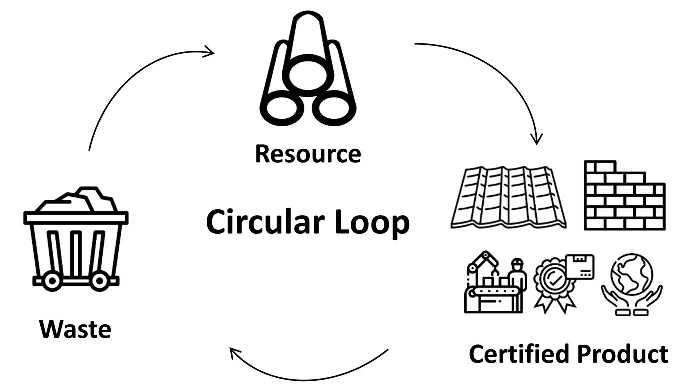
Rationale

Mining and quarrying waste is the second biggest waste stream in the EU-27 and, according to Eurostat, in 2018 it represented 27 wt% (621 Mt) of the total waste output. The space shortage, landfill regulations and costs, resource scarcity, environmental and health hazards, and social inequality of nearby population make this type of waste, especially sulphidic mining waste, prone to acid mine drainage, a key priority for a more sustainable and resource efficient Europe.

Main goal

To investigate the possibilities to replace primary raw materials used in building ceramics by (cleaned) sulphidic mine waste materials, taking into account production processes behaviour, product quality criteria, and environmental compliance.



Milestones

- Develop building ceramics in which primary raw materials, such as clay and sand, are partly or totally replaced by (cleaned) mine waste materials (0-40%), matching production and product quality criteria.
  - ✓ **Roof tiles:** 5, 10 and 20 wt% of uncleaned mine waste partly replacing local clay and sand;
  - ✓ **Inner-wall blocks:** 10 and 20 wt% of uncleaned mine waste totally replacing local sand and partly replacing imported filler;
  - ✓ **Pavers:** 10 and 20 wt% of uncleaned mine waste partly replacing the ready-to-use paver mix;
  - ✓ **Facing bricks:** 20 and 40 wt% of uncleaned and cleaned mine waste partly replacing local loam and imported filler.
- Meet raw materials and building products requirements of Flemish (VLAREMA) and European (Landfill Directive) environmental regulations, and perform a life cycle assessment (LCA) of waste-derived ceramics.
  - ✓ **VLAREMA tests** (total metal(loid)s and organic compounds, column leaching test) performed on mining waste materials for potential use as non-shaped building materials;
  - ✓ **VLAREMA tests** for service life (diffusion leaching test) and second life (column leaching test) assessment of standard and waste-derived ceramics;
  - ✓ **Landfill directive test** for landfill disposal (batch leaching test) assessment of mine waste materials, standard and waste-derived ceramics;
  - ✓ **Life-Cycle Assessment (LCA)** of standard and waste-derived ceramics (cradle-to-gate scenario).

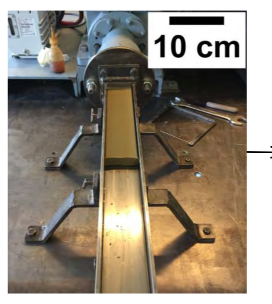
Materials, Methods and Results

1. Characterisation of (cleaned) mine waste materials

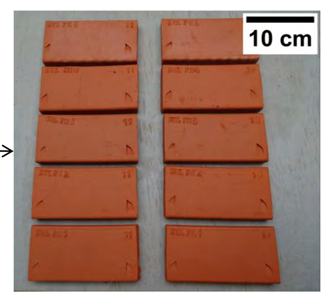
Sample picture			
Sample location	Plombières (Belgium)	Neves Corvo (Portugal)	Neves Corvo (Portugal)
Sample code	SUL_PL_62_I[1]	SUL_NC_01_CL_FLOT[2]	SUL_NC_03_CL_FLOT[2]
Sample description	Uncleaned stored tailing (pond)	Cleaned fresh waste-rock (mine flow)	Cleaned stored waste-rock (open-air pile)
Geochemistry (Carbon + Sulphur)	**NDIR (firing at 1450°C and flow ± 2.5 lpm, based on ISO 10694-1995 §7.2)**		
	wt%	wt%	wt%
C	0.1	0.4	0.5
S	0.01	0.7	1.0
Geochemistry (Trace elements)	**ICP-OES ($HNO_3/HClO_4$/HF digestion, based on ISO 14869-1:2001)**		
	mg/kg	mg/kg	mg/kg
Ba	366.3	408.6	444.2
Co	19.6	31.1	16.0
Cr	60.4	127.5	91.2
Cu	23.1	401.6	1180.5
Ni	30.6	48.7	35.9
Pb	29.8	776.7	349.3
Zn	136.8	1299.5	462.0
Geochemistry (Soluble sulphates)	**IC (H_2O_2/Na_2CO_3-$NaHCO_3$ solution, based on NBN EN ISO 10304-1 (2009))**		
	wt% (18 g/150 ml)	wt% (20 g/200 ml)	wt% (20 g/200 ml)
SOx	0.01	0.40	0.73

Grain size distribution (mechanical shaker + sedigraph), mineralogy (XRD quantitative) and major elements composition (XRF Omnian) analyses were also performed. Environmental characterisation results of mine waste materials are shown in section 5.

2a. Preparation of vacuum-extruded test pieces (roof tiles and inner-wall blocks)

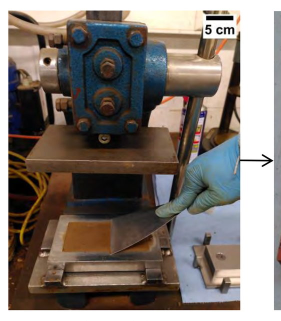


Vacuum extruder

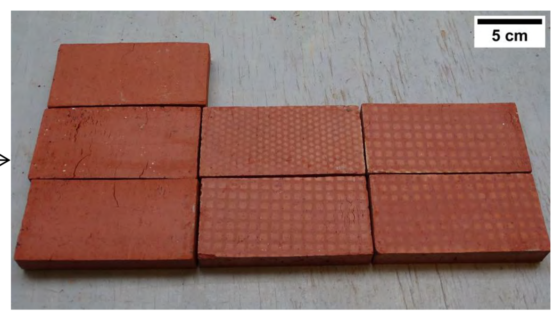


Fired test pieces

2b. Preparation of hand-molded test pieces (pavers and facing bricks)



Hand-mold press

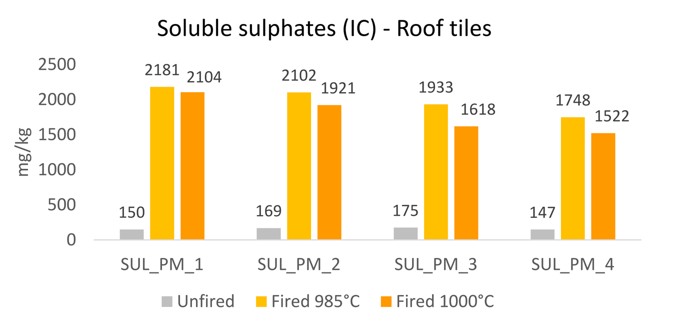


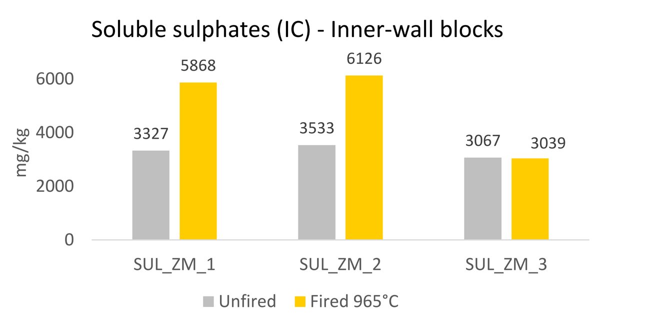
Fired test pieces

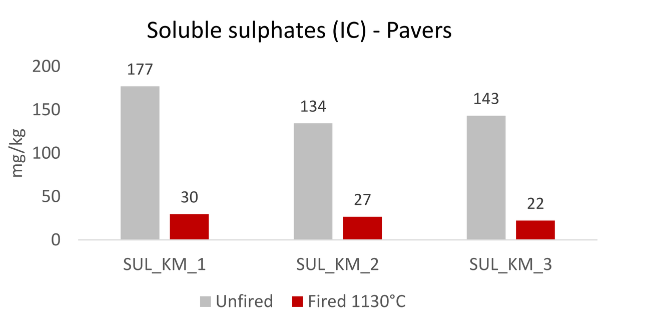
3. Composition and technical properties of ceramic test pieces

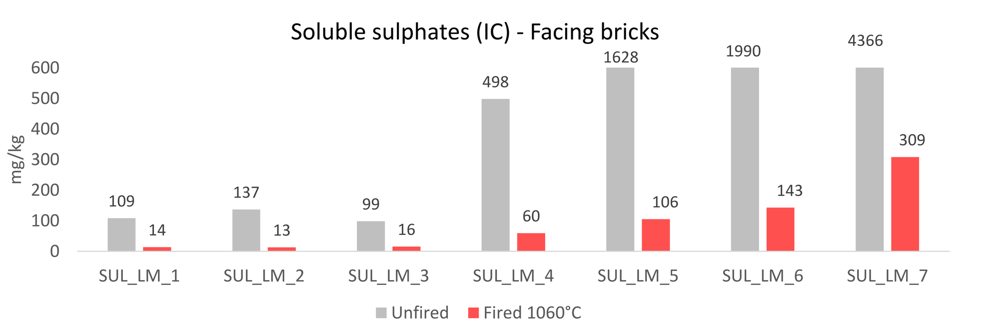
Blend codes	Composition	Water absorption	Saturation level	E-modulus
		wt%	%	GPa
Roof tiles		**Lab fired 985°C / 1000°C**		
SUL_PM_1	Reference (standard)	7.2 / 6.2	71 / 66	18.1 / 19.6
SUL_PM_2	5 wt% SUL_PL_62_I	7.5 / 6.6	73 / 68	17.4 / 19.3
SUL_PM_3	10 wt% SUL_PL_62_I	8.0 / 7.1	74 / 70	16.5 / 18.1
SUL_PM_4	20 wt% SUL_PL_62_I	8.7 / 7.9	78 / 74	15.0 / 16.8
Inner-wall blocks		**Lab fired 965°C**		
SUL_ZM_1	Reference (standard)	10.5	NA	17.6
SUL_ZM_2	10 wt% SUL_PL_62_I	10.5	NA	18.2
SUL_ZM_3	20 wt% SUL_PL_62_I	12.6	NA	13.5
Pavers		**Lab fired 1130°C**		
SUL_KM_1	Reference (standard)	3.0	NA	27.6
SUL_KM_2	10 wt% SUL_PL_62_I	1.7	NA	28.6
SUL_KM_3	20 wt% SUL_PL_62_I	1.9	NA	30.5
Facing bricks		**Lab fired 1060°C**		
SUL_LM_1	Reference (standard)	8.1	NA	11.4
SUL_LM_2	20 wt% SUL_PL_62_I	7.7	NA	13.4
SUL_LM_3	40 wt% SUL_PL_62_I	6.8	NA	15.2
SUL_LM_4	20 wt% SUL_NC_01_CL_FLOT	6.5	NA	18.4
SUL_LM_5	40 wt% SUL_NC_01_CL_FLOT	4.9	NA	28.3
SUL_LM_6	20 wt% SUL_NC_03_CL_FLOT	7.0	NA	17.0
SUL_LM_7	40 wt% SUL_NC_03_CL_FLOT	6.1	NA	21.8

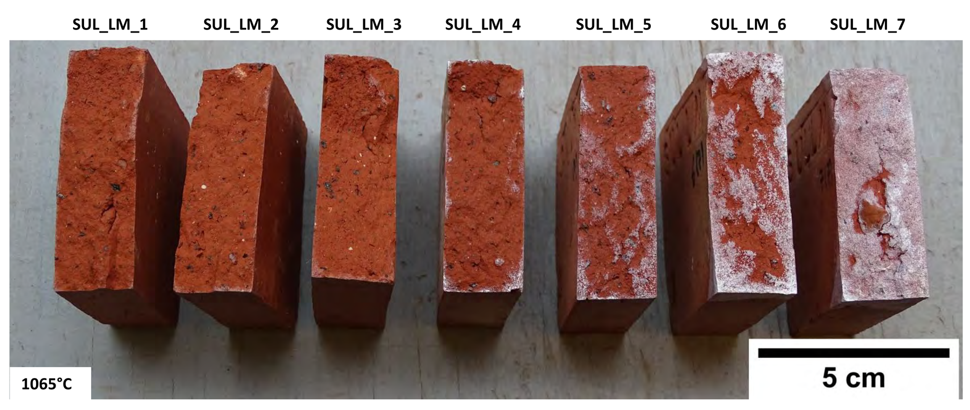
4. Chemical and aesthetical properties of fired ceramic test pieces











Efflorescence test result - Facing bricks

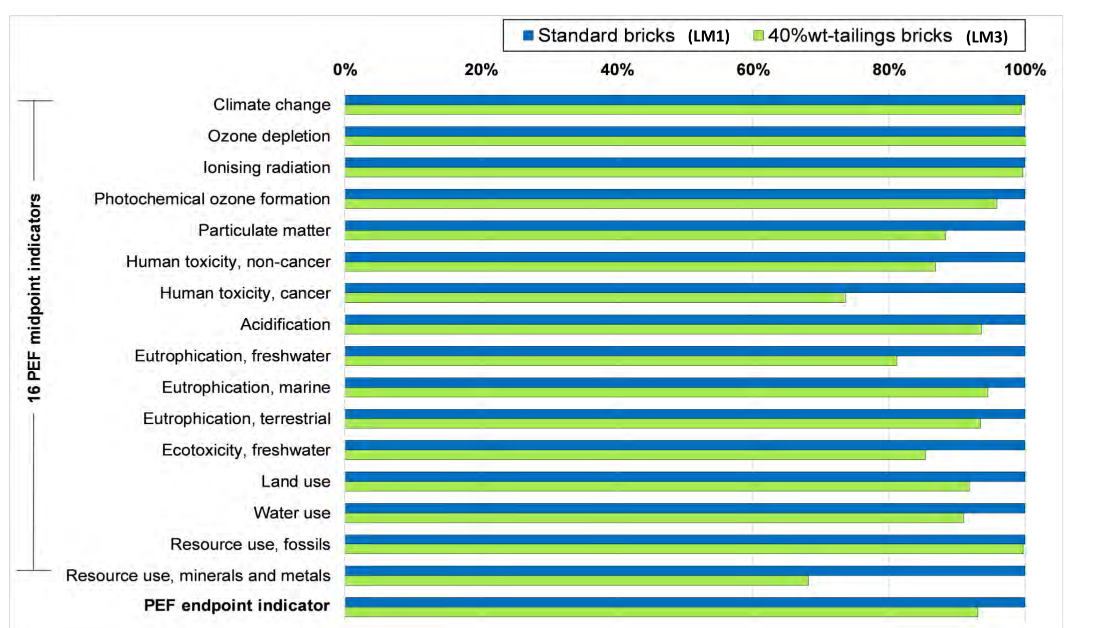
5. Environmental regulatory tests on mine waste materials and ceramic test pieces

Diffusion/tank leaching test

Column leaching test

	pH	Cumulative release quantity of metal(loid)s							
		As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Diffusion leaching test (service life)[3]	**L/A=80 l/m²**	**mg/m²**	**mg/m²**	**mg/m²**	**mg/m²**	**mg/m²**	**mg/m²**	**mg/m²**	**mg/m²**
Limit values (VLAREMA, 2012)		285	12	555	255	8.2	136	609	924
SUL_LM_1 (1060°C)	6.3	7.5	<LOD	<LOD	<LOD	NM	<LOD	<LOD	<LOD
SUL_LM_3 (1060°C)	6.1	7.0	<LOD	<LOD	<LOD	NM	<LOD	<LOD	<LOD
SUL_LM_5 (1060°C)	6.0	34.7	<LOD	<LOD	7.20	NM	<LOD	<LOD	2.00-6.80
SUL_LM_7 (1060°C)	6.2	**350.0**	<LOD	<LOD	1.10-3.50	NM	<LOD	<LOD	<LOD
Column leaching test (second life)[4]	**L/S=10 l/kg**	**mg/kg**	**mg/kg**	**mg/kg**	**mg/kg**	**mg/kg**	**mg/kg**	**mg/kg**	**mg/kg**
Limit values (VLAREMA, 2012)		0.8	0.03	0.5	0.5	0.02	0.75	1.3	2.8
SUL_PL_62_I	7.6	<LOD	<LOD	0.055-0.065	<LOD	<LOD	<LOD	<LOD	0.16-0.18
SUL_NC_01_CL_FLOT	7.8	0.001-0.04	0.01	<LOD	0.004-0.05	0.00012-0.0005	0.10-0.12	0.002-0.05	**4.35**
SUL_NC_03_CL_FLOT	5.4	0.004-0.04	**0.04**	<LOD	**1.40**	<LOD	**0.80**	0.001-0.1	**7.91**
SUL_PM_1 (985°C)	8.2	0.64	<LOD	0.18	0.001–0.03	NM	0.01–0.03	<LOD	0.03
SUL_PM_4 (985°C)	8.2	0.32-0.40	<LOD	0.27-0.32	<LOD	NM	<LOD	<LOD	<LOD
SUL_ZM_1 (965°C)	8.2	0.32	<LOD	0.12	<LOD	NM	0.02–0.04	<LOD	0.02
SUL_ZM_3 (965°C)	8.1	0.13-0.21	<LOD	0.18-0.23	<LOD	NM	<LOD	<LOD	<LOD
SUL_KM_1 (1130°C)	8.6	0.42	<LOD	0.002-0.10	0.002-0.10	NM	<LOD	<LOD	<LOD
SUL_KM_3 (1130°C)	8.6	0.63	<LOD	0.003-0.10	0.001-0.10	NM	<LOD	<LOD	<LOD
SUL_LM_1 (1060°C)	7.3	0.40	<LOD	0.001-0.10	0.001-0.10	NM	<LOD	<LOD	0.004-0.20
SUL_LM_3 (1060°C)	6.8	0.47	<LOD	0.001-0.10	<LOD	NM	<LOD	<LOD	<LOD
SUL_LM_5 (1060°C)	8.7	**2.49**	<LOD	0.03-0.12	0.002-0.10	NM	<LOD	<LOD	<LOD
SUL_LM_7 (1060°C)	7.6	**20.41**	<LOD	0.005-0.10	0.01-0.10	NM	<LOD	<LOD	0.01-0.20

6. Life-cycle assessment (cradle-to-gate) of facing brick test pieces



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During this PhD research around 120 trees were planted. Join the movement and use ecosia.org as your default web search engine for surfing the web while planting trees! 45 searches = 1 tree

[1] Dried 105˚C overnight.

[2] Crushed + sieved (9 and 2.5 mm) + dried 45°C + grinded (ball mill) + wet sieved (87 µm) + wet grinded (<65 µm) + froth flotation (KAX 50g/t, MIBC 100 g/t, pH 9, S/L= 300 g/l).

[3] Cumulative release of metal(loid)s from leachates collected during diffusion leaching test, performed according to method CMA/2/II/A.9.1, based on NEN 7375:2004 with L/A_{cum}= 80 l/m² (8 fractions, 64 days). Limit values, excluding Hg, according to VLAREMA (2012) Annex 2.3.2.C (immission in mg/m² over 100 years).

[4] Cumulative release of metal(loid)s from leachates collected during the column leaching test, performed according to method CMA/2/II/A.9.1, based on NEN 7373:2004 with L/S_{cum}= 10 l/kg (7 fractions, 21 days). Limit values, excluding Hg, according to VLAREMA (2012) Annex 2.3.2.B (leachability in mg/kg).

NA: not applicable; S/L: solid/liquid ratio; L/S: liquid/solid ratio; L/A: liquid/area ratio; LOD: limit of detection; NM: not measured (ceramic blends were fired between 965°C-1130°C, thus no mercury should be present due to its boiling point being at around 360°C); PEF: product environmental footprint.

Conclusions

- The **uncleaned Plombières tailing material** can be used directly (without the need of any pre-treatment):
  - ➢ As non-shaped building material (e.g., cover layers, earth-rock filled dams);
  - ➢ In shaped building products, such as roof tiles (5 wt%), inner-wall blocks (10 wt%), pavers (10 and 20 wt%), and facing bricks (20 and 40 wt%), without compromising the production processes, product quality criteria and environmental performance during service life (application), second life (recycling) or at the end-of-life (landfill). For the Plombières tailing-derived (40 wt%) bricks, a cradle-to-gate LCA showed environmental benefits when compared to the standard bricks (1/3 reduction of natural resource usage, 10% reduction of particulate matter emissions, human and eco-toxicity, and eutrophication.
- The **(cleaned) Neves Corvo waste rock materials** cannot be used as non-shaped building materials, due to leaching of metals above regulatory limits. When incorporated in a specific blend for facing bricks (20 and 40 wt%), the waste rock materials induced aesthetical and environmental problems in the fired bodies, mainly due to their still high content in metal(loid)s, sulphur and soluble sulphates.

Further perspectives

- In order to incorporate higher percentages of **Plombières tailing material** in roof tiles (10 and 20 wt%) and blocks (20 wt%), new blends should be worked out. The **(cleaned) Neves Corvo waste rock materials** are not yet suitable for building ceramics; therefore an optimisation of the cleaning procedure needs to be performed before incorporation in new ceramic blends.
- This research demonstrated the added value of a complete characterisation of alternative raw materials, such as mining waste, in order to assess their potential in replacing primary raw materials in building ceramics and understand their behaviour, not only considering production and product quality criteria but also environmental performance of waste-derived building ceramics. The economic benefit of (re)mining such alternative materials for the ceramic industry is essential to complement the waste valorisation assessment.
- The methodological approach used in this research for the characterisation of waste materials and waste-derived building ceramics can be applied to any mine waste material for potential use in building ceramics.

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