

# Belite cement clinker from autoclaved aerated concrete waste – A contribution towards CO<sub>2</sub>-reduced circular building materials

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## Abstract

The processing of belite cement clinker in a rotary kiln at about 1000°C is a new recycling option for autoclaved aerated concrete (AAC) waste that otherwise must be landfilled. The clinker produced can partially substitute ordinary portland cement (OPC) in AAC production. Waste quantities and landfill costs are minimized, while at the same time CO<sub>2</sub> emissions and the primary resource consumption of AAC production are reduced. The technology is currently under development. New analytical possibilities and modeling have made it possible to optimize the process conditions to such an extent that the use of belite cement clinker in aerated concrete production has already been technically tested. Particularly large effects on CO<sub>2</sub> emissions can be achieved through the electrical heating of the rotary kiln and the coupled sequestration of the released CO<sub>2</sub> in other secondary products such as recycled aggregate for concrete production from waste concrete. Comparable concepts for the AAC cycle are currently being worked on together with the industry partner Xella. Although decentralized plant concepts would be useful in order to minimize transportation, small plants are currently not economical according to initial estimates. In the long term, emission-free product cycles are aimed at.

## KEYWORDS

belite cement clinker, circular building materials, post-demolition AAC, Raman spectroscopy, x-ray-diffraction

## Zusammenfassung

Die Herstellung von Belit-Zementklinker in einem Drehrohrofen bei ca. 1000°C ist eine neue Recyclingoption für Porenbetonabfälle, die sonst deponiert werden müssen. In der Porenbetonherstellung kann der erzeugte Klinker Portlandzement teilweise ersetzen. Nicht verwertbarer Porenbetonabfall und damit Deponiekosten werden vermieden, während gleichzeitig die CO<sub>2</sub>-Emissionen und der primäre

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Ressourcenverbrauch der Porenbetonproduktion sinken. Die Technologie befindet sich derzeit in der Entwicklung. Durch neue analytische Möglichkeiten und Modelle konnten die Prozessbedingungen sehr schnell optimiert und der Einsatz von Belit-Zementklinker in der Porenbetonproduktion technisch erprobt werden. Besonders große Effekte auf die CO<sub>2</sub>-Emissionen lassen sich durch die elektrische Beheizung des Drehrohrofens und die gekoppelte Sequestrierung des freigesetzten CO<sub>2</sub> in andere Sekundärprodukte wie z.B. rezyklierte Gesteinskörnung für die Herstellung von Beton erzielen. An vergleichbaren Konzepten für den Porenbetonkreislauf wird derzeit gemeinsam mit dem Industriepartner Xella gearbeitet. Obwohl dezentrale Anlagenkonzepte sinnvoll wären, um den Transportaufwand zu minimieren, sind Kleinanlagen nach ersten Schätzungen derzeit nicht wirtschaftlich. Langfristig werden emissionsfreie Produktkreisläufe angestrebt.

## 1 | INTRODUCTION

Waste from the demolition of buildings and infrastructures accounts for the largest share of waste generation in Germany – even though this material flow is often only incompletely recorded statistically. The recycling of such waste has improved in recent decades. Nevertheless, considerable amounts are landfilled, while landfill capacities are decreasing at the same time. High-quality recycling remains the exception; in most cases, only low-quality recovery is achieved. The demand for many primary raw materials, such as limestone, is thus hardly reduced. This is particularly true for the recycling of concrete, a hardened mixture of water, cement, and aggregates, especially sand and gravel. After processing, crushed old concrete can replace natural aggregates in concrete production. However, this is only done to a small extent and is limited to certain areas of application. The processing steps necessary for recycling always result in fractions that are not economically usable and, in the worst case, have to be landfilled.

This applies in particular to the finest fraction of the concrete mix, which is heavily enriched with cement paste. Recycling cement as a binding agent is not yet possible.

Compared to other building materials, autoclaved aerated concrete (AAC) is still a young building material. Its intensive use only began in the second half of the last century. With a lifespan of about 50 years, the annual volume of post-demolition AAC waste (pd-AAC) is therefore expected to quadruple to about 4 million m<sup>3</sup> in 2050 [1]. Currently, the recycling rate of pd-AAC is low. One reason for this is the low volume generated to date. On the other hand, pd-AAC does not fit into the established recycling processes for other construction waste because of its high porosity and limited compressive strength. Therefore, special recycling methods have to be developed.

There are various approaches to making material flows recyclable. Starting with a recycling-friendly product design, this continues with direct reuse and finally ends with reprocessing of materials after sorting and pretreatment. A special characteristic of building materials is their long-lasting life cycle. The majority of the products from a century of construction activity are still present today in the built environ-

ment. Optimized technologies for sorting, processing, and secondary use are necessary to close the material loops. Possible technological approaches for the high-quality recycling of pd-AAC are currently being investigated in the research project REPOST, which is funded by the German Federal Ministry of Education and Research, BMBF [2]. Mechanical processing, which is an inevitable step before recycling, always produces fractions that are difficult to use or cannot be used at all. As a rule, most of these fractions are fines and finest particles. Their composition is similar to that of a raw meal for cement production diluted by sand.

An interesting new possibility for the utilization of previously unusable fine fractions from pd-AAC is to substitute natural raw materials in the processing of cement clinker. This could reduce the consumption of natural materials and reduce CO<sub>2</sub> emissions.

One newly developed technology is the production of belite cement clinker at only 1000°C (low-temperature belite cement clinker [LT-BCC]), which is currently being developed at the Karlsruhe Institute of Technology. On the one hand, this development aims to produce an energy-efficient and economical substitute for Portland cement clinker (PCC) and, on the other hand, to minimize the emission of greenhouse gases [3].

## 2 | BELITE CEMENT CLINKER

Belite is the name given to the alpha and beta modifications of the cement clinker mineral Ca<sub>2</sub>SiO<sub>4</sub>, which are formed at high temperatures and remain stabilized at room temperature due to impurities in the manufacturing process. Pure belite decomposes into the gamma modification during cooling, which is not suitable as a clinker mineral. Belite is a major constituent of PCC [4] and as such is one of the most abundant technically produced minerals in the world.

In the 19th century, so-called Roman cement was produced that contained belite as the main mineral [5]. However, in addition to its inhomogeneity due to the production process in shaft kilns, this type of cement had very slow setting and hardening properties, which is not

compatible with modern construction technologies. For this reason, Roman cement was replaced towards the end of the 19th century by Portland cement, whose clinker is burnt in a rotary kiln at much higher temperatures (1450°C). In addition to belite, it contains the clinker mineral alite ( $\text{Ca}_3\text{SiO}_5$ ) as its main constituent, which is responsible for its rapid hydration characteristics.

The production of belite-rich clinker in a rotary kiln is technically possible and offers several advantages. Compared to Portland cement, belite clinker requires a slightly lower lime content for its production. Its processing is possible at significantly lower temperatures around 1200°C. The disadvantage of the slower hydration rate is complemented by a significantly lower heat of hydration compared to Portland cement. Belite-based types of cement are therefore used, for the construction of massive concrete structures such as dams, especially in China, where the dissipation of the hydration heat of ordinary Portland cement would lead to major construction delays [6].

## 2.1 | Low-temperature belite cement clinker for the recycling of waste fines from construction

The use of waste fines from construction as raw material for the production of cement clinker is limited by two main factors: calcium oxide and  $\text{SiO}_2$  content. A raw meal for PCC possesses a molar ratio of CaO to  $\text{SiO}_2$  of about three. A belite cement clinker (BCC) requires a ratio of about two. Secondary raw materials are almost always significantly richer in  $\text{SiO}_2$  than the required raw meal. When using these materials for clinker production, the dilution with  $\text{SiO}_2$  must therefore be compensated either in the form of natural limestone or in the form of secondary CaO sources (e.g., lime slurries). Since BCCs require a lower calcium content in the raw meal, recycling is facilitated.

In addition, there is no clear differentiation between Portland cement and belite cement in European standards, so at least partial replacement of PCC by BCC in standardized cement seems possible. However, a final assessment of this question has not yet been carried out.

In addition, the handling of waste constituents that are harmful to human health, the environment, or the product and that cannot simply be separated before the burning process is of central importance. Since many of these substances are volatile at the high burning temperatures of Portland cement clinker (1450°C), pollutant-enriched partial streams are produced that require special flue gas cleaning. The formation of belite in LT-BCC, on the other hand, begins at temperatures just above 600°C; however, in this temperature range, the reaction is very slow and incomplete. Carbonates in secondary raw materials are often very fine and already deacidify at 500–700°C. Using suitable additives, a good conversion is possible at below 1000°C. In this temperature range, many pollutants can be trapped in insoluble mineral phases (concept of the inner barrier [7]). In this case, however, normative problems do arise when using the clinker produced as a cement component.

Furthermore, the lowered final clinkering temperature of about 1000°C considerably reduces energy consumption. Technological specifications for the kiln are less stringent, and electric heating is

possible. This may become particularly important if smaller plant scales are to be operated in the future since secondary raw materials from demolition are hardly available in consistent quality and quantity to operate kilns with the standard throughput of today's cement plants.

## 2.2 | Processing of low-temperature belite cement clinker

For the processing of LT-BCC, a calcium-rich siliceous secondary raw material is required, for example fines from the processing of pd-AAC, fiber cement waste, or the fine fraction of a stationary concrete recycling plant equipped according to the state of the art. This raw material is thoroughly analyzed, dried, if necessary, homogenized with primary or secondary limestone, and ground to a raw meal of usual fineness in cement processing. A molar ratio of CaO/ $\text{SiO}_2$  of two is aimed at, which corresponds to the stoichiometric composition of belite. Various mineralizing agents such as alkali carbonates or  $\text{CaCl}_2$  can be added to the raw meal to accelerate the reaction kinetics and thus shorten the residence time in the kiln.

The raw meal is fired in a rotary kiln at temperatures up to 1000°C in a pure  $\text{CO}_2$  atmosphere, which is self-created by the deacidification of the limestone contained in the raw meal. Subsequently, the product LT-BCC, which mainly consists of dicalcium silicate or belite, is ground or stored as an intermediate product for further processing.

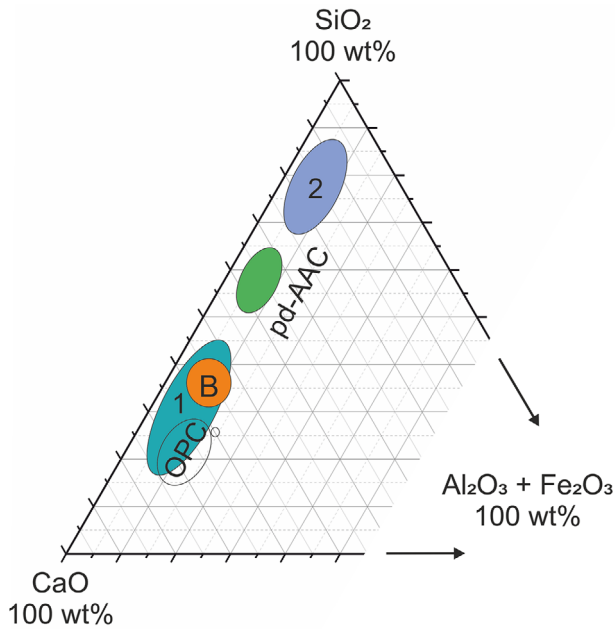
### 2.2.1 | Requirements for secondary raw materials

Suitable secondary raw materials consist of at least 80 wt.% CaO and  $\text{SiO}_2$ , whereby the CaO/ $\text{SiO}_2$  molar ratio is as high as possible. Also important are homogeneity, pollutant levels, and water content. Other important criteria are the regularly available quantities, which are decisive for the security of supply and the possible plant size. Finally, the use as secondary material in clinker production should save disposal costs and contribute economically.

Figure 1 shows that the above-mentioned requirements are fulfilled in particular for fiber cement, aerated concrete, and fines from concrete with calcitic aggregate. For concretes with siliceous aggregate, the quality of the comminution process is decisive. Modern plants can achieve a CaO content in the fine fraction in the range of aerated concrete.

### 2.2.2 | Acceleration of clinker formation below 1000°C

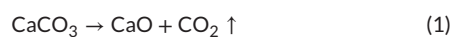
To achieve rapid clinker formation, several factors are crucial: (a) thorough mixing during processing – this is achieved by the rotary kiln; (b) a particle size that is as uniform as possible; (c) a firing temperature that is sufficient to form a melt to initiate sintering and to increase the diffusion rates in solids and melts.



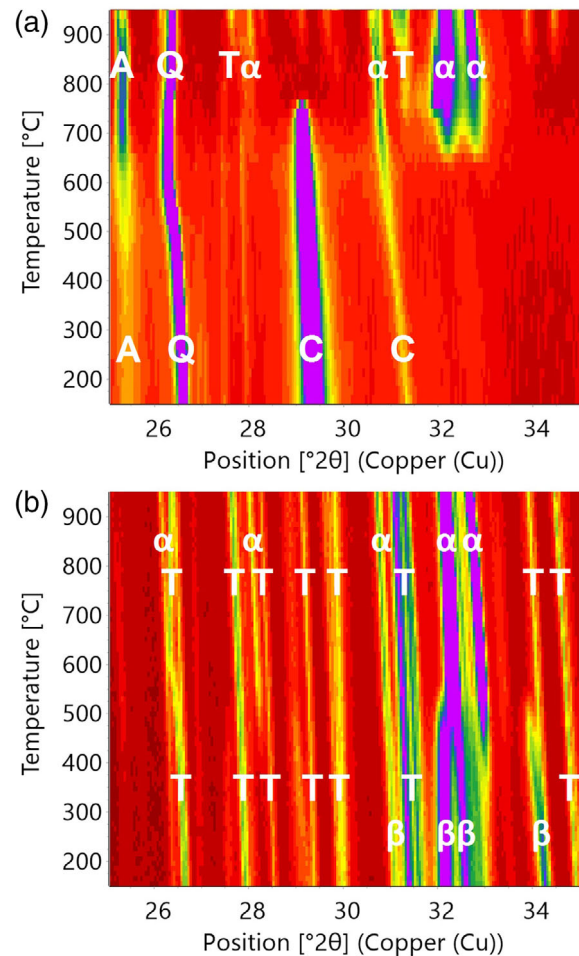
**FIGURE 1** Composition of secondary raw materials suitable for processing of LT-BCC, shown as a projection on the three component system CaO-SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>+Fe<sub>2</sub>O<sub>3</sub>. (1) fiber cement waste and concrete with calcitic aggregate; (2) concretes with siliceous aggregate processed according to today's average standard. B: LT-BCC.

In the binary system CaO-SiO<sub>2</sub>, the conditions for the formation of belite are very well known. However, the first eutectic melt forms only at 1460°C [8]. In addition, the high calcination temperature requires a long heating time and promotes the transfer of volatile elements into the gas phase, which in turn increases the melting temperatures. The growth of intermediate phases during heating also changes the uniform particle size.

The lowering of the eutectic temperature in the process developed at Karlsruhe Institute of Technology (KIT) for the production of LT-BCC is achieved in particular by a CO<sub>2</sub> atmosphere in the rotary kiln. The atmosphere increases the decalcification temperature of CaCO<sub>3</sub> according to equation (1), which for ideal (small) crystals in air already occurs at about 500°C, to 860°C and stabilizes the existence of carbonate melts in the technical system to just below 1000°C. Thus, the crystal growth of the intermediate product CaO according to equation (1) is strongly reduced. The reaction of CaO and SiO<sub>2</sub> to belite according to equation (2) occurs almost simultaneously with reaction (1).



The melt quantity and the content of SiO<sub>2</sub> and CaO in the melt can be additionally increased by adding mineralizers such as sodium carbonate or CaCl<sub>2</sub>. The combination of mineralizers and CO<sub>2</sub> atmosphere



**FIGURE 2** Isolines plot of the in-situ x-ray diffraction measurements in a heating camera: Heating (a) and cooling (b) of LT-BCC raw meal from sulphate-rich pd-AAC. The main phases are marked at the lines as α-α'-H-C<sub>2</sub>S, β-β'-C<sub>2</sub>S, A-anhydrite, Q-quartz, T-ternesite (from Ullrich et al. [9]).

increases the reaction rate of the crystallization of belite according to equation 2 at the final temperature of 1000°C by a factor of about 20.

### 2.2.3 | Processing of low-temperature belite cement clinker from pd-ACC in the laboratory

As part of the REPOST project [2], LT-BCC was produced in a laboratory rotary kiln and a muffle oven. As an example, the clinkering of a sorted pd-AAC from real demolition material with a high sulfate content in an x-ray heating camera is shown in Figure 2 [9]. After homogenization of the pd-AAC powder with CaCO<sub>3</sub> to a raw meal, approximately 50 wt.% of the sample is X-ray amorphous, that is, it consists of highly disordered, very small, or gel-like portions such as calcium silicate hydrate (C-S-H). Because of the required acquisition time in the heating camera, the residence time of the sample above 500°C is more than 10 h. Therefore, the addition of a mineralizer to accelerate

the reaction was not necessary. Also, the CO<sub>2</sub> partial pressure could not be controlled.

During heating, initially, the network plane distances in the crystal structures increase due to thermal expansion. This brings the associated reflections to larger diffraction angles  $2\theta$ . Already at below 800°C, crystalline calcite is completely consumed. At the same time, with  $\alpha'$ -H-C<sub>2</sub>S a high-temperature modification of belite emerges. Due to the high sulfate content of the pd-AAC, which is released during the reaction, anhydrite, CaSO<sub>4</sub>, and, through reaction with belite, ternesite, Ca<sub>5</sub>(SiO<sub>4</sub>)<sub>2</sub>(SO<sub>4</sub>), crystallize. Ternesite is a clinker mineral, common in certain calcium sulfate belite types of cement [10].

During cooling, the network plane distances of the clinker phases contract again. At around 500°C, the high-temperature modification  $\alpha'$ -H-C<sub>2</sub>S mainly transforms into the modification  $\beta$ -C<sub>2</sub>S, which remains as a metastable phase at room temperature, which is also typical for PCC.

### 2.3 | The concept of the inner barrier – use of CaCl<sub>2</sub> as a mineralizing agent

Even with the greatest care in deconstruction, transport, and storage of waste materials from demolition, contamination of the individual material streams, for example, by old paints, plastic residues, fibers etc, is unavoidable. If these pollutants, for example heavy metals, are bound into hydraulic or soluble clinker minerals, they are uncontrollably remobilized during later use. This can impair the hydraulic properties, the performance of the product, and the environmental compatibility. A possible solution to the problem is the targeted addition of additives like CaCl<sub>2</sub>, that are suitable as mineralizing agents and promote the formation of insoluble mineral compounds in which the undesirable components of the raw meal are permanently bound. This concept of the Inner Barrier [7] has already been applied to different technical products such as fly ash and slags from waste incineration [11]. Ellestadite has a particularly suitable structure for storing pollutants. In addition to the incorporation of sulphate, silicate, phosphate, and carbonate as well as fluoride, chloride, and hydroxyl groups, the incorporation of lead, chromate, and strontium has been proven in its apatite type structure [11, 12],

#### 2.3.1 | Low-temperature belite cement clinker from pd-AAC with mineralizer CaCl<sub>2</sub>

The formation of LC-BCC in the presence of CaCl<sub>2</sub> is much more complex than the processing of the same system without mineralizer. Already at about 600°C a eutectic melt forms and drastically increases the reaction rate already at low temperatures.

In addition to the formations of belite, ternesite, and anhydrite, which were already observed without mineralizer, ellestadite Ca<sub>10</sub>(SiO<sub>4</sub>)<sub>3</sub>(SO<sub>4</sub>)<sub>3</sub>Cl<sub>2</sub>, spurrite Ca<sub>5</sub>(SiO<sub>4</sub>)<sub>2</sub>(SO<sub>4</sub>), chlormayenite Ca<sub>12</sub>Al<sub>14</sub>O<sub>32</sub>Cl<sub>2</sub>, and CaO occur in larger quantities. To achieve the

highest possible conversion of C<sub>2</sub>S and at the same time incorporate sulfate completely in ellestadite, a temperature in the range of 950–1000°C is ideal. The maximum yield of  $\alpha$ -C<sub>2</sub>S,  $\beta$ -C<sub>2</sub>S, and ellestadite in various formulations based on pd-AAC was 83%, independent of the sulfate content [13].

The total sulphate content of pd-AAC determines the yield of ellestadite. At a final temperature below 950°C, large amounts of the non-hydraulic mineral spurrite remain in the product. At temperatures higher than 1000°C, chlorine begins to evaporate into the process gas.

#### 2.3.2 | Special analytics

In addition to the classical analysis of chemical and mineralogical compositions as a function of process conditions, which averages over the entire sample and thus allows a representative statement, high-resolution analytics are playing an increasingly important role in understanding local, non-equilibrium processes.

As an example, dusted specimens of LC-BCC synthesized with the addition of CaCl<sub>2</sub> were examined with the scanning electron microscope [14]. The images and elemental distributions acquired with the scanning electron microscope (SEM) did not allow the assignment of individual mineral phases, since the excitation “bulb” of this method is much larger than the sampled crystals and the phases are intensively intermixed. By combining SEM and confocal Raman spectroscopy it was possible to identify individual chemically different areas. Additionally, based on the characteristic oscillations in the crystal lattice, it was also possible to identify regions with enrichments of individual structures such as ellestadite, spurrite, calcite, and anhydrite (Figure 3). In the core of the imaged aggregate, even C<sub>2</sub>S, which is probably formed by the decomposition of spurrite, was detected.

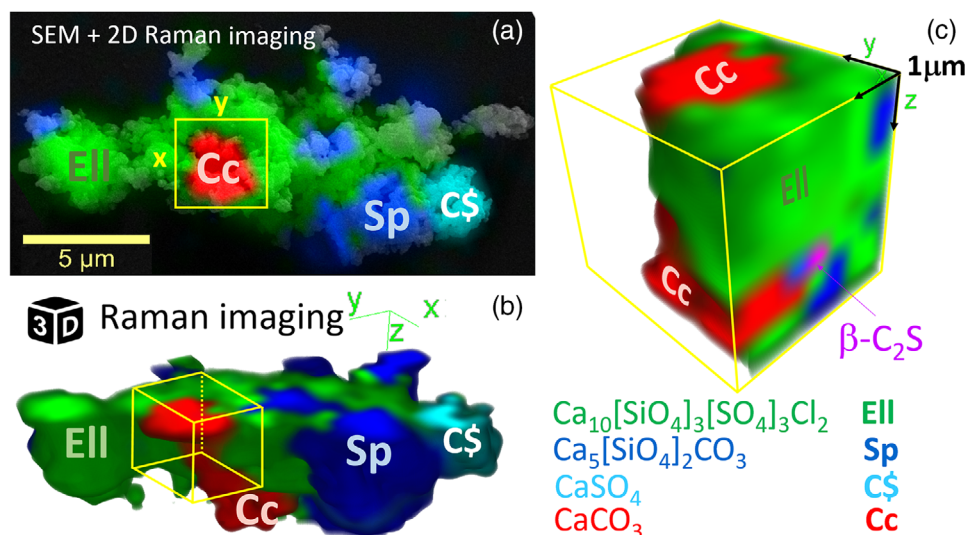
#### 2.3.3 | Non-stoichiometry in low-temperature chlorellestadite

X-ray diffraction analyses with Rietveld refinements of the mineral phases in LT-BCC from pd-ACC with CaCl<sub>2</sub> revealed a deviation of the structure of ellestadite in the temperature range between 700 and 950°C [14].

The generally accepted composition Ca<sub>10</sub>(SiO<sub>4</sub>)<sub>3</sub>(SO<sub>4</sub>)<sub>3</sub>Cl<sub>2</sub> and the symmetry of the crystal structure are modified due to the replacement of



This substitution of sulphate against carbonate can also be seen in the Raman spectra: In addition to the characteristic vibrations of carbonate in spurrite and calcite, a third carbonate band appears, which is characteristic for the incorporation of carbonate ions in ellestadite. The close intergrowth of ellestadite, calcite, and spurrite in syntheses up to 950°C could be shown by individual point analyses.



**FIGURE 3** Top: Overlay of an electron micrograph (SE) of RC-Belite clinker powder made from pd-AAC at 800°C with a corresponding Raman image of the phase distribution in false colors. Bottom: 3D image of the same aggregate with cube-shaped area marked in yellow. Right: clipped 3D image of the cube marked at bottom shows a sub  $\mu\text{m}$   $\beta\text{-C}_2\text{S}$  crystal between spurrite, ellestadite, and calcite.

## 2.4 | Tools for improvement of raw meal composition and process conditions

The complex starting materials and the large number of simultaneously occurring reactions require an efficient tool for process improvement that is based on thermodynamic and fluid-dynamic models.

These models are currently being developed based on the acquired extensive analytical data. Unfortunately, the thermodynamic data available are very incomplete. This applies to individual phases but is especially true for solid solutions and melts. In the above example, it is currently only possible to model the mixed silicate and carbonate-sulphate and halide-containing melts occurring in the process as three partial melts using the FactSage software package [15]. To improve this situation, the database is constantly being expanded based on experimental measurements.

This is all the more astonishing because the systems involved are of high technical relevance. For example, a pseudo-binary section through the system  $\text{Ca}_2\text{SiO}_4\text{-CaCO}_3$  in the  $\text{CaCO}_3$ -rich part of the system at 1 bar  $\text{CO}_2$  partial pressure shows a melt that already forms at low temperatures next to spurrite and only disappears again at about 1000°C. Under similar process conditions, the formation of spurrite is known, for example, as spurrite rings in cement plants, which is promoted by melt formation [16].

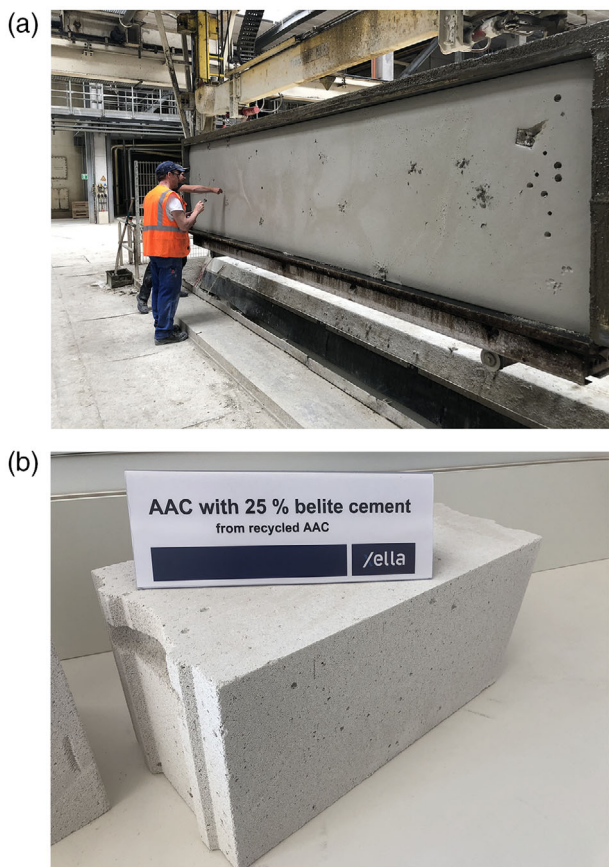
## 3 | STATE OF DEVELOPMENT – PILOT PLANT

The newly developed process has already been tested with various primary and secondary feedstocks and has been optimized on a laboratory scale – first in batches and then in a continuously operating laboratory rotary kiln. Examples of secondary input materials are pd-

AAC, waste fines from concrete recycling plants, fly ash from hard coal and lignite-fired power plants, flotation residues from ore processing and various slags. The complex composition of secondary raw materials requires the balancing and detailed description of element-specific material flows in the clinking process. These data were essential for the design of a small pilot plant with a throughput of 10 kg/h which is currently being built at the KIT [17]. It will be used for further upscaling and improvement of the respective process parameters. For large-scale tests of applications, LC-BCC processing was carried out on a ton scale at a service provider.

### 3.1 | Application of low-temperature belite cement clinker in AAC production

Belite cement clinker is very well suited as a starting material for the production of hydrothermal processed building materials such as AAC. The main issue to solve if ordinary portland cement (OPC) is replaced partly by belite clinker is the slow hydration of belite, which causes a reduction in the initial strength of the green body, which has to be compensated for by other measures. Within the framework of the REPOST project, a total of more than 100 kg of belite clinker was burnt from four different grades of pd-AAC. High and fluctuating sulfate contents in the secondary raw material could be bound by suitable mineralizers. In the small and large-scale pilot plant of the project partner Xella Technologie und Forschungsgesellschaft mbH, formulations for autoclaved aerated concrete were developed that allow the use of high quantities of LT-BCC while complying with the standard and approval-relevant material parameters of the product. Currently, test productions are being carried out in Xella plants based on these new recipes. The aim is to obtain recipe approvals and production recordings. [18].



**FIGURE 4** Production of 5 m<sup>3</sup> autoclaved aerated concrete (YtongPP2-0.35) with 25 wt.% ordinary portland cement (OPC) substitution by LT-BCC in the large-scale pilot plant of Xella Technologie-und Forschungsgesellschaft mbH [18].

The development of ACC formulations containing up to 50% LT-BCC from pd-AAC was successfully completed on a small-technical scale. Recipe validations through upscaling in the large-scale pilot plant are in preparation.

LT-BCC from a 1.5 t small-scale firing test behaves in the same way in application (Figure 4).

### 3.2 | Application as a constituent of cement

When used as a hydraulic binder, belite clinkers hydrate much more slowly than Portland cement clinker but achieve high final strength. The reactivity of belite can be increased by various measures and is the subject of further optimization. First of all, a partial replacement of Portland cement clinker of about 30% by belite clinker is aimed at concerning European standards.

### 3.3 | Firing for the production of LT-BCC

The pilot plant currently being built at KIT will help to investigate and solve essential chemical and product-specific questions. However, cen-

tral technical questions remain open. To generate a CO<sub>2</sub> atmosphere, combustion heating with oxyfuel or electric heating is in principle conceivable. Electrical systems based on resistance heating are limited in size. Other options, such as microwave heating, exist only on a laboratory scale.

In contrast, the oxyfuel technology for cement production is being intensively developed [19]. Further questions concern heat recovery and clinker cooling. This is particularly relevant for the reactivity of the clinker. Currently, clinker coolers are being developed in the cement industry that can be operated in pure CO<sub>2</sub> [20].

## 4 | ENERGY AND MASS BALANCE

A comparative rough estimate of the energy and mass balance of the production of LC-BCC from pd-AAC with CaCl<sub>2</sub> as mineralizer starts with the thermodynamic modelling of the process.

For this purpose, three possible plant configurations for the rotary kiln were compared: (a) heating through methane combustion in air; (b) heating with oxyfuel; and (c) electric heating. Subsequently, the efficiencies for individual variants were roughly estimated in comparison to OPC (Table 1).

The CO<sub>2</sub> saving per kg of clinker is highest for an electrically heated system, although the CO<sub>2</sub> emissions from electricity generation are not considered if the electricity does not come from renewable sources. It is noticeable that the additional CO<sub>2</sub> savings which are possible through oxyfuel are comparatively low. However, the concentration of CO<sub>2</sub> in the exhaust gas rises sharply, which opens up better recycling possibilities.

### 4.1 | Comparative energy balance

Based on estimated thermal efficiencies, the energy balance shown in Table 2 was created. Regardless of the accuracy of the estimation, a drastically increased electricity demand results when switching to a rotary kiln operated in a CO<sub>2</sub> atmosphere.

### 4.2 | Utilization of CO<sub>2</sub>

Although the recycling of cement-like building materials into LT-BCC has the potential to minimize CO<sub>2</sub> emissions, high residual emissions remain. Since only partial streams of the secondary building materials are used for BCC production and the remaining partial streams often have a great potential for sequestering CO<sub>2</sub>, a coupling of LF-BCC production and sequestration may be useful. Currently, various technologies are being developed for the sequestration of CO<sub>2</sub> in construction waste.

It is attractive, in particular, to incorporate CO<sub>2</sub> into crushed coarse aggregates from concrete recycling. Since the fines necessary for the production of LT-BCC clinker are produced during concrete recycling, clinker production and CO<sub>2</sub> sequestration could take place

**TABLE 1** Mass balance of LT-BCC production per kg of clinker for three heating modes of the rotary kiln

Mass Balance					
	Input	g/kg Clinker	Output	g/kg Clinker	Total (Reduction*)
Clinkering Electric Heating	pd-AAC	607	RC-Belite Clinker	1000	
	Tobermorite	363	Belite	852	
	Quartz	144	Ellestadite	138	
	Water	31	CaO	10	
	Gypsum	69			
	CaCO <sub>3</sub>	840	H <sub>2</sub> O	107	107
	CaCl <sub>2</sub> ·6H <sub>2</sub> O	34	CO <sub>2</sub>	369	369 (55%)
	<b>Total</b>	<b>1482</b>	<b>c(CO<sub>2</sub>) off-gas approx. 100%</b>	<b>1476</b>	
+ Oxyfuel	+ CH <sub>4</sub>	47	+ H <sub>2</sub> O	103	210
	+ O <sub>2</sub>	197	+ O <sub>2</sub>	9	9
			+ CO <sub>2</sub>	132	502 (39%)
			<b>c(CO<sub>2</sub>) off-gas approx. 95%</b>		
	<b>Total</b>	<b>1725</b>	<b>1721</b>		
+ Combustion in air	+ CH <sub>4</sub>	12	+ H <sub>2</sub> O	24	235
	+ O <sub>2</sub>	49	+ O <sub>2</sub>		9
	+ N <sub>2</sub>	800	+ N <sub>2</sub>	800	800
			+ CO <sub>2</sub>	38	540 (34%)
			<b>c(CO<sub>2</sub>) off-gas approx. 36%</b>		
	<b>Total</b>	<b>2586</b>	<b>2584</b>		
PCC			CO <sub>2</sub>	820	

For comparison, the specific CO<sub>2</sub> emissions per kg of PCC are given based on the combustion of natural gas.

**TABLE 2** Comparative energy balance for three heating modes of the rotary kiln

Energy Balance							
	Thermal efficiency*	Heat Demand kJ/kg	el. Heating kWh/t	Milling kWh/t	Oxygen production kWh/t**	Oxygen std. m <sup>3</sup> /t	Total el. demand kWh/t
Clinkering Electric Heating	60%	1937	538,08	110			648
Oxyfuel	50%	2606	CH <sub>4</sub>	110	350,23	700	460
Combustion in air	60%	3243	CH <sub>4</sub>	110			110

\* estimation.

\*\* assumption 0.5 kWh/std. m<sup>3</sup> oxygen.

without transport. About half of the sequestration potential of waste concrete remains in the coarse aggregate. In addition, the physical properties of the aggregate, especially its water absorption, are improved by the carbonation of the cement matrix. However, the currently achievable reaction rates are very low. KIT is working on a hydrothermal process that drastically increases the sequestration rate. The final goal is an emission-free circular concrete (Figure 5).

## 5 | SUMMARY

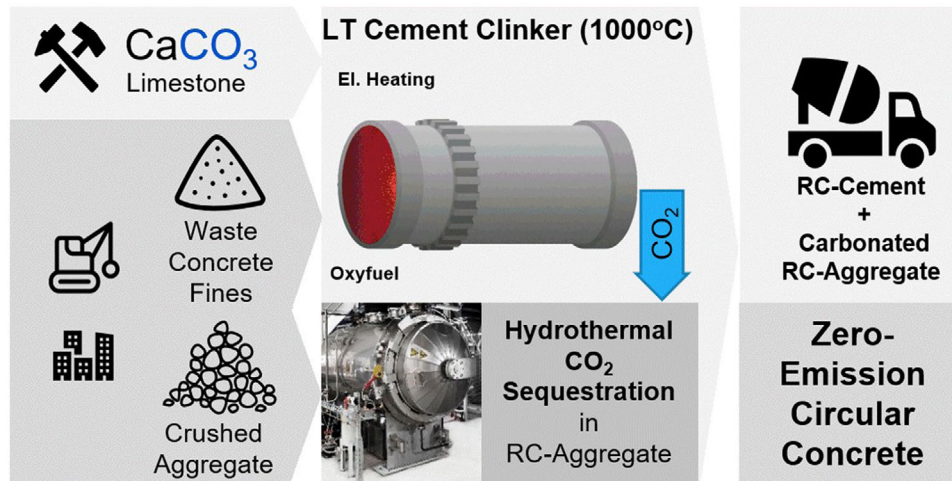
From a technical point of view, the production of LT-BCC clinker for the recycling of pd-AAC is feasible. However, considerable efforts are still

needed in plant and product development as well as in scale-up to gain a secure basis for technology transfer.

The ongoing improvements in the field of analytics and modeling of the reactions and processes involved make it possible to effectively investigate and optimize process conditions and recipes already based on laboratory installations.

Given the current economic conditions, the production and use of LT-BCC can presumably only be economical in individual cases. The economic viability is dominated by the size of the plant and the investment costs, the energy costs and here in particular the electricity price as well as waste disposal fees, CO<sub>2</sub> price and transport costs. Almost all technical aspects are being intensively developed. Some external cost factors such as energy and CO<sub>2</sub> prices, on the other hand, can hardly be planned.





**FIGURE 5** Concept for zero-emission circular concrete by integrated recycling of coarse and fine concrete fractions to LT-BCC and hydrothermally carbonated coarse aggregate.

At the same time as efforts are being made to establish a circular economy for building materials, which have to be processed regionally due to their large masses and low economic value, recycling routes will also have to be adapted and intensified for other material flows, especially for mixed plastic waste and EPS-based composite thermal insulation systems. For these wastes, it is the low density that makes transport very costly. As far as mechanical recycling is no longer possible in the case of plastic waste, chemical recycling processes, based on pyrolysis, should be used, which after regional processing and separation of mineral fractions deliver a transportable intermediate product and replace the classic waste incineration plant.

This creates the need for a new regional architecture of waste disposal and recycling that integrates and networks various partial streams from different recycling processes.

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