



FACULTY OF TECHNOLOGY

**UTILISING PHLOGOPITE AS A  
SUPPLEMENTARY CEMENTITIOUS MATERIAL  
(SCM)**

Chayapa Techavissavakit

PROSESSITEKNIIKAN KOULUTUSOHJELMA

Kandidaatintyö

Helmikuu 2026

## ABSTRACT

The production of ordinary Portland cement (OPC) is the reason for carbon dioxide emissions and high energy consumption, making it important to explore more environmentally friendly ways in concrete production. One effective approach to reducing the environmental impacts of cement is the partial replacement of OPC with supplementary cementitious materials (SCMs). This thesis investigates the potential of phlogopite, a magnesium-rich mica mineral obtained as a mining by-product from Siilinjärvi, Finland, as an SCM in Portland cement-based systems.

To improve its reactivity, phlogopite was examined through mechanical milling and chemical pre-treatment using sulfuric acid ( $H_2SO_4$ ) and 3,4-dihydroxybenzoic acid (DHZ), combined with sonication. Cement mortars were prepared with a 20 wt.% replacement of OPC by phlogopite, and compressive strength was measured after 3, 7, and 28 days of curing times.

The results indicate that the incorporation of raw, untreated phlogopite as a SCM hindered the strength development of the mortar specimens. In contrast, disk-milled phlogopite led to an enhancement in mechanical strength compared with the reference samples composed of 100% Portland cement. Additional chemical treatment of phlogopite did not result in further improvements in mechanical performance; however, the strength values remained higher than those observed for mortars containing raw phlogopite.

Overall, mechanically activated phlogopite demonstrates potential as a SCM. The utilization of this mining by-product could contribute to reducing clinker consumption and enhancing the sustainability of cement-based materials.

*Keywords: Mechanochemical Activation, Phlogopite, Portland Cement, Sonication, Supplementary Cementitious Materials.*

# TIIVISTELMÄ

Tavallisen portlandsementin (OPC) valmistus aiheuttaa merkittäviä hiilidioksidipäästöjä ja se kuluttaa paljon energiaa, minkä vuoksi ympäristöystävällisempien ratkaisujen tutkiminen betonintuotannossa on tärkeää. Yksi tehokas tapa, joka voi vähentää sementin ympäristövaikutuksia on sijoita osa OPC:stä seosaineilla eli SCM materiaalia. Tässä kandidityö tarkastellaan Siilinjärven kaivostoiminnan sivutuotteena saatavan magnesiumrikkaan kiilemineraalin, flogopiitin, soveltuvuutta seosaineeksi portlandsementtipohjaisissa järjestelmissä.

Reaktiivisuuden parantamiseksi flogopiittia käsiteltiin mekaanisella sekä kemiallisella esikäsitteily käyttäen rikkihappo ( $H_2SO_4$ ) ja 3,4-dyhydroksibentsoehappo (DHZ) yhdessä sonikoinnin kanssa. Sementtilaastit valmistettiin korvaamalla 20 painoprosenttia OPC:stä flogopiitilla, ja puristuslujuus mitattiin 3,7 ja 28 vuorokauden kovettumisajan jälkeen.

Tulokset osoittivat, että käsittelemätön flogopiittin käyttö seosaineena heikensi laastinäytteiden lujuuden kehittymistä. Sen sijaan mekaanisesti jauhettu flogopiitti paransi näytteiden mekaanista lujuutta verrattuna vertailunäytteisiin, joka sisältää 100% portlandsementtiä. Flogopiitin kemiallinen lisäkäsitteily ei parantanut mekaanista lujuutta enää lissä. Näytteiden lujuusarvot olivat kuitenkin korkeammat kuin laasteissa, joissa käytettiin käsittelemätöntä flogopiittia.

Yhteenvedona voidaan todeta, että mekaanisesti aktivoitulla flogopiitilla on potentiaalia seosaineeksi. Tämän kaivostoiminnan sivutuotteen hyödyntäminen voisi vähentää klinkkerin käyttöä ja parantaa sementtipohjaisten materiaalien kestävyyttä.

*Asiasanat: Flogopiiti, Mekanokemiallinen Aktivointi, Portlandsementti, Sonikointi, Seosaineet*

# TABLE OF CONTENTS

ABSTRACT .....	2
TIIVISTELMÄ.....	3
TABLE OF CONTENTS.....	4
SYMBOLS AND ABBREVIATIONS .....	5
1 INTRODUCTION.....	6
2 LITERATURE REVIEW .....	8
2.1 Ordinary Portland Cement in Concrete .....	8
2.2 Environmental impact of cement.....	9
2.3 Supplementary cementitious materials (SCMs) .....	10
2.4 Phlogopite as a Potential Supplementary Cementitious Material .....	11
2.5 Improving reactivity: milling, Acid treatment, and sonication.....	12
3 MATERIALS AND METHODS.....	13
3.1 Materials .....	13
3.2 Milling of phlogopite.....	14
3.3 Chemical Treatment and Sonication.....	14
3.4 Separation, Filtration, Drying, and Grinding.....	16
3.5 Mix design and Casting .....	18
3.6 Curing and testing.....	19
4 RESULTS .....	21
5 REWIEW OF RESULTS.....	22
6 CONCLUSION .....	24
7 REFERENCES .....	25

## SYMBOLS AND ABBREVIATIONS

The following table presents the symbols and abbreviations used throughout this thesis.

### Symbols

M molar concentration [mol/L]

rpm revolutions per minute [ $min^{-1}$ ]

wt.% Weight percentage [%]

### Abbreviations

BFS Blast furnace slag

C-S-H Calcium silicate hydrate

$CO_2$  Carbon dioxide

DHZ 3,4-Dihydroxybenzoic acid

DI Deionized water

$H_2SO_4$  Sulfuric acid

SCMs Supplementary cementitious materials

# 1 INTRODUCTION

The single most used modern construction material in today's world is Concrete. Concrete is a highly reliable material that can withstand harsh conditions while also being quite cheap to produce. The most pressing downside to the production of concrete is the usage of Cement. The process of producing cement is very taxing on the environment due to the calcination of limestone and its high energy demand. To produce one ton of Clinker, an approximately 90 litres of oil and 180kg of coal are required (Ali et al., 2015).

Global cement production has increased more than global fossil energy in the last 2 decades and around 25 billion tonnes per year. Concrete represents the largest portion of construction and demolition waste by weight in the United States, totalling over 220 million tonnes in 2014 (Nguyen et al., 2021). The emissions from the cement industry release to the atmosphere around 8% of global carbon dioxide emissions (Andrew, 2019; Scrivener, et al. 2018).

One common way to make concrete less harmful to the environment is to use supplemental cementitious materials (SCMs) to partially replace Portland cement. SCMs is one of the most efficient solutions to minimise carbon emissions and make concrete that lasts longer and is better for the environment. It can reduce cost and make better use of resources. By replacing the Portland cement with 15% of SCMs can cut  $CO_2$  emissions by 5.3%. This reduction can be even greater if the SCM content is increased to 20% (Kang, et al. 2025; Hossain, et al. 2021).

In addition of standard supplementary cementitious materials, there is increasing interest in using mining by-products as alternative SCMs. This is done to cut down on mining waste and the amount of clinker used in cement, which lowers  $CO_2$  emissions. Mining activities in the European Union can produce a large amount of waste, approximately 30% in 2012. Such a big amount of the waste can affect the environment and people's health. (European commission, 2016)

Many mining by-products contain aluminosilicate phases that can react within cementitious systems. When appropriately processed and incorporated, these materials can contribute to strength development and improved long-term performance of cement-

based composites (Juenger et al., 2011). Phlogopite is a mineral that is often found in large amounts as a by-product of mining activities (Puustinen, 1973). Phlogopite ( $KMg_3(AlSi_3O_{10})(OH)_2$ ) has a crystalline chemical structure that includes magnesium, silica and alumina. However, its use as a further cementitious material in Portland cement-based systems is still not well known (Niu et al., 2020). Increasing the reactivity of phlogopite through mechanical and chemical pre-treatment could improve its use in more sustainable cement-based materials.

This study investigates the potential of phlogopite as a supplementary cementitious material by replacing 20 wt.% of Portland cement. The effects of mechanical activation through milling and chemical treatment, including acid treatment with and without sonication, on elemental dissolution and compressive strength development were examined. All the samples were cured under controlled conditions, and their compressive strength was measured after 3, 7 and 28 days.

## 2 LITERATURE REVIEW

### 2.1 Ordinary Portland Cement in Concrete

Ordinary Portland Cement (OPC) is the most common binder used in construction, because it is cheap, strong, long-lasting, and flexible. To make OPC, you grind together clinker, which is made by heating limestone and clay to a high temperature, and a little bit of gypsum to control the setting time and expansion (Ali et al., 2015).

Making clinker uses the most energy in the cement-making process. It uses more than 90% of the energy used in the cement industry. Limestone, silica stone, coal ash, and iron sources are the main raw materials used to make clinker. These materials are heated to 1450 degrees Celsius to make the main clinker phases (Elakneswaran et al., 2019).

Ordinary Portland cement is a type of hydraulic binder, which mean it hardens when it comes into contact with water. The main minerals in it are tricalcium silicate ( $Ca_3SiO_5$ ), dicalcium silicate ( $Ca_2SiO_4$ ), tricalcium aluminate ( $Ca_3Al_2O_6$ ), and calcium aluminoferrite ( $Ca_4(AlFe)_4O_{10}$ ). Changes in the composition of clinker and the way it is made it possible to make cements with certain feature, such as quick setting, stronger early-age strength, and less heat generation during hydration phases (Neville, A. M. (2011)).

To make concrete, cement is mixed with aggregates and water. Water is one of these parts that is very important for building strength since it starts hydration reactions in the cement pastes. In order to make sure the cement hydrates properly and the fresh concrete is easy to work with; the amount of water must be carefully controlled. Calcium silicate hydrate (C-S-H) is the main hydration product that make concrete stronger (Neville, A. M. (2011); Elakneswaran et al., 2019).

## 2.2 Environmental impact of cement

The production of ordinary Portland cement is the main cause of carbon footprint, which difficult to deny it, particularly in terms of carbon dioxide emissions and energy consumption. Limestone utilized in cement manufacturing is causing global warming. Fossil fuels are utilised to produce heat for the process of breaking down limestone during cement manufactures, which causes emissions (Noor-ul-Amin, 2012; Ali et al., 2015).



When calcium carbonate ( $CaCO_3$ ) is heated, it breaks down into calcium oxide (CaO) and carbon dioxide ( $CO_2$ ). This chemical process is responsible for around 65% of all emissions connected to cement. This is because making clinker uses a lot of energy. Cement manufacturing facilities release out a lot of sulphur dioxide, nitrogen oxides, and carbon monoxide, which can have several negative effects on health and the environment (Mishra et al., 2022; Ali et al., 2015).

Nitrogen oxide ( $NO_x$ ) is harmful to the environment by causing acid rain and global warming. It may harm people with lung disorders like asthma and destroy lung tissue in people who work or exercise outside. Sulphur dioxide ( $SO_2$ ) can make it difficult to breathe and may cause heart problems in some people. Carbon monoxide (CO) can have severe effects on health by limiting the amount of oxygen that gets to the body's organs and tissues. It also effects the cardiovascular and central nervous systems (Ali et al., 2015).

Because of the energy use during (OPC) manufacture, each tonne of OPC releases between 0.82 and 1 metric tonnes of  $CO_2$ . Researchers are investigating new, eco-friendly materials for reducing  $CO_2$  emissions (Alsaman et al., 2021).

Blended cement has been suggested to decrease usage on OPC, consequently reducing the associated carbon footprint. Blended cement contains supplemental cementitious materials (SCMs) that left over from the mining process, could reduce  $CO_2$  emissions by about 13-22%. The solution for lowering the release of carbon dioxide depends on several

of important factors, such as the amount of SCMs mixed, the source of the raw materials, and the type of production facilities (Alsalman et al., 2021).

### **2.3 Supplementary cementitious materials (SCMs)**

Supplementary cementitious materials (SCMs) are non-clinker mineral components that are added to Portland cement blends to partially replace OPC, improve performance of concrete, and make it less harmful to the environment. SCMs can lowering the amount of clinker needed. Fly ash, ground granulated blast furnace slag (GGBFS), natural pozzolan, silica fume, calcined clay and ground glass are common SCMs used in concrete manufacturing. These materials usually react with calcium hydroxide released during cement hydration, producing extra calcium silicate hydrate (C-S-H) and improving the pore structure of the hardened paste. As a result, concrete containing SCMs often shows better long-term strength, lower permeability, and longer-lasting durability than regular OPC concrete (CEMEX, n.d.; O'Quinn et al., 2025).

The use of SCMs reduces the amount of Portland cement needed in concrete, which reducing carbon dioxide emissions, as the embodied emissions associated with clinker production are lowered. For example, commonly used SCMs such as fly ash and GGBFS can reduce CO<sub>2</sub> emissions by 20-30% when used in appropriate proportions. Traditional SCMs such as fly ash and blast furnace slag are actively utilised, however, their availability is based on industrial by-products, pushing the research of other possibilities (CEMEX, n.d.; Juenger et al., 2011).

Phlogopite has been mainly studied as a component in alkali-activated binder systems, although its application as a partial substitute for Portland cement remains limited. Phlogopite is an aluminosilicate mineral that contains important elements such as silicon (Si), aluminium (Al), and magnesium (Mg), which are relevant for cementitious reactions. Because of this chemical composition, phlogopite has the potential to participate in hydration reactions and contribute to the formation of binding phases in cement-based systems. Previous studies have also shown that phyllosilicate minerals such as phlogopite can be considered for use in SCMs. (Ponomar et al., 2022; Akbarzadeh Khoei et al., 2025).

## 2.4 Phlogopite as a Potential Supplementary Cementitious Material

Phlogopite is a magnesium-rich mica mineral that contains significant amounts of silica and alumina, which makes it a potential candidate for use in cementitious systems (Fig 1). Phlogopite belongs to the mica subgroup of phyllosilicates (sheet silicates) (Sreenivasan et al., 2017).

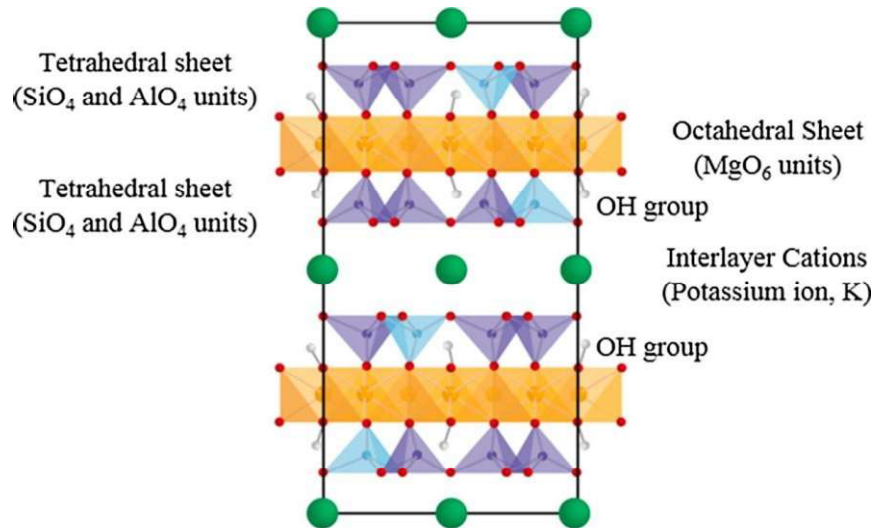


Figure 1. The crystal structure of perfect phlogopite. (Sreenivasan et al., 2017)

Characterised by a layered crystal structure. Its structural unit consists of one octahedral sheet sandwiched between two tetrahedral silicate sheets, forming a so-called T-O-T (tetrahedral-octahedral-tetrahedral) layer. These layers are separated by interlayer cations, that can be represented as  $KMg_3AlSi_3O_{10}(OH)_2$ . In Finland, phlogopite is found in mining at the Siilinjärvi deposit as a by-product (Sreenivasan et al., 2017).

Phlogopite, despite its advantageous chemical composition, in its natural crystalline form is expected to exhibit limited reactivity, as studies show that significant dissolution and strength development generally occur after mechanical or chemical activation (Niu et al., 2020; Ponomar et al., 2022).

This study investigates the potential of phlogopite as a supplementary cementitious material and how mechanical and chemical treatment procedures affect its performance. In particular, the study evaluates milled and chemically treated phlogopite as a 20%.wt partial replacement of Portland cement and investigates its strength development.

## 2.5 Improving reactivity: milling, Acid treatment, and sonication

Due to the limited intrinsic reactivity of crystalline phlogopite, modification of its physical and chemical properties may be required to enhance its suitability as a supplementary cementitious material. Previous studies on aluminosilicate minerals have demonstrated that mechanical and chemical activation methods can significantly influence mineral dissolution and reactivity in cementitious systems (Niu et al., 2020; Juenger et al., 2011).

High-energy milling can significantly change the structure of layered silicate minerals, making them more reactive. The process changes the structure and partial amorphization, which improves certain elements dissolve better and improves pozzolanic behaviour. These changes suggest that mechanochemical treatment is a useful pre-treatment step for activating otherwise low-reactivity minerals for use in cementitious systems (Baki et al., 2022). Similar approaches have been applied to phlogopite in previous research, where milling was used to modify its structure prior to further treatment (Aho, 2025).

In this study, chemical activation using sulfuric acid ( $H_2SO_4$ ) and 3,4-Dihydroxybenzoic acid to modify the phlogopite structure. Acid treatment is known to affect phyllosilicate minerals through proton exchange and layer attack, which can release cations and alter surface properties (Komadel, 2003).

Sonication introduces ultrasonic waves into a liquid medium, generating cavitation effects that enhances mass transfer and accelerate dissolution processes. The rapid formation and collapse of microbubbles can create localized high temperatures and pressures, which may facilitate mineral surface modification and increase reactivity (Suslick & Price, 1999).

### 3 MATERIALS AND METHODS

#### 3.1 Materials

Ordinary Portland cement (OPC) was used as the primary binder in all cement paste mixtures. Phlogopite ( $KMg_3(AlSi_3O_{10})(OH)_2$ ), sourced from the **LKAB, Siilijärvi, Finland** and obtained as a mining by-product, was replace Portland cement at a replacement level of 20 wt.%.

Standard sand was added to the paste mixtures to ensure consistency between samples and deionized (DI) water was used for all cement paste mixing and chemical treatment steps to minimize the influence of dissolved impurities.

A 0.1M solution of  $H_2SO_4$  was prepared from sulfuric acid (95% wt. Merck, Darmstadt, Germany) and a 0.1M solution of DHZ was prepared using 3,4-Dihydroxybenzoic acid (97% wt. Merck, Darmstadt, Germany) were used for the chemical pre-treatment of phlogopite. These treatments were applied to enhance mineral dissolution and modify the surface reactivity of phlogopite prior to its incorporation into cement paste.

A polycarboxylate-based superplasticizer (Fig. 2, Hyperflow 3000, Rhein-Chemotechnik GmbH, Germany) was used to improve the workability of the cement paste mixtures. All materials were stored at room temperature and handled under laboratory conditions.



Figure 2. A polycarboxylate-based superplasticizer (Hyperflow 3000, Rhein-Chemotechnik GmbH, Germany)

### 3.2 Milling of phlogopite

Phlogopite was mechanically activated by milling using a **Retsch Vibrating Disk Mill (RS200)**. Milling was carried out at a rotational speed of **1400 rpm for 10 minutes** (Fig. 3). The purpose of milling step was to reduce particle size, increase surface area, and enhance the reactivity of phlogopite prior to chemical treatment and cement mixing.

After milling, the phlogopite powder was collected and stored in sealed containers at room temperature until further processing.



Figure 3. The disk mill RS200 used for milling phlogopite.

### 3.3 Chemical Treatment and Sonication

Chemical treatment was applied to the milled phlogopite in order to enhance mineral dissolution and modify surface reactivity prior to its incorporation into cement paste. Two different chemical treatments were investigated: sulfuric acid ( $H_2SO_4$ ) treatment and sulfuric acid combined with DHZ.

A 0.1 M sulfuric acid ( $H_2SO_4$ ) solution was prepared for the chemical treatment. In the first treatment, 22 g of milled phlogopite was mixed with 100 mL of deionized (DI) water and 60 mL of 0.1 M sulfuric acid ( $H_2SO_4$ ) in a 250 mL beaker. In the second treatment,

22 g of milled phlogopite was mixed with 100 mL of deionized (DI) water, 0.25 g of DHZ, and 60 mL of 0.1 M sulfuric acid ( $H_2SO_4$ ) in a 250 mL beaker.

The initial pH of both samples after mixing was approximately 3. An immediate color change was observed upon contact between the acid solution and the milled phlogopite, indicating rapid surface reactions. (Fig. 4)



Figure 4. A sample after mixing milled phlogopite with acids ( $H_2SO_4$  & DHZ).

For the leaching step, the mixtures were sonicated for 30 minutes at 80% amplitude. The beaker was placed into the sonicator (Hielscher UP 400s (Germany) ultrasonicator) and cold water was added to the box around the beaker so that the mixture does not overheat (Fig.5).

Following sonication, further colour changes of the sample was observed and recorded. The pH of the samples were measured again after sonication and was found to be approximately 7.



Figure 5. The picture on the left was the leaching process of the phlogopite sample using 0.1M of sulfuric acid ( $H_2SO_4$ ) and picture on the right was the leaching process of the phlogopite sample using 0.1M of DHZ and 0.1M of sulfuric acid ( $H_2SO_4$ ).

### **3.4 Separation, Filtration, Drying, and Grinding**

After completion of the chemical treatment and sonication, the samples were processed to separate the solid and liquid phases. Initial separation was carried out using a centrifuge, which enabled efficient settling of solid particles. The samples were transferred into centrifuge containers and centrifuged for 5 minutes at 5000 rpm. (Fig. 6)



Figure 6. Centrifuge machine

The remaining solid residues were collected and dried under laboratory conditions to remove residual moisture. After drying, the solid materials were manually ground using a mortar and pestle to obtain a homogenous powder prior to weighing and cement mixing. (Fig. 7)



Figure 7. Grinding sonicated samples after drying.

The ground phlogopite samples were then weighed prior to using the cement paste mixtures to ensure accurate and consistent mix proportions.

The prepared solid samples were stored in sealed containers at room temperature until further use.

### 3.5 Mix design and Casting

The experimental design was developed to evaluate the feasibility of using phlogopite as a supplementary cementitious material (SCM) by partially replacing Portland cement at a replacement level of 20 wt.%. A reference mixture without phlogopite was also prepared for comparison (Table 1).

Table 1. Mix design of samples.

Sample	Portland cement (g)	Deionized water (mL)	Phlogopite (g)	Standard Sand (g)
Ref-Cement	100 g	50 mL	-	300 g
Raw-Phlog-Cement	80 g	50 mL	20 g	300 g
Mill-Phlog-Cement	80 g	50 mL	20 g	300 g
$H_2SO_4$ -Cement	80 g	50 mL	20 g	300 g
DHZ-Cement	80 g	50 mL	20 g	300 g

The dry components (cement and phlogopite) were first blended to ensure homogenous distribution of the materials. Deionized water and sand were then gradually added while mixing continued until a uniform paste was obtained. During the process, the superplasticizer was added in this stage to improve flowability and ensure consistent mixing.

After mixing, the fresh cement mortar was poured into  $2 \times 2 \times 8$  cm moulds and compacted using a **jolting table** to remove entrapped air and improve sample uniformity. Excess material was removed from the mould surface to ensure even specimen dimensions. The moulded samples were then prepared for curing under controlled laboratory conditions. (Fig. 8)

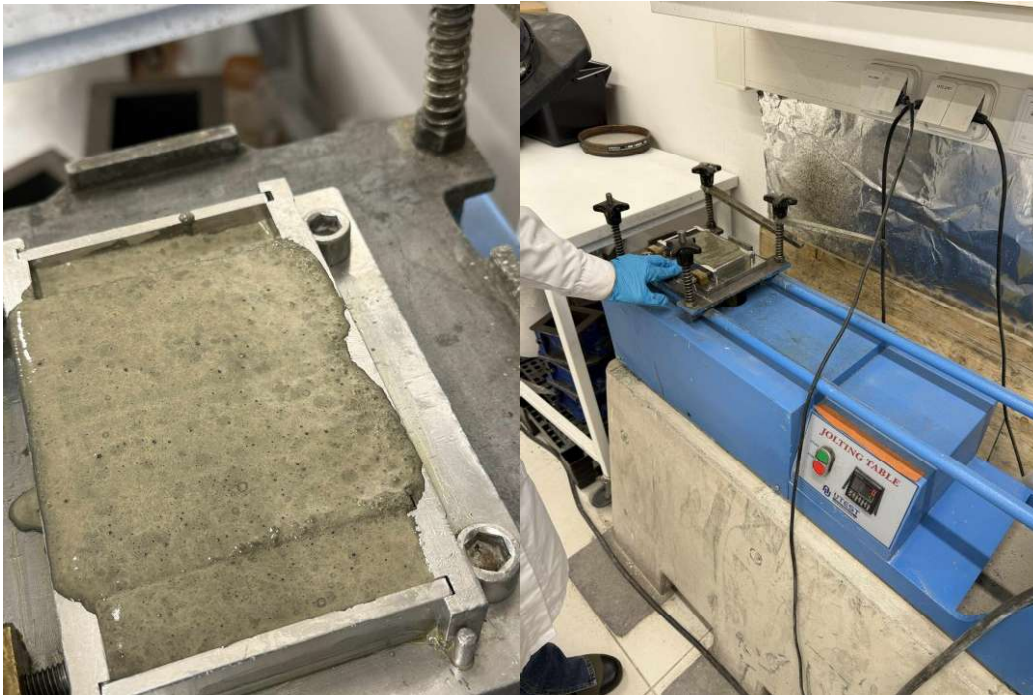


Figure 8. Mould samples using  $2 \times 2 \times 8$  cm and Jolting table (UTCM-0890)

### 3.6 Curing and testing

After casting, all samples were stored under controlled laboratory conditions and allowed to cure at room temperature. The samples were kept undisturbed in their moulds during the initial curing period to ensure consistent hydration and strength development.

Compressive strength tests were performed after curing periods of **3,7 and 28 days**. Prior to testing, the samples were removed from the moulds. The compressive strength of each sample was determined by recording the maximum load at failure using **a compression testing machine**, in accordance with laboratory testing procedures (Fig. 9).

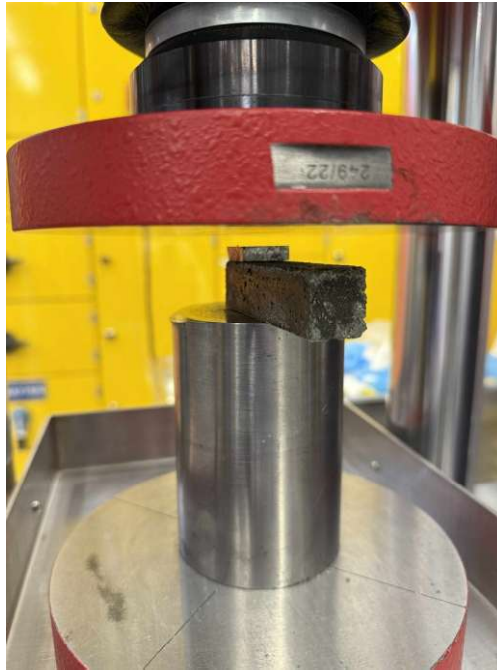


Figure 9. Compression testing machine (Toni technik Hydraulic Test Frame, Berlin, Germany)

## 4 RESULTS



Figure 10. All samples after curing arranged from left to right: Ref-Cement, Raw-Phlog-Cement, Mill-Phlog-Cement,  $H_2SO_4$ -Cement and DHZ-Cement.

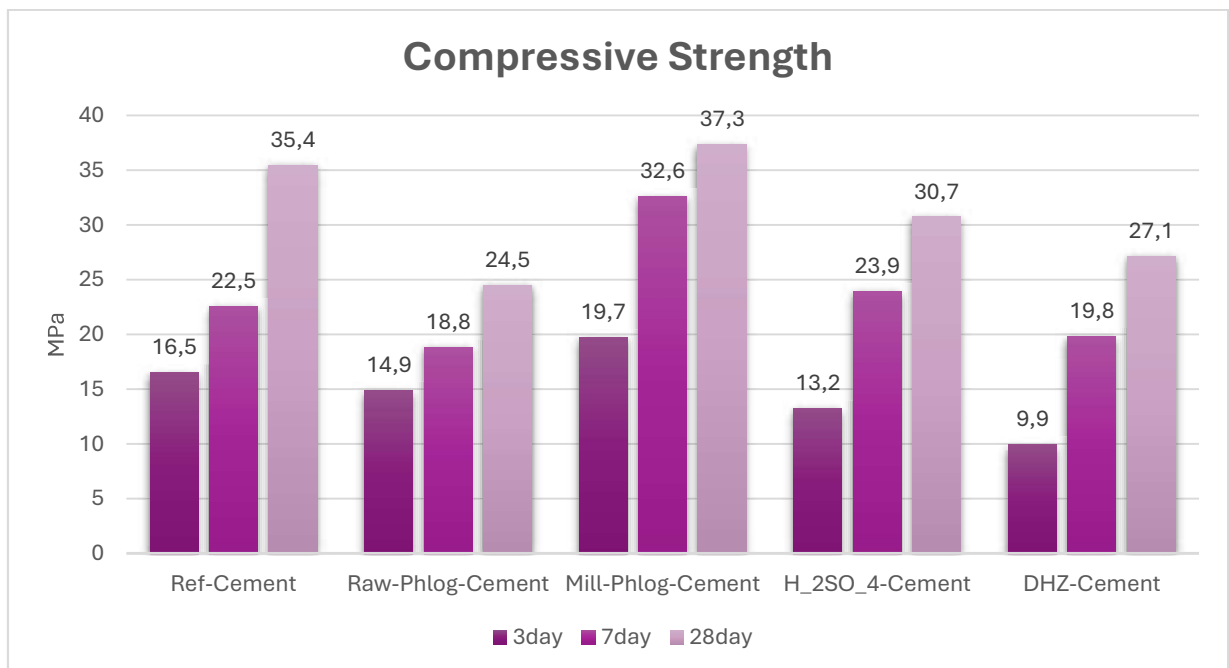


Figure 11. Compressive strength results of five samples, shown from left to right (Ref-Cement, Raw-Phlog-Cement, Milled-Phlog-Cement,  $H_2SO_4$ -Cement and DHZ-Cement) after 3,7 and 28 days of curing.

## 5 REVIEW OF RESULTS

The compressive strength results of the mortar samples are presented in Figure 11. The study focused on the effects of mechanical activation (milling), and chemical treatment (sulfuric acid ( $H_2SO_4$ ) treatment and sulfuric acid combined with DHZ) with and without sonication on the strength development of cement mortars containing phlogopite.

The Ref-Cement sample, which contained only Portland cement, was used as a baseline for comparison. As expected, the Ref-Cement sample showed steady strength development over time during the curing periods of 3,7 and 28 days.

Raw-Phlog-Cement sample was used as a 20 wt.% replacement of Portland cement, the compressive strength values were lower than those of the Ref-Cement at all curing times. This indicates that untreated phlogopite has limited reactivity in cement-based systems and therefore hinders the strength development of the mortar.

In contrast, the sample Mill-Phlog-Cement showed significantly higher compressive strength compared with the Raw-Phlog-Cement sample. Mechanical activation through milling improves the reactivity of phlogopite by reducing particle size and increasing surface area. As a result, more reactive aluminosilicate species can dissolve during hydration reactions, which contribute to the formation of stronger cementitious phases. After 28 days of curing, the Mill-Phlog-Cement sample even showed higher compressive strength than the Ref-Cement sample.

The chemically treated samples ( $H_2SO_4$ -Cement and DHZ-Cement) showed intermediate strength values. Although the acid treatments appeared to modify the material and improve its reactivity compared with Raw-Phlog-Cement sample, the strength development was still lower than that observed for the Mill-Phlog-Cement sample. This suggests that mechanical activation through milling had a stronger effect on improving the performance of phlogopite in cement mortars than the applied chemical treatments.

Overall, the results indicate that mechanical activation is important in improving the performance of phlogopite as a supplementary cementitious material. Milling significantly enhanced the compressive strength of mortar samples, while additional

chemical treatments did not provide further improvements compared with milling phlogopite alone.

## 6 CONCLUSION

Concrete is still one of the most important construction materials in the world. However, its production affects the environment, which means that more environmentally friendly options need to be explored. One possible solution is replacing the Ordinary Portland cement (OPC) with supplementary cementitious materials (SCM) derived from mining waste or industrial by-products.

This study investigated the potential of phlogopite as a supplemental cementitious material through mechanical and chemical activation. The results showed that untreated phlogopite had limited reactivity and reduced the strength development of the mortar samples. However, mechanically milled phlogopite showed better result improvement in compressive strength, outperforming the reference cement, raw phlogopite cement and chemically treated cement after 28 days of curing.

Mechanical activation through milling improved the performance of phlogopite by increasing its surface area and enhancing its reactivity. In comparison, chemical treatments using sulfuric acid and DHZ slightly improved the strength compared with raw phlogopite, but the results were still lower than those obtained with milling alone.

Overall, the results suggest that mechanical activation is an effective method for improving the performance of phlogopites as a supplementary cementitious material. The utilisation of phlogopite mining waste could reduce the use of Portland cement and contribute to more environmentally sustainable cement-based materials.

## 7 REFERENCES

Aho, E. (2025, January 9). Utilization of phlogopite as alkali activated material [Kandidaatintyö]. E. Aho. Laturi.Oulu.Fi. <https://oulurepo.oulu.fi/handle/10024/53617>

Akbarzadeh Khoei, M., Kurtulus, R., Alzeer, M. I. M., Sirviö, J. A., & Yliniemi, J. (2025). Combined Role of Organic Ligands and Ultrasound on the Dissolution of Phlogopite at pH 4 and 7. *Langmuir*, 41(23), 14607–14617. <https://doi.org/10.1021/acs.langmuir.4c04307>

Ali, N., Anwer, M., Alwi, S. K. K., & Anjum, M. N. (2015). *The Greenhouse Gas Emissions Produced by Cement Production and Its Impact on Environment: A Review of Global Cement Processing*. 2(2).

Alsaman, A., Assi, L. N., Kareem, R. S., Carter, K., & Ziehl, P. (2021). Energy and CO<sub>2</sub> emission assessments of alkali-activated concrete and Ordinary Portland Cement concrete: A comparative analysis of different grades of concrete. *Cleaner Environmental Systems*, 3, 100047. <https://doi.org/10.1016/j.cesys.2021.100047>

Andrew, R. M. (2019). Global CO<sub>2</sub> emissions from cement production, 1928–2018. *Earth System Science Data*, 11, 1675–1710. <https://doi.org/10.5194/essd-11-1675-2019>

Baki, V. A., Ke, X., Heath, A., Calabria-Holley, J., Terzi, C., & Sirin, M. (2022). The impact of mechanochemical activation on the physicochemical properties and pozzolanic reactivity of kaolinite, muscovite and montmorillonite. *Cement and Concrete Research*, 162, 106962. <https://doi.org/10.1016/j.cemconres.2022.106962>

Elakneswaran, Y., Noguchi, N., Matumoto, K., Morinaga, Y., Chabayashi, T., Kato, H., & Nawa, T. (2019). Characteristics of Ferrite-Rich Portland Cement: Comparison With Ordinary Portland Cement. *Frontiers in Materials*, 6. <https://doi.org/10.3389/fmats.2019.00097>

Hossain, Md. U., Dong, Y., & Ng, S. T. (2021). Influence of supplementary cementitious materials in sustainability performance of concrete industry: A case study

in Hong Kong. *Case Studies in Construction Materials*, 15, e00659.

<https://doi.org/10.1016/j.cscm.2021.e00659>

Juenger, M. C. G., Winnefeld, F., Provis, J. L., & Ideker, J. H. (2011). Advances in alternative cementitious binders. *Cement and Concrete Research, Conferences Special: Cement Hydration Kinetics and Modeling, Quebec City, 2009 & CONMOD10, Lausanne, 2010*, 41(12), 1232–1243. <https://doi.org/10.1016/j.cemconres.2010.11.012>

Kang, I., Kim, G., An, T., Lee, J., Shin, S., & Kim, J. (2025). Carbon neutrality in the Korean cement industry: Reducing carbon emissions by increasing supplementary cementitious material content in ordinary Portland cement. *Construction and Building Materials*, 494, 143391.

Komadel, P. (2003). Chemically modified smectites. *Clay Minerals*, 38(1), 127–138. <https://doi.org/10.1180/0009855033810083>

Mishra, U., Sarsaiya, S., & Gupta, A. (2022). A systematic review on the impact of cement industries on the natural environment. *Environmental Science and Pollution Research*, 29, 1–12. <https://doi.org/10.1007/s11356-022-18672-7>

Neville, A. M. (2011). *Properties of concrete*. Harlow Pearson Education.

Nguyen, W., Martinez, D. M., Jen, G., Duncan, J. F., & Ostertag, C. P. (2021). Interaction between global warming potential, durability, and structural properties of fiber-reinforced concrete with high waste materials inclusion. *Resources, Conservation and Recycling*, 169, 105453. <https://doi.org/10.1016/j.resconrec.2021.105453>

Niu, H., Kinnunen, P., Sreenivasan, H., Adesanya, E., & Illikainen, M. (2020). Structural collapse in phlogopite mica-rich mine tailings induced by mechanochemical treatment and implications to alkali activation potential. *Minerals Engineering*, 151, 106331. <https://doi.org/10.1016/j.mineng.2020.106331>

O’Quinn, K., Bernal, S. A., & Juenger, M. C. G. (2025). Blending portland limestone cement with supplementary cementitious materials to minimize clinker content.

Construction and Building Materials, 505, 144657.  
<https://doi.org/10.1016/j.conbuildmat.2025.144657>

Ponomar, V., Yliniemi, J., Adesanya, E., Ohenoja, K., & Illikainen, M. (2022). An overview of the utilisation of Fe-rich residues in alkali-activated binders: Mechanical properties and state of iron. *Journal of Cleaner Production*, 330, 129900.  
<https://doi.org/10.1016/j.jclepro.2021.129900>

Puustinen, K. (1973). Tetraferriphlogopite from the Siilinjärvi carbonatite complex, Finland. *Bulletin of the Geological Society of Finland*, 45(1), 35–42.  
<https://doi.org/10.17741/bgsf/45.1.005>

REPORT FROM THE COMMISSION TO THE EUROPEAN PARLIAMENT, THE COUNCIL, THE EUROPEAN ECONOMIC AND SOCIAL COMMITTEE AND THE COMMITTEE OF THE REGIONS on the Implementation of Directive 2006/21/EC on the Management of Waste from Extractive Industries and Amending Directive 2004/35/EC (2016). <https://eur-lex.europa.eu/legal-content/en/TXT/?uri=CELEX%3A52016DC0553>

Scrivener, K., Martirena, F., Bishnoi, S., & Maity, S. (2018). Calcined clay limestone cements (LC3). *Cement and Concrete Research*, 114, 49–56.  
<https://doi.org/10.1016/j.cemconres.2017.08.017>

Sreenivasan, H., Kinnunen, P., Heikkinen, E.-P., & Illikainen, M. (2017). Thermally treated phlogopite as magnesium-rich precursor for alkali activation purpose. *Minerals Engineering*, 113, 47–54. <https://doi.org/10.1016/j.mineng.2017.08.003>

Suslick, K. S., & Price, G. J. (1999). APPLICATIONS OF ULTRASOUND TO MATERIALS CHEMISTRY. *Annual Review of Materials Research*, 29(Volume 29, 1999), 295–326. <https://doi.org/10.1146/annurev.matsci.29.1.295>

What are Supplementary Cementitious Materials (SCMs)? (2023). Cemex US.  
<https://www.cemexusa.com/w/what-are-supplementary-cementitious-materials-scms>