Supplementary materials for

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Potential of mechanochemically activated sulfidic mining waste rock for alkali activation

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A. lab-scale analysis of feeding materials for establishing HSC Sim Flowsheet simulations

Figure S1 and Table S1 show particle size distribution and mineralogy composition of different size classes. Table S2 to S3 indicate the basic information to establish the simulated grinding process via HSC Sim.

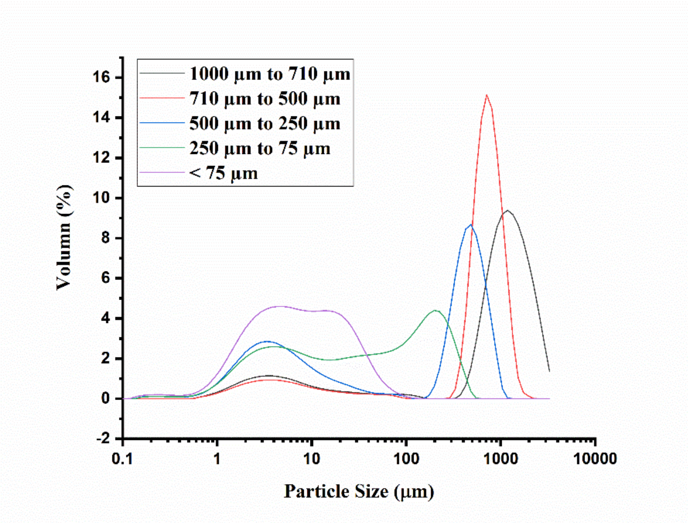

Figure S1. Particle size distribution of initial feed mining waste rock.

Table S1 Mineral composition of each sieving classes by QXRD

Phase Name	> 1000 µm	710 µm - 1000 µm	500 µm - 710 µm	250 µm - 500 µm	75 µm – 250 µm	< 75 µm
	wt% Rietveld	wt% Rietveld	wt% Rietveld	wt% Rietveld	wt% Rietveld	wt% Rietveld
Inert minerals	52.7	59.1	58.6	63.9	52.5	52.9
Aluminosilicate minerals	47.3	40.9	41.4	36	47.6	47

Table S2 Mineralogy of input data for simulation (Stoichiometric minerals are used, taken from HSC Geo module)

Element	Qtz	Py	Gyp	Ms	Cch
O	53.26		55.76	48.20	51.82
Si	46.74			21.15	15.16
Fe		46.55			
S		53.45	18.62		
Ca			23.28		
H			2.34	0.51	1.45
Al				20.32	9.71
K				9.82	
Mg					21.86

Mineral	Bulk	Unit
Quartz	48.62857	%
Pyrite	6.114286	%
Gypsum	1.314286	%

Muscovite	6.942857	%
Clinochlore	37	%

Table S3 Suitable equipment according to simulation process

Simulation unit	Upscaled equipment	Vendor	Power consumption
Jaw Crusher	Pilot Crushtec VS100	Metso Outotec	55 kW
Double Roller Crusher	Φ 600×750	Haiwang	22 kW
Sieving	ZKJ1007-D5	Haiwang	1.8 × 2 kW
Pump Sump	40ZJ-A19	Haiwang	6.25 kW
Hydro cyclone	SkimAir 80	Metso Outotec	11 kW
HIGmill	Outotec HIGmill 130	Metso Outotec	130 kW
Conditioner	OKTOP 3200	Metso Outotec	2.2 kW
Thickener	NZS6	Metso Outotec	6 kW
Pressure Filter	OUTOTEC LAROX PF 12 SERIES	Metso Outotec	33 kW

B. Further descriptions of HSC Sim 10 Flowsheet simulations and the HIGmill equipment

In the first configuration (route 1), the coarse waste rock is crushed by both jaw crusher and double roller crusher, followed by a sieving process set at 250 µm to maximize the aluminosilicate-rich minerals (muscovite and clinochlore) which content increases in the fraction finer than 250 µm, thereafter, the stream of aluminosilicate-rich minerals flows to a HIGmill for mechanochemical activation preventing the useless energy consumption on inert minerals, such as quartz, which is not the reactive precursor. Nevertheless, those inert minerals can be both coarse aggregates in geopolymer mortar manufacturing rather than waste. The special design of HIGmill (see further details below) requires the feed stream to be in a slurry form; therefore, a cyclone equipment can be used to split the ultra-fine particles prior to HIGmill to avoid the unnecessary grinding. In addition, a large proportion of dense stream flows to HIGmill to be mechanochemically activated. Finally, all the slurry flows to the thickener through a conditioner and it is possible to obtain the low-water-content products for subsequent alkali activation after drying treatment.

The HIGmill (Metso Outotec Oy) is fine and ultra-fine grinding mill technology, that uses gravitational forces and proprietary rotor technology to produce a finer grind for mineral liberation. This enables chemical activation processes to occur, as assumed in the process modelling in section 2.5 LCA [1].

The model is optimized by replacing the thickener with pressure filter and additive of water recycling system where the cyclone was remained (route 2). In this modified simulation, the operational environment is considered; for instance, it is difficult to maintain the thickener in extreme places such as Finland where the temperature is practically below zero-degree Celsius during the whole wintertime. Water in the thickener will be frozen, thereby blocking the whole process. Water recycling is another critical issue according to the sustainability of the process. Therefore, the pressure filter can be a promising equipment for solving both problems of maintenance and sustainability. The cake is produced as the final product for subsequent geopolymer manufacturing, while a large proportion filtrate is able to be recycled to pump sump 1.

The third optimized model (route 3) is achieved by adding an additional sieving machine to substitute the function of a cyclone in the second route, which is expected to separate particles in a dry form. Thus, the ultra-fine particles directly flow to the product tank whereas relatively coarse particles flow to HIGmill for mechanochemical activation, thereby saving the energy and improving the efficiency. Although it is possible to screen ultra-fine particles especially the size < 75 µm, the dry residues can easily clog the mesh holes compared to the wet cyclone separator. It is also important to take water recycling into account by replacing the thickener with pressure filter. In summary, these are the overall findings.

- Mass flow of products for geopolymer binder making: Different model indicates the mass flow of the mineral processing, which is basically from crusher, sieving to HIGmill. All the models have the same amount of initial waste rock stream of 100 t/h; however, different amounts of products are generated. The sieving process produces undesired residues of inert minerals such as quartz and pyrite, which can be the aggregates in the subsequent geopolymer mortar manufacture.
- Water recycling: Route 1 is the only model which has no water recycling. It means that a considerable amount of water becomes waste as the tails. The solid/water ratio was fixed at 1 prior to the slurry 1 flows to cyclone since it prevents the clog of cyclone equipment. In terms of water consumption, nearly 90% water is recycled in the other two configurations (i.e., route 2 and 3). As for waste water, it can be purified after specific treatments such as separation, oxidation and neutralization [2].
- Electricity consumption: The suitable equipment in the entire process is selected according to the mass balance and flow of the simulation, where the supporting technical sheet provides the equipment information. Route 1 has the lowest electricity consumption, whereas route 2 has the highest power utilization resulting from the higher energy demand for pressure filter, cyclone, and extra pumps. However, when it is normalized to the production of precursors, the results are in reverse. Larger mass flow of products in route 2 and 3 imply lower overall energy consumption, despite the higher electricity inputs due to the presences of additional processing equipment.

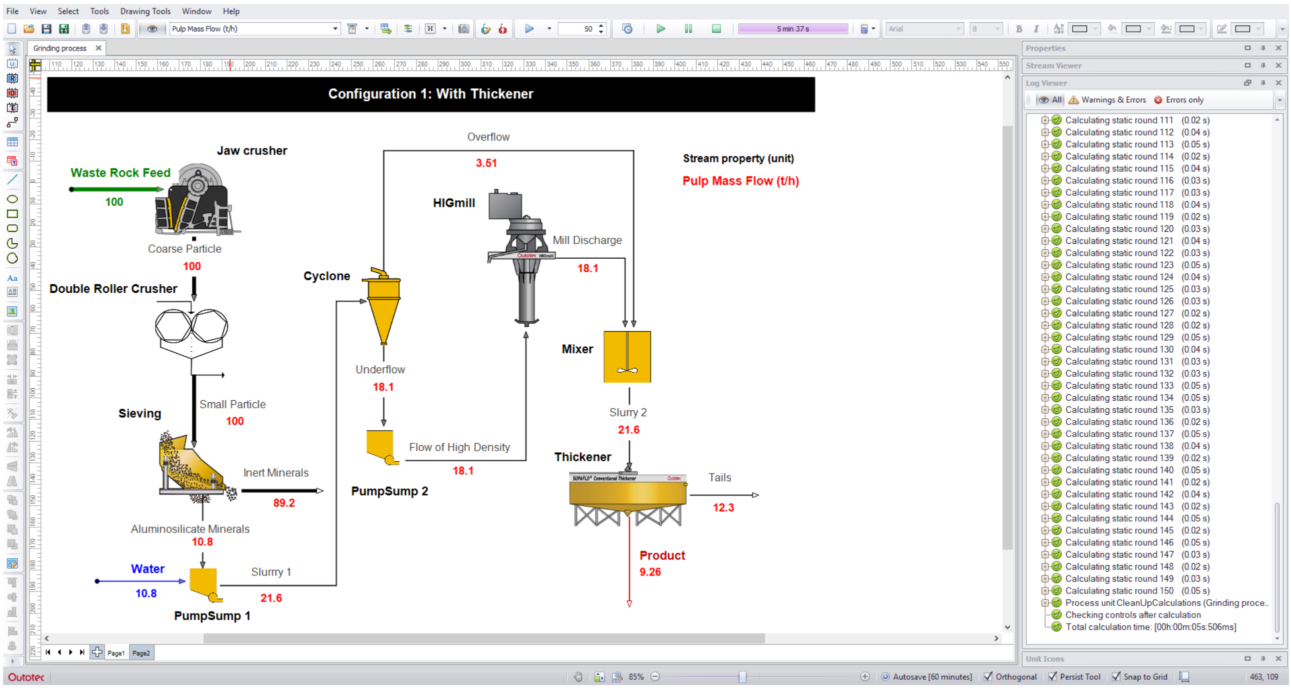

Figure S2. Configuration 1 of mechanochemical activation: Flowsheet simulation in HSC Sim (Route 1).

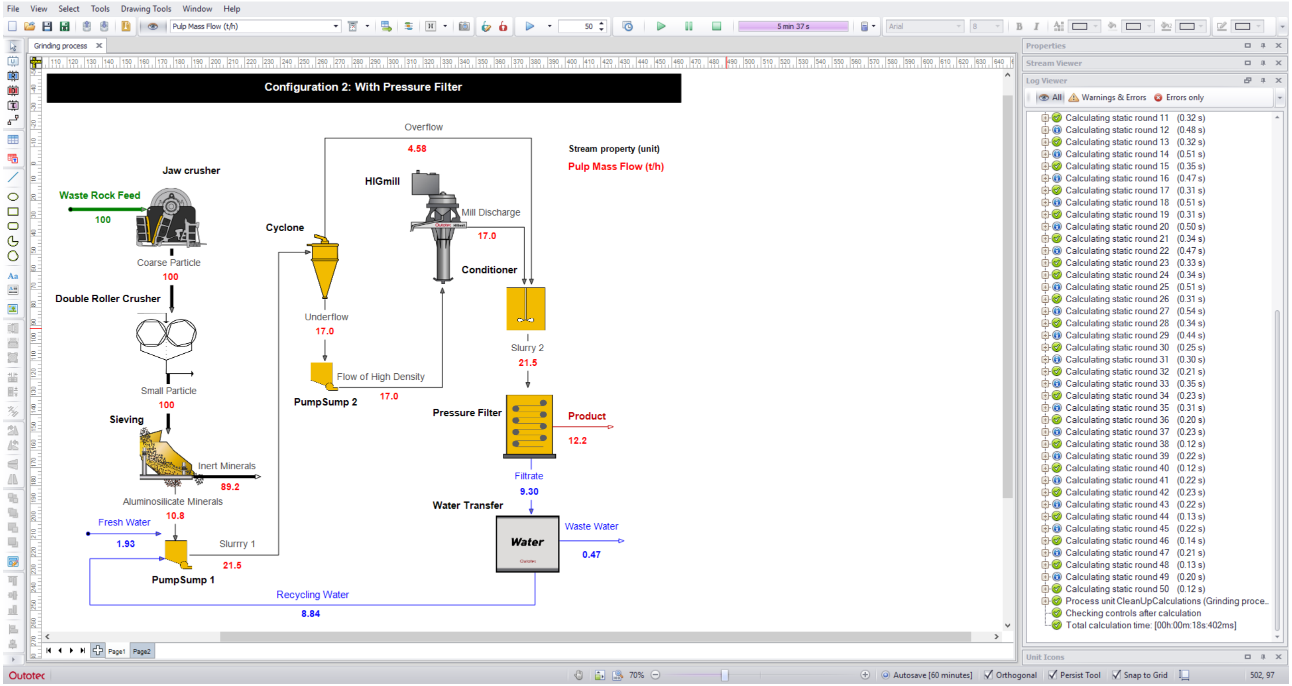

Figure S3. Configuration 2 of mechanochemical activation: Flowsheet simulation in HSC Sim (Route 2).

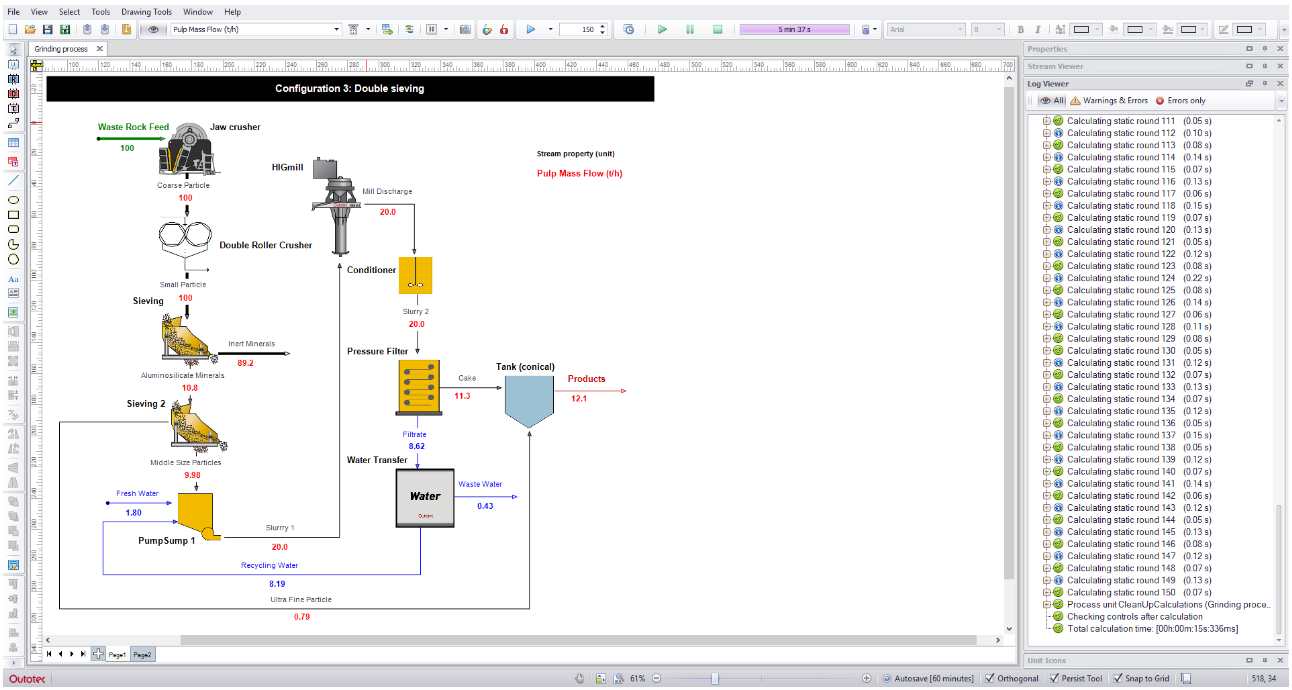

Figure S4. Configuration 3 of mechanochemical activation: Flowsheet simulation in HSC Sim (Route 3).

C. Further supporting graphs and tables for the manuscript

Figure S5 to Figure S8 show the Rietveld refinement of ground samples and samples after solubility test, indicating G16LiCl obtains relatively higher amorphous phase (R_{wp}< 10).

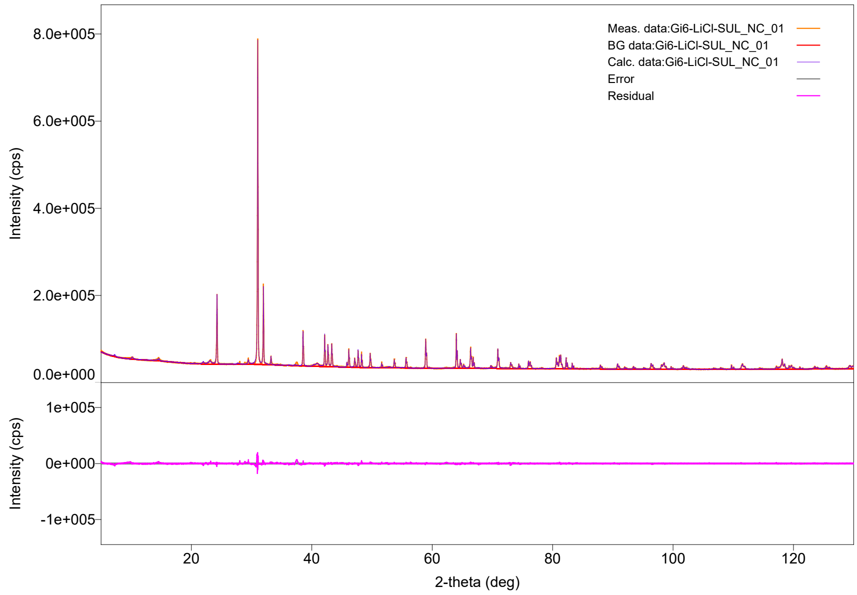

Figure S5. WPPF Rietveld refinement of G16LiCl with internal standard (amorphous phase content: 38.2%).

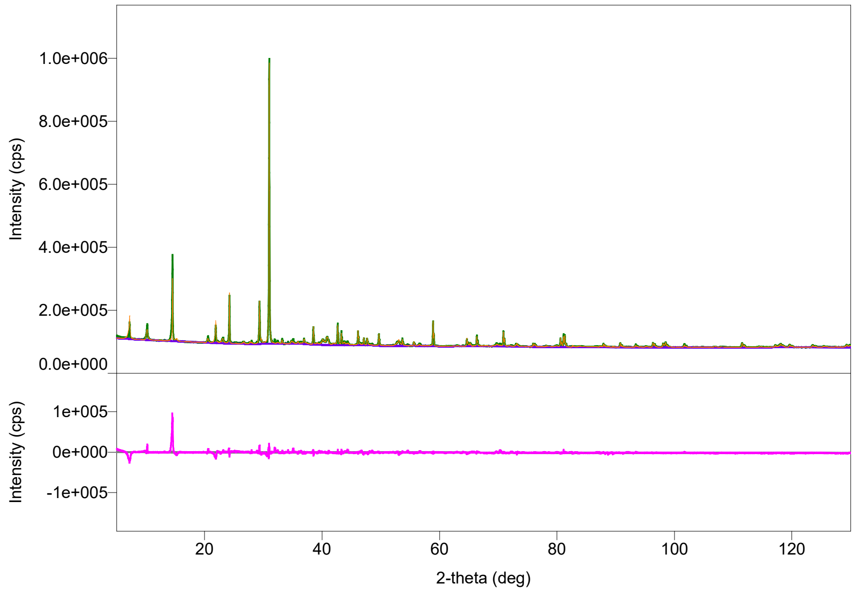

Figure S6. WPPF Rietveld refinement of G1 after solubility test (amorphous phase content: 26.8%).

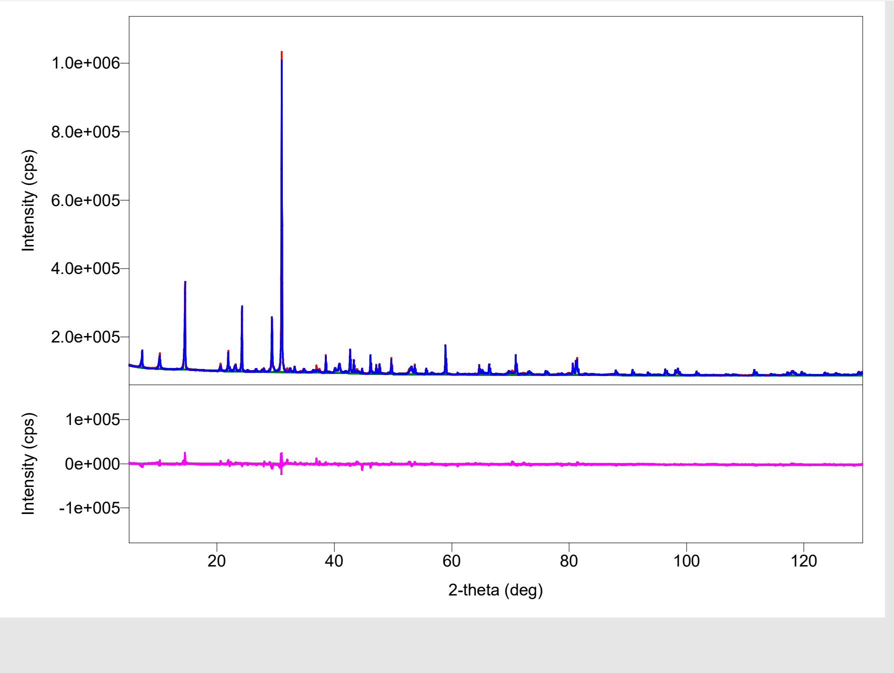

Figure S7. WPPF Rietveld refinement of G16 after solubility test (amorphous phase content: 27.2%).

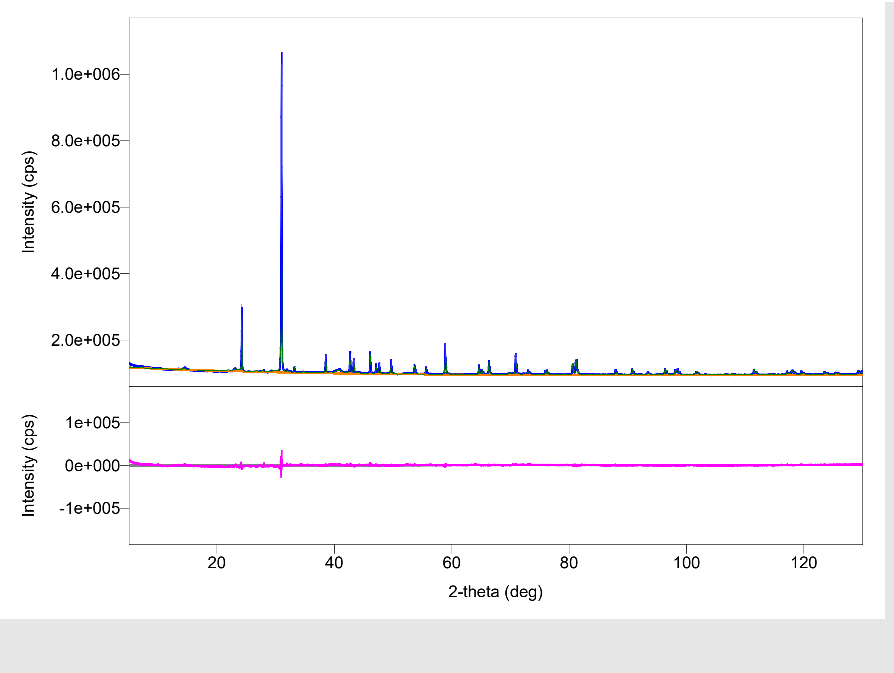

Figure S8. WPPF Rietveld refinement of G16LiCl after solubility test (amorphous phase content: 43.1%).

Figure S9 and Figure S10 display the STEM elemental mapping of G16 samples with and without lithium chloride grinding aid after solubility, where G16 retains crystallinity while G16LiCl shows a gel-like structure.

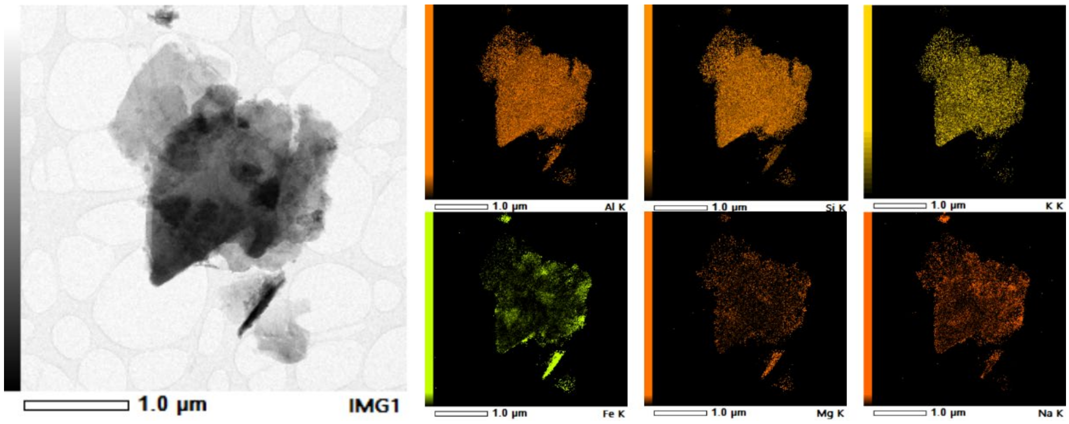

Figure S9. STEM elemental mapping of G16 after solubility test.

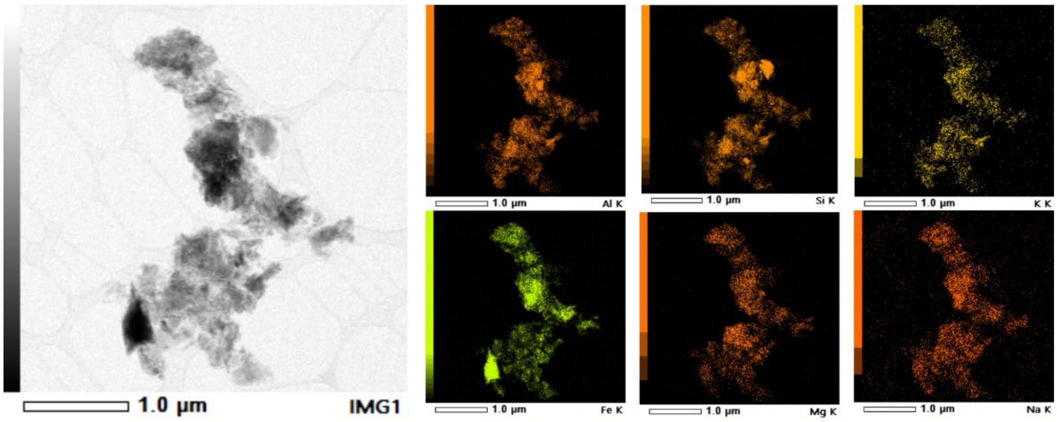

Figure S10. STEM elemental mapping of G16LiCl after solubility test.

D. Unit process data for foreground processes

All the background data used in this study are linked with the Ecoinvent 3.7 LCA Database. The modelled inventory for all systems is depicted in Table S4.

Table S4 Unit process data for foreground processes in this study

	Comparison of systems to produce binder materials				
	Lab scale	Route 1 with thickener	Route 2 with pressure filter	Route 3 additional double sieving	OPC cement
Precursor production	**Inputs**: Waste rock (zero burden approach) Market for tap water 1.17 kg, Europe Electricity medium voltage 3.35 kWh (direct measurements), Europe Lithium chloride 0.005 kg, GLO Output: 1 kg of precursor	**Inputs**: Waste rock (zero burden approach) Market for tap water 1.17 kg, Europe Electricity medium voltage 0.025 kWh, Europe Lithium chloride 0.005 kg, GLO Output: 1 kg of precursor	**Inputs**: Waste rock (zero burden approach) Market for tap water 0.16 kg, Europe Electricity medium voltage 0.021 kWh, Europe Lithium chloride 0.005 kg, GLO Output: 1 kg of precursor	**Inputs**: Waste rock (zero burden approach) Market for tap water 0.15 kg, Europe Electricity medium voltage 0.02 kWh, Europe Lithium chloride 0.005 kg, GLO Output: 1 kg of precursor	/

Binder making (recipe from own study, see Table 3)	**Inputs**: Precursor 0.6096 kg Sodium silicate 0.0722 kg, RER Sodium hydroxide 0.0145 kg, RER Tap water 0.303 kg, Europe **Output**: 1 kg of geopolymer binder	**Inputs**: Precursor 0.6096 kg Sodium silicate 0.0722 kg, RER Sodium hydroxide 0.0145 kg, RER Tap water 0.303 kg, Europe **Output**: 1 kg of geopolymer binder	**Inputs**: Precursor 0.6096 kg Sodium silicate 0.0722 kg, RER Sodium hydroxide 0.0145 kg, RER Tap water 0.303 kg, Europe **Output**: 1 kg of geopolymer binder	**Inputs**: Precursor 0.6096 kg Sodium silicate 0.0722 kg, RER Sodium hydroxide 0.0145 kg, RER Tap water 0.303 kg, Europe **Output**: 1 kg of geopolymer binder	**Output**: 1 kg Ordinary Portland Cement, Europe without Switzerland

Supplementary information Reference:

1. Åstholm M, Paz A, Jamieson E (2012) High intensity grinding at its finest

2. Lottermoser BG (2010) Sulfidic Mine Wastes. In: Mine Wastes: Characterization, Treatment and Environmental Impacts. Springer Berlin Heidelberg, Berlin, Heidelberg, pp 43–117