

Mineralogical effects on the intensified mineral carbonation of steel slags: kinetics, conversion, basicity and products

R.M. Santos (1), M. Bodor (2), J. Van Bouwel (1), L. Kriskova (3), J. Elsen (4), M. Vlad (2), and T. Van Gerven (1)

(1) Dept. Chemical Engineering, KU Leuven, Heverlee, Belgium (rafael.santos@cit.kuleuven.be; thomas.vangerven@cit.kuleuven.be), (2) Dept. Environmental Engineering and Metallurgical Technological Systems, University of Galati, Galati, Romania (bodormarius@yahoo.com; maria.vlad@ugal.ro), (3) Dept. Metallurgy and Materials Engineering, KU Leuven, Heverlee, Belgium (lubica.kahalova@mtm.kuleuven.be), (4) Dept. Earth and Environmental Sciences, KU Leuven, Heverlee, Belgium (jan.elsen@ees.kuleuven.be)

Mineral carbonation, the reaction of carbon dioxide with alkaline minerals, is an attractive carbon sequestration approach owing to the geochemical stability of carbonates. When applied to low-value or hazardous industrial alkaline residues, it can result in the reduction of basicity and leaching, enabling valorisation or safe disposal. However, process limitations including high energy intensity, low reaction conversion, and slow reaction kinetics, have thus far prevented mineral carbonation of wastes from being widely applied. These barriers are caused by inefficient processing, but also by mineralogical aspects inherent to the materials.

Alkaline waste materials are typically composed of several mineral phases that may or may not be susceptible to mineral carbonation, and which, if reactive to CO₂, may exhibit varying degrees of carbonation kinetics and influence on the material's basicity. The formation and character (e.g. thickness, porosity) of passivating layers can also be affected by the relative solubility of the minerals. Most carbonation studies to date, however, have focussed on the chemical composition of these materials rather than on their mineralogical composition. The commonly used Steinour equation, for example, relies solely on the amounts of alkali oxides to predict CO₂ uptake capacity. Though stoichiometrically accurate, this prediction can be overly optimistic, causing doubts whether carbonation processes are ineffective in reaching complete conversion (due to insufficient process severity or formation of passivating layers), or if the unreacted material is inert to carbonation.

In the present work, three types of steel slags are utilized for mineral carbonation: argon oxygen decarburization slag (AOD), continuous casting slag (CC), and basic oxygen furnace slag (BOF). This study aims to provide insight on the carbonation behaviour of various minerals commonly found in steel slags, including: lime, portlandite, periclase, brucite, γ - and β -dicalcium silicates, srebrodolskite, bredigite, cuspidine, merwinite, åkermanite, and gehlenite. The carbonation products, which include various forms of calcium and magnesium carbonates (e.g. calcite, aragonite...), silica and intermediate products (e.g. wollastonite) are also characterized, both with respect to composition and to the process conditions that promote their formation (temperature, ionic concentrations, sonication).

Three experimental approaches of increasing levels of process severity have been tested and optimized, and are discussed in this work: (i) mild wet carbonation; (ii) accelerated slurry carbonation, and (iii) rapid high temperature direct carbonation. A comparison is made of the characteristics of these processes and the results achieved, with respect to carbonation kinetics, mineral conversion and basicity reduction. In addition to carbonation of the innate slags, high purity single minerals (synthetic and geological) were also prepared and tested to reveal in more detail their carbonation behaviour. Mineralogical assessment was made by Rietveld refinement technique of XRD diffractograms.

The understanding of the mineralogical behaviour of alkaline materials towards mineral carbonation can enable the optimal development of intensified processes that better meet energy and economical demands of carbon sequestration technologies, as well as materials valorisation requirements. The outcome includes better choice of carbonation technology, optional hot-stage tuning of residue composition, and a priori prediction of carbonation effects, such as CO₂ uptake, basicity and leaching behaviour.