V International Conference Progress of Recycling in the Built Environment
10-12 October 2023 – Weimar, Germany

ZERO-EMISSION CIRCULAR CONCRETE

Peter Stemmermann (1), Krassimir Garbev (1), Angela Ullrich (1), Günter Beuchle (1)

(1) Institute for Technical Chemistry (ITC), Karlsruhe Institute of Technology, Germany

Abstract

The project Zero Emission Circular Concrete develops a CO_2-neutral, high-quality, and resource-efficient concrete cycle starting with end-of-life concrete. A cement clinker with a reduced CO_2 footprint is processed from waste concrete fines at a strongly reduced temperature of approximately 1000°C. The main clinker mineral is belite, Ca_2SiO_4. The residual CO_2 is released in concentrated form and used for the technical carbonation of either waste concrete fines as supplementary cementitious material or of coarse crushed waste concrete. The coarse fraction is treated in a new process based on a pressurized autoclave, where hardening by carbonation improves the properties of the recycled aggregate. Both carbonation options are investigated on a laboratory scale.

Recycling cement is developed from belite cement clinker, Portland cement clinker, and other substitutes in a joint project with the industry. A 30% substitution rate of Portland cement clinker compared to European cement standards is targeted. Subsequently, formulations for recycling concrete will be developed from recycled cement and recycled aggregate. The processing of concrete products and precast concrete elements will be tested in plant trials.

A pilot plant for belite cement clinker is currently under construction to bring its technology readiness level to four.

1. Introduction

The production of concrete is responsible for about 6 to 9 % of all man-made CO_2 emissions, whereby these emissions are almost exclusively linked to the upstream production of Portland cement clinker (PCC). CO_2 from the clinkering process originates to about 60% from the calcination of limestone (process emissions) and about 40% from fuels.

Waste concrete, on the other hand, could capture up to 80% of the process emissions generated during cement manufacture by recarbonation. During service life, demolition, and subsequent crushing, a value of about 5% is observed [1].

Today's standard procedure for the recycling of concrete waste neither includes its use for carbon sequestration nor for the production of cement clinker. Waste concrete is usually processed into crushed aggregate as a substitute for natural aggregate in road construction.

Therefore, an excess of crushed concrete fines remains, which is hardly usable. In Germany, the economic value of the recycled product is currently about 1/3 of fresh concrete, which is why it's called downcycling. In the considered case, downcycling reduces the demand for natural raw materials but does not contribute to reducing CO_2 emissions in the cement industry.

In principle, it is possible to use waste concrete fines in particular as a raw meal component for cement clinker production in existing cement plants, whereby CO_2 emissions could be slightly reduced. Economically, however, the transport of concrete waste is limited to a distance of about 20 km, so at best a very small proportion of concrete waste could be used. The situation will become even worse in the future, when the need to implement carbon capture, use, and storage (CCUS) will require investments in expensive infrastructure for the transport of CO_2, which, according to the current state of the art, is only economical for very large plants. A technical solution for high-quality closed-loop recycling of waste concrete with an average transport distance of 20 km of secondary raw materials is missing.

2. Zero Emission Circular Concrete at KIT

The KIT project for the production of zero-emission circular concrete is therefore aiming at technologies that are designed for stand-alone plants with a cement clinker capacity of about 50,000 t/a, far smaller than existing cement plants. The objective of the project is to reduce CO_2 emissions from clinker production by using crushed concrete fines as a second component besides limestone in the production of cement clinker (figure 1). The unavoidably released CO_2 is then to be fixed as completely as possible in other mineral wastes, preferably in crushed aggregate from old concrete. This is done by crystallizing carbonates in a hydrothermal process. Infrastructure e.g. pipelines for CO_2 transport thus become obsolete. The structural properties of the aggregate are even improved due to pore closure induced by the growth of carbonates. Carbonated crushed aggregate shall be used together with recycled cement clinker for the production of recycled concrete without the need for an increased cement content compared to standard concrete formulations.

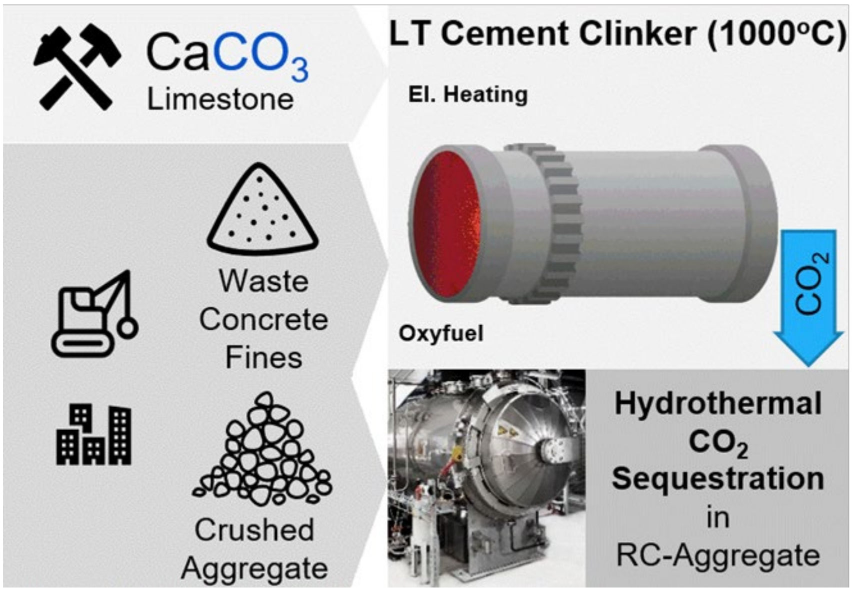

Figure 1: Combined low-temperature processing of belite cement clinker and carbonated recycled aggregate from waste concrete and limestone aiming at zero CO_2 emission.

3. Low-temperature belite cement clinker (LT-BCC) from waste concrete

To process belite cement clinker at low temperatures, a new cement raw meal characterized by a lower lime content and the addition of mineralizing agents as well as new process conditions were investigated [2]. The technology consists of a rotary kiln with a maximum process temperature of 1000ºC, preferably operated electrically or with oxyfuel combustion in an almost pure CO_2 atmosphere. The combined presence of CO_2 and selected mineralizing agents, e.g. $CaCl_2$ or Na_2CO_3, results in the formation of a carbonate-rich melt at temperatures below 700ºC, which allows extremely fast clinkering. Concrete-damaging components such as sulfate and chloride are bound in dedicated and insoluble reservoir minerals [3].

In the raw meal, limestone is partially replaced by the hardened cement paste contained in the crushed concrete fines. A further reduction of the limestone content compared to PCC is achieved by the low molar CaO/SiO_2 ratio of 2, which overall cuts the process emissions by about 30%.

The LT-BCC produced consists to a large extent (>60 wt.%) of the calcium silicate belite (Ca_2SiO_4) present in both polymorphs: β-C_2S and $\alpha`_H$ C_2S. The most common clinker mineral of today's PCC, alite (Ca_3SiO_5) is not present. LT-BCC is intended to at least partially replace PCC in accordance with cement standards. The initial target in the research project URBAN [4] is a substitution rate of 30 wt.%. High sulfate contents in the waste fines may result in the formation of ternesite or ellestadite if $CaCl_2$ is used as mineralizing agent. Besides clinker, highly concentrated CO_2 exits the kiln (90-95 wt.%).

LT-BCC processing has been successfully tested using different wastes e.g. post demolition autoclaved aerated concrete (AAC) [5] and has been demonstrated on a 1.5-ton scale using waste concrete fines. The replacement of 25 wt.% Portland cement by LT-BCC in the production of AAC has been demonstrated on a technical scale [6]. The total reduction of emissions depends in particular on the available electricity mix. If only electricity from renewable sources is used, 400 kg CO_2 per t LT-BCC was calculated [7].

A pilot plant with a capacity of 10kg/h is currently being built at KIT funded by the Ministry of the Environment Baden-Württemberg [8]. The closing of the concrete cycle is being optimized in the URBAN project [4] together with several industrial companies (cement manufacturers, construction chemicals companies, and manufacturers of precast concrete parts). As part of the Innopool project EuK [9], a superstructure approach is being developed in collaboration with the Jülich Research Centre [10] to identify scenarios under which the production can become economically and ecologically viable.

4. Hydrothermal carbonatization of recycled aggregate from waste concrete

The mechanical processing of waste concrete leads preferentially to fractures in the hardened cement paste. Therefore, about 50% of the cement content of waste concrete is found in the crushed fine fraction, although it represents only about 30% of its total mass. The other half of the cement remains in the recycled aggregate and increases both, its specific surface area and porosity, which degrades its properties compared to natural aggregate. In the production of RC concrete, this is usually compensated for by increasing the cement content.

One possibility to close the porosity of hardened cement paste is carbonation, which causes an increase in volume. However, under standard conditions, turnover and reaction rate are very limited. Freshly precipitated calcium carbonate closes pores and cracks near the surface thus preventing the further transport of carbonate ions into the interior of the grain.

One approach to increase the carbonation yield is to further increase the specific surface area by crushing. However, the material can then no longer be used as concrete aggregate, but only as a substitute for sand or as supplementary cementitious material (SCM). The use as SCM is pursued in the Marie Skłodowska-Curie Doctoral Network CO2Valorize funded by the European Commission.

The aim of the approach pursued in the zero-emission circular concrete project is to reduce clogging using suitable reaction control in a technical autoclave, similar to that used for AAC production. The effectiveness of the approach has been demonstrated on a laboratory scale. For carbonation, the concentrated CO_2 from the clinker kiln is used. A conversion of > 80% of the sequestration capacity within a treatment period of 6h is aimed at. The autoclave is heated by the waste heat of the clinker kiln.

11. Conclusions

High quantities of cementitious mineral wastes such as waste concrete, post-demolition autoclaved aerated concrete (AAC), and others on the one hand, and decreasing quantities of landfill capacities for mineral residues on the other hand require higher recycling rates in Germany. The approach of zero emission circular concrete developed at KIT combines high-quality closed-loop recycling with the sequestration of released CO_2 into the generated products. The concept is primarily designed for small, decentralized plants with an output of about 50,000 t/a cement clinker, which corresponds according to own calculations to the waste concrete fines generated annually in an urban area of about 1 to 1.5 million inhabitants, and within an average transport distance of 20 km.

The zero-emission circular concrete plant consists of a clinker line for LT-belite cement clinker, which is coupled with an autoclave line for the carbonation of the aggregates. Both modules could also be operated independently. An LT-BCC line at the site of an existing cement plant would be possible.

The variable composition of secondary raw materials derived from wastes and the potential contamination with constituents harmful to concrete requires, on the one hand, an elaborate and as yet not established analytical routine and mixing technology. On the other hand, cement standards may have to be adapted if the concept of insoluble reservoir minerals presented is to be used for general applications.

Acknowledgments

The research was supported by the Ministry of the Environment Baden-Württemberg, the German Federal Ministry of Education and Research, the German Federal Ministry of Economics and Climate Protection, and the Helmholtz Association.

The article was originally published in Rilem Proceedings PRO 137, Proceedings of the V International Conference Progress of Recycling in the Built Environment, 10-12-October 2023, Weimar, Germany, edited by Ines Döring, Yury Villagrán Zaccardi and Anette Müller.

References

[1] Achternbosch, M. and Stemmermann, P., "The carbon uptake by carbonation of concrete structures – some remarks by perspective of TA," 1000131215, 2021, doi:10.5445/IR/1000131215.

[2] Hunsinger, H., Beuchle, G., Stemmermann, P., Schweike, U. et al., "Verfahren zur Herstellung von Dicalciumsilikat," WO 2014/019656 A1.

[3] Ullrich, A., Garbev, K., Schweike, U., Köhler, M. et al., "$CaCl_2$ as a Mineralizing Agent in Low-Temperature Recycling of Autoclaved Aerated Concrete: Cl-Immobilization by Formation of Chlorellestadite," *Minerals*, 2022, doi:10.5445/IR/1000150781.

[4] "Verbundvorhaben URBAN - CO_2-reduzierter Beton durch Upcycling von Reststoffen aus der Betonaufbereitung und CCU," FKZ 03EE5130A BMWK, 7. Energieforschungsprogramm, 2022 bis 2025.

[5] "ReziProK - Verbundvorhaben REPOST - Recycling-Cluster Porenbeton: · Optimierung von Rückbau und Sortierung von Altporenbeton und Erarbeitung neuer Optionen für die Kreislaufführung von Porenbeton - TP 4: Dicalciumsilikat aus Porenbetonresten aus Abbruchmaßnahmen,"(FKZ 033R249D), 2019-2022.

[6] Stemmermann, P., Ullrich, A., Beuchle, G., Garbev, K. et al., "Belite cement clinker from autoclaved aerated concrete waste – A contribution towards CO_2 -reduced circular building materials," in *CE/Papers 2022 – 2022 Xella International Colloquium*:17–26.

[7] Stemmermann, P., Volk, R., Steins, J.J., and Beuchle, G., "Recycling belite cement clinker from post-demolition autoclaved aerated concrete – assessing a new process," *submitted to Journal of Industrial Ecology*, 2023.

[8] "R-Zement: CO_2-arme Herstellung des Klinkerminerals Dicalciumsilikat aus Recycling - Baustoffen,"(L7521115), 2021-2024.

[9] "Innopool Projekt Energiewende und Kreislaufwirtschaft (EuK): Dekarbonisierung und Flexibilisierung energieintensiver Recycling-Prozesse," 1/2021-12/2023.

[10] Paredes, A.Y.O., Sandaka, G., Beuchle, G., Stemmermann, P. et al., "Superstructure optimization for a novel cement production process utilizing secondary raw materials," *Abstracts*, Helmholtz Energy Conference, Koblenz, 12. - 13.6., 2023.