Accelerated Carbonation of Construction Materials by Using Slag from Steel and Metal Production as Substitute for Conventional Raw Materials

M. Prokein[a], K. Fuchs[b], N. Mölders[a], M. Renner[a], E. Weidner[a,b]

[a]*Fraunhofer UMSICHT, Osterfelder Str. 3, 46047 Oberhausen*

[b]*VTP Ruhr University Bochum, Universitätsstraße 150, 44801 Bochum*

**Corresponding author: michael.prokein@umsicht.fraunhofer.de*

1. Introduction

Quicklime, sands and water are the raw materials for the production of lime sandstone. In terms of raw materials and energy consumption, the production process of this building material is of high concern. The production of quicklime from limestone causes high CO_2 emissions, the availability of sands for the construction industry decreases and the energy consumption for hydrothermal curing is enormous. Hydrothermal curing operates at about 15 bar and 200 °C for 12 h. For this reason, we investigate the possibility to substitute quicklime and sand for the production of building blocks by varoius types of slag as calcium rich waste from the steel making process. Instead of conventional hydrothermal curing, we perform CO_2-curing.

2. Materials and Methods

Six types of slag (Linz-Donawitz (LD), ferrochrome (FeCr), ladle (LS), stainless steel (SS), ladle furnace (LF), electric arc furnace (EAF)) provided by »thyssenkrupp MillServices & Systems GmbH« were milled to powders (particle sizes between 12 and 25 µm) at »Loesche GmbH«. Cylindrical blocks with a diameter of 100 mm and a height of 40 mm consisting of pure slag powder as well as mixtures of slag powder and sand were mingled with 7 wt.-% of water, shaped and compacted in a press at 12 MPa. The influences of pressure, temperature and time on the CO_2-curing process of LD-slag based blocks were investigated in a 2 L high-pressure autoclave (P_{max} = 30 MPa; T_{max} = 300 °C). Three blocks with the same composition were treated per trial. Two trials were performed per parameter set. Pressures between 0.1 and 5 MPa, temperatures between 25 and 140 °C and times between 1 and 100 h were considered. The quality of the CO_2-cured blocks was assessed and compared to conventional lime sandstones by measuring the compressive strength and frost resistance by »Ruhrbaustoffwerke GmbH & Co. KG«. The degree of carbonation was assessed by thermogravemetry (TG) and X-ray diffraction (XRD) measurements. The pH-trends in the cross section of the blocks were observed using phenolphthalein as liquid pH indicator. The parameter set that lead to the material with highest performance was tested on all slag types. In addition, the process was scaled to steel-slag based building blocks (240 mm x 115 mm x 60 mm) provided by »Ruhrbaustoffwerke GmbH & Co. KG« and CO_2-cured in a 20 L high-pressure autoclave (P_{max} = 320 bar; T_{max} = 80 °C).

3. Results and discussion

The results show that CO_2-curing of building blocks consisting of pure wetted LD-slag cause severe cracking of the cylindrical samples. This is because the high carbonation efficiency of the calcium rich material. The high CO_2 uptake cause the samples to expand. However, by using 35 wt.-% of LD-slag for substituting quicklime totally and sand partially building blocks without a change in geometry and high compressive strength are produced.

The experiments for the determination of the optimal pressure and temperature indicate 2 MPa and 50 °C as promising parameter set for the CO_2-curing process. At these parameters and after 1 h of CO_2-curing, the compressive strength of the blocks reaches values of conventional lime sandstones. By increasing the time to 3 h, the compressive strength of LD-slag based blocks reaches the highest mediate value of nearly 50 N/mm^2. This is more than twice as high compared to conventional lime sandstones. Longer CO_2-curing times do not lead to higher compressive strength. The increase in overall compressive strength due to the increase in curing time is caused by raising the degree of carbonation of the Ca-rich material to the center of the sample. Figure 1 shows the trend of the pH-value assessed by phenolphthalien in the cross section of three slag based blocks after CO_2-curing for 1, 3 and 24 h. Between 1 and 3 h of CO_2-curing a carbonation front in the cross section is observed. XRD and TG measurements confirmed the formation of carbonates. The carbonate content increased from 2 wt.-% of the not treated stones to more than 20 wt.-% after CO_2-curing. The amount of inorganic carbon increased from 1 wt.-% to 11 wt.-%.

2 MPa, 50 °C, 1 h | 2 MPa, 50 °C, 3 h | 2 MPa, 50 °C, 24 h

Figure 1: Pictures of the cross section of LD-slag based blocks after CO_2-curing at 2 MPa, 50 °C and different times, 1h (left), 3 h (middle), 24 h (right).

Figure 2 shows the influence of the different slag types on the compressive strength after CO_2-curing at 2 MPa, 50 °C and 3 h. All slag based stones show higher compressive strengths compared with conventional lime sandstones. However, the type of slag influences the compressive strengths values significantly. XRD measurements indicate that this is due to variating carbonation efficiencies and mineralogy of the different slag types. In addition, the particle size of the slag powders influence the achievable compressive strength, significantly. Thereby, finer particles allow higher compressive strength.

The results of the tests in the 20 L equipment were in good agreement with the results of the 2 L trials.

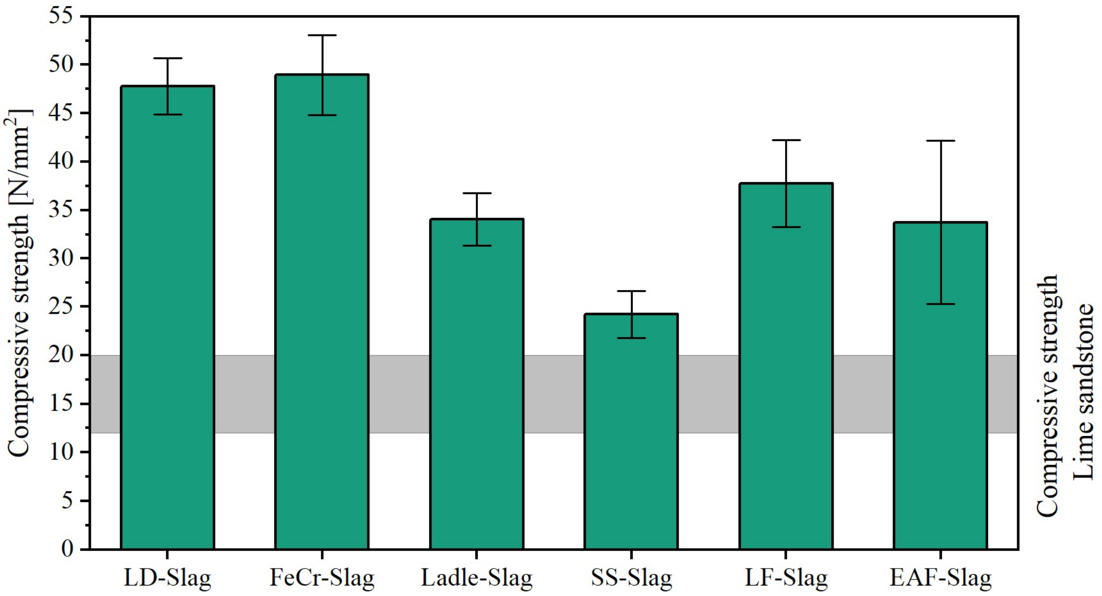

Figure 1: Compressive strength of various slag based building blocks with a slag content of 35 wt.-% after CO_2-curing at 2 MPa, 50 °C and 3 h compared with the compressive strength of conventional lime sandstone.

4. Conclusions

In the conventional process, lime sandstones produced from quicklime and sand are hydrothermal cured in a saturated steam atmosphere at about 15 bar, 200 °C for 12 h. The desired process parameters for CO_2-curing are 2 MPa, 50 °C and 3 h. Since autoclaves for the production of lime sandstones are already used in the construction industry, the CO_2-curing process with its moderate operating conditions has high potential for industrial application. The CO_2-curing process consumes high amounts of CO_2 instead of emitting it and lead to high quality building blocks. The development is still in progress and the project includes an LCA study. Data for the comparison of the conventional lime sandstone process with the carbonation method will allow assessing the CO_2-curing process in detail in the near future.