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Author(s): Honkanen, Hannariina; Pennanen, Teemu; Turunen, Leena

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TESTING OF APPLICABILITY OF PULP PRODUCTION WASTE TO CONCRETE AND CONCRETE-LIKE MATERIALS

Hannariina Honkanen, principal lecturer, School of Technology, JAMK University of Applied Sciences, PO BOX 207, FI-40101 Jyväskylä, Finland

Email: hannariina.honkanen@jamk.fi ; Phone: +358 50 432 5820

Teemu Pennanen, laboratory engineer, School of Technology, JAMK University of Applied Sciences; Email: teemu.pennanen@jamk.fi

Leena Turunen, laboratory engineer, School of Technology, JAMK University of Applied Sciences; Email: leena.turunen@jamk.fi

ABSTRACT: Forest industry produces solid waste materials, which are still experienced globally problematic from the continuing use perspective. JAMK coordinated industry cooperated research project focuses on utilization of forest industry waste to increase the environmental friendliness of concrete used in construction. In addition, the common benefit is promoting cooperative initiatives between industry sectors. Project introduces manufacturing of test pieces of concrete mixed in various ways, including cement, water, aggregate and green liquor dregs (GLD) as raw materials. This paper focuses on the project parts containing selected groups of prism tests determining flexural and compression strength, GLD suitability for concrete constituent and test pieces of geopolymers and freeze-thaw durability tests. The usability tests were conducted for the material mixes in JAMK laboratories to examine three Finnish pulp factories' GLD as part of the concrete raw material mix. Receipts included also compilation of tests of shares of combined GLD and blast furnace slag. Results show, that small amounts of cement can be replaced with GLD without significant changes in strength. One essential indication from the results was the fact, that concrete-like masses are possible to be made totally without cement.

Keywords: concrete, green liquor dregs, waste, geopolymer

1 APPROACH AND SCIENTIFIC RELEVANCE

1.1 Background for utilization of by-products in concrete

Concrete is the most used manmade material in world. Concrete is also second most consumed substance in the world after water. History of concrete starts probably at least 4000 BC. After the Roman Empire collapsed, use of concrete became rare. The technology was developed again in the mid-18th century. A method of producing Portland cement was developed in England in 1824.

Portland cement itself is a complex material, manufactured by first burning an intimate mixture of limestone and clay or shale in a kiln at temperatures in the range of 1400°C–1500°C, and then intergrinding the resulting clinker with a small amount of gypsum. Concrete is a composite material. It is typically made of aggregates, water and cement. In addition, modern concretes usually contain one or more chemical admixtures to modify the properties of either the fresh or the hardened concrete, or both. By-products from coal power plants and steel industry like fly ash, blast furnace slag and silica fume are commonly used in concrete or in cement.

Material Economics [1] states, that "the EU uses currently more than two tonnes of concrete per person per year, of which 325 kg is cement". World cement production in 2017 was 4.1 billion tons. China used almost 60% of all the cement made in 2017. The production of cement is expected to rise to 5.5 billion tons by 2050. [2]

The cement industry is creating up to 8% of CO₂ worldwide [3]. This however varies from country to country. In EU "For every kilogram of cement that is produced, 0.7 kg of CO₂ is released into the air" [1]. For example Illikainen et al. [4] have tested by-products like slag and pretreated paper sludge for substituting the ordinary Portland cement in more sustainable construction materials. In addition, Rui et al. [5] have presented results of dregs used for the first time as filler in geopolymeric mortars production. The target for using by-products promotes circular economy and offers possibilities to cut down environmental impacts and greenhouse gas

emissions.

However, it is difficult to replace cement as a binder in concrete. The availability of alternative materials that can be used as other constituents varies considerably. For example, granulated blastfurnace slag availability depends on the location and output of blastfurnaces for pig-iron production equipped with slag granulation facilities, whilst fly ash use is dependent on supply from sufficiently close coal-fired power plants. The availability of pozzolans depends on the local situation and only a limited number of regions have access to this material for cement production.

Green liquor dregs (GLD) is a side product of pulp industry. In Finland over 50 000 tons (as a dry matter) green liquor dregs is placed to landfills in year and it is greatest waste fraction in pulp and paper industry [6]. Some part of generated GLD is utilized as infrastructure construction materials. GLD has already been used in cement production in commercial industrial products in Austria. According to Kinnarinen et al. [7], GLD offers great possibilities for industrial utilization due to its good availability. It has strong alkalinity and contains quite low levels of hazardous substances in total. Utilization is still subjected to environmental license in Finland. [8]

1.2 Industrial cooperation project KBB

JAMK University of Applied Sciences coordinates ongoing national research and development project "Sustainable bioresidual concrete" (2018-2020). The project drives on the interests of essential group of industrial players in sectors of forest and concrete industry acting in Finland. The aim of the project is to improve the environmental friendliness of concrete used in construction by utilizing pulp industry waste materials, which are still experienced problematic from the continuing use perspective. In addition to enhancing the utilization of waste materials from local origin, the common benefit is promoting cooperative initiatives between industry sectors. As concrete objective, project aims at finding out the new material mixtures' long-term

durability and strength and porosity properties meeting the goals set for construction. Project introduces manufacturing of test pieces of concrete mixed in various ways, including cement, water, aggregate and green liquor dregs (GLD). Concrete samples and their required strength and frost-resistance properties are determined in JAMK's accredited Concrete Testing Laboratory.

This study presents results from using three different GLD sample batches, originated from three different Scandinavian kraft pulp mills. Every GLD sample batch separates each other with particle size, chemistry and dry matter content. Nevertheless, every GLD batch were prepared in the laboratory in same way: drying, milling and screening before mixing with water and aggregates.

2 METHODS

2.1 Tests of using GLD in cement and concrete

Firstly, the conducted tests aimed at understanding the suitability of GLD to replace cement in concrete. Groundwork phase of the research was to carry out prism tests according to cement testing standard EN 196-1, casting 40x40x160 mm pieces and conducting flexural and compression strength tests (Figure 1). Recipe complied with the standard, and standard sand was used as aggregate. GLD's fineness was determined using Blaine method (according to EN 196-6). By using standard sand in the mass, one variable was eliminated simplifying the recipe and excluding some factors for uncertainties.

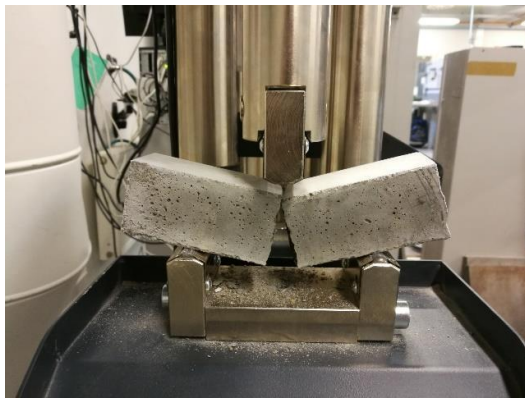


Figure 1: Example of prism test in JAMK laboratory.

Pretreatment of fresh green liquor dregs were carried out before the actual experiments. After GLD was received from the pulp mills the first step was to determine the moisture content of the material and dry the whole batch for further processing (drying temperature was 105 °C). The moisture content usually ranged from about 40 to 50 percent. After the drying procedure GLD was ground in a cutting mill with a sieve size of 0.25 mm and finally material was sieved with a vibrating screen with a sieve size of 0.063 mm (Figure 2).



Figure 2: Grinded and unprocessed green liquor dregs.

The second step of the research was to examine the waste materials' usability in concrete. The objective was either to replace cement or fine aggregate (filler) with GLD. The test resulted casting 100 mm cube-shaped test pieces with selected recipes, and testing compression strength according to EN 12390-3 and density according to EN12390-7 standards (Figure 3). Comparison was carried out to test pieces casted with and without waste-derived raw materials. All applications followed the same base recipe. GLD replaced either parts of cement or the share of aggregate.

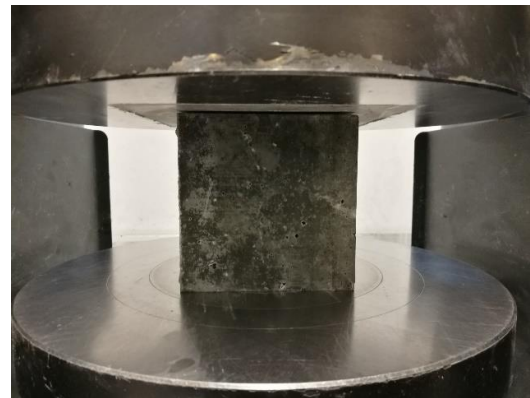


Figure 3: Example test cube in compressive strength test.

2.2 Geopolymers

The project was also interested to look into geopolymers, which are concrete-like products excluding cement as raw material. Geopolymers can be produced with utilizing industrial by-products, which offers incentives from the circularity and sustainability aspect in processes. Geopolymerization including setting and curing is physically based on exposing suitable material mixture to alkali activity and heat. [9]

Some experimental test pieces were manufactured for demonstration in the project. The approach for the recipes totally differed from the cement recipes with by ways of experience and creativity. Besides GLD, also blast furnace slag was used in the mass mixtures. Slag is already common recovered material especially in earth construction in Finland. From the material mix, cube-shaped test pieces were casted for determining values of compression strength.

Both concrete and geopolymers were also tested for

freeze-thaw durability as long-term test according to standard EN 12390-9. Freezing tests are essential when considering applications in northern demanding climate. The test included exposing the test pieces freeze-thaw cycles with 3% NaCl solution stress.

3 RESULTS

3.1 Prism strength tests

Prism tests included both compressive and flexural strength tests. Figure 4 shows the results of compressive strength test for different test pieces, where cement is partially replaced with green liquor dregs. 5 % share of GLD quite good results, but already with 10 % share strength shows clear decline.

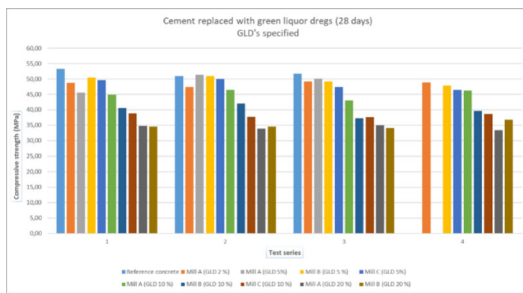


Figure 4: Comparison of recipes, with three factories' (mill A, mill B and mill C) GLD shares replacing cement, compressive strengths.

3.2 Cube compressive strength tests

Cube tests were conducted with method for concrete testing. Figure 5 presents the reference concrete compressive strength values at different test ages.

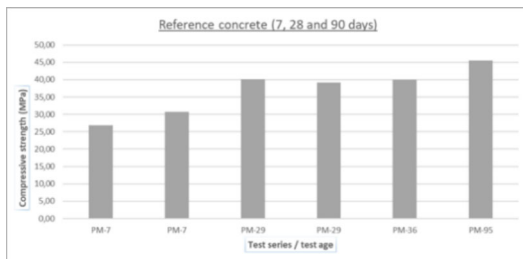


Figure 5: Reference concrete compressive strength results with different test ages.

Figure 6 shows the results when GLD is used for replacing fine aggregate. It is seen, that the use of GLD does affect from few to ten percents difference on compressive strength – even when tested with different testing age. When test age is 7 days, strength seems to be higher than for reference concrete. However, there are differences between different factory originated dregs.

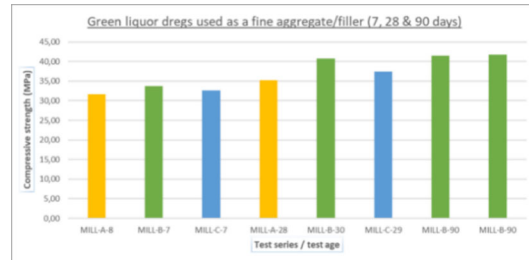


Figure 6: Comparison of recipes, with three factories' GLD shares, compressive strengths. Different compression test ages are seen in the figure.

The findings pointed out, that the strength is in relation to the share of GLD in the mass by cutting down the strength when GLD share is increased. However, with low share of GLD, 5 %, the difference of influence for strength change is insignificant when compared to regular mass.

The replacement of filler material in concrete by GLD was successful at least with smaller shares added in the mass. Workability properties did not show significant variations, and strength feature did not suffer significant drop. Furthermore, workability can be adjusted with using additives such as super plastisizers.

3.3 Geopolymer tests

In geopolymer tests, the observations included, that the proportions of different raw materials in the mix have great significance. In addition, water amount variation does not regulate properties in geopolymer as in concrete manufacturing. One important finding was that the blast furnace slag and GLD seem to work together well in geopolymer (Figure 7). The hardening process is however rather slow. The project is looking now into different kind of solutions to make the hardening process faster. The typical accelerators used in concrete doesn't seem to work with geopolymer masses. One of the most important findings in this project is the possibility to use GLD as an alkali activator. The development of this kind of one-part geopolymer mixes ("just add water") alkali-activated materials is an important step forward in the commercialization of these alternative, low carbon binders. [10]

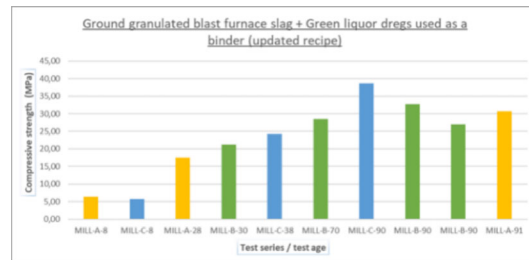


Figure 7: Comparison of three factories' GLD mixed with blast furnace slag as a binder, compressive strengths. Different compression test ages are seen in the figure.

In addition, the hybrid geopolymer mixes with OPC (ordinary portland cement) were tested in the project (Figure 8). When 20% of the binder was cement and the rest 60% of GGBF and 20% GLD the results showed

rather good results. They can be compared to the samples made only with OPC and GGBF in the age of 7 days.

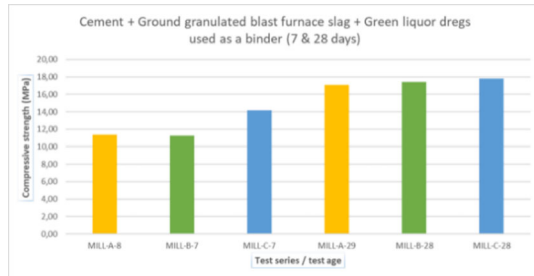


Figure 8: Comparison of hybrid geopolymer mix (cement + GLD + blast furnace slag) as a binder, compressive strengths. Different compression test ages are seen in the figure

The results can be compared to the reference compressive strength results for cement and blast furnace slag as a binder (Figure 9).

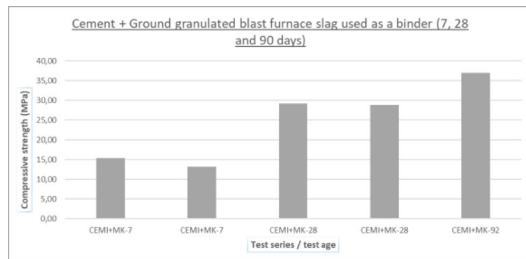


Figure 9: Reference compressive strength results for cement and blast furnace slag used as a binder, with different test ages.

Heat treatment seems to have positive effect on the strength at least in early age (Figure 10). The effect is not so obvious after 28 days. Usually heat treatment improves the strength of geopolymers. However, heat treatment is not a standard procedure in concrete industry.

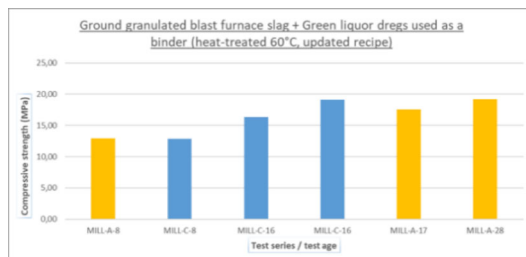


Figure 10: Example of the effect of heat treatment to compressive strengths.

3.4 Ongoing development

The results from freeze-thaw are still pending. The preliminary results showed several decomposition damage in test pieces since special protective agents (air entraining admixtures) for frost resistance were not used. Also the samples made with OPC were damaged the same way as GLD samples. Next step is to test samples made with air entraining admixture, which hopefully improves the resistance to freezing. Results are pending.

The next focus in the project is to accelerate the hardening process. NAOH solution is already commonly used in the production of geopolymer concretes. Test results show that adding of NAOH in the mixture of GLD and GGBF accelerates the hardening.

4 CONCLUSION

In the project, one of the original ambitious goals was also to test biobased ash resulting from the pulp production process as a substitutive agent in concrete. The focus in the research has now centered to GLD since its focal status as the most problematic waste to recover, produced in high volumes from forest industry in Finland. At this point, testing biobased ash in similar demonstrations were not conducted in this project.

The results show that small amounts of cement can be replaced with GLD without significant changes in strength. Actually, it was found out that the replacing 5% of cement with GLD had very little affect in the strength of the test pieces.

Probably one the most important outcomes in this project is the possibility to make concrete-like mixtures totally without cement. What makes it even more significant is the fact that geopolymers can be made without NaOH and sodium silicate solutions. This will influence the chemicals used in the process, and in the same way, effect greatly to the environmental effect. Essentially these kind of geopolymer concretes could be made totally from recycled materials if the aggregates would also be replaced with recycled materials such as crushed concrete.

GLD can also be used as fine aggregate in concrete. That makes it possible to reduce the utilization of virgin materials such as sand. GLD can also be used in hybrid geopolymer mixes with cement and GGBF, which is interesting aspect to look more carefully into.

5 REFERENCES

- [1] Material economics. 2019. Industrial transformation 2050 – Pathways to Net-Zero Emissions from EU Heavy Industry.
- [2] Activity report. 2018. CEMBUREAU.
- [3] Andrew, R, Global CO₂ emissions from cement production. *Earth System Science Data* (2018) 10: 195-217.
- [4] M. Illikainen, E. Adesanya, K. Ohenoja, T. Luukkonen, P. Kinnunen, One-part geopolymer cement from slag and pretreated paper sludge, *Journal of Cleaner Production* (2018) 185: 168-175.
- [5] Rui. M., Novais, J. Carvalheiras, L. Senff, J.A. Labrincha, Upcycling unexplored dregs and biomass fly ash from the paper and pulp industry in the production of eco-friendly geopolymer mortars: A preliminary assessment, *Construction and building materials* (2018) 182: 464-472.
- [6] Waste statistics. Finnish forest industries federation. 2019. <https://www.forestindustries.fi/statistics/environment/>
- [7] T. Kinnarinen, M. Golmaei, E. Jernström, A. Häkkinen, Separation, treatment and utilization of inorganic residues of chemical pulp mills, *Journal of Cleaner Production* (2016), 133: 953-964.

- [8] K. Manskinen, Utilisation aspects of ashes and green liquor dregs from an integrated semichemical pulp and board mill (2013), pag. 137. Doctoral dissertation 102/2013. Aalto University.
- [9] K. De Weerd, Geopolymers, State of the Art (2011), pag. 37. Sintef.
- [10] Luukkonen, T., Abdollahmejad, Z., Yliniemi, J., Kinnunen, P., Ilikainen, M. One-part alkali-activated materials: A review. Cement and Concrete Research (2018) 103: 21-34.

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