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Investigation of Industrial Residues and Waste Materials to Expand the Raw Material Base for the Production of Lightweight Aggregates †

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Abstract: More than 218 million tonnes of mineral construction waste are produced in Germany every year. In view of the shortage of domestic raw materials and an increasing demand for lightweight aggregates and gypsum, it is important to find alternative sources of raw materials for the production of light aggregates. The main focus of our investigations is on construction and demolition waste and industrial by-products, which have so far only been used at a low level. Chemical analyses can be used to classify potential substances in the ternary diagram according to RILEY and to examine their basic suitability. However, the results show that the suitability of a raw material for the production of lightweight aggregates cannot be determined solely on the basis of the classification in the ternary diagram. Experimental investigations are necessary in any case. From the range of residual and waste materials investigated, the rhyolite fractions showed the best bloating properties. Without the addition of bloating agents, bloating values of 1.4 can be achieved. The addition of iron slurry to a reduction in the bloating temperature by about 100 °C, however, is associated with a slight reduction in the bloating value. With our investigations, we were able to show that an expansion of the raw material base for the production of lightweight aggregates is also possible beyond the use of classic, heterogeneous building rubble containing bricks.

Keywords: lightweight aggregates; bloating; mineral residues; mineral processing

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1. Introduction

More than 218 million tonnes of mineral construction waste are produced in Germany every year. In addition, there are further not inconsiderable quantities of mineral by-products from industrial processes [1]. Both mineral construction waste, such as mixed masonry rubble, and industrial residues, such as rock powders, are severely limited in their recyclability. The main reason for the poor recyclability is the heterogeneity of the physical properties and grain sizes. Their use is therefore limited to applications with low quality requirements. In the case of high sulphate contents, the use of even the simplest applications such as landfilling may not be permissible for reasons of groundwater protection. In order to avoid cost-intensive and noncyclical disposal in landfills, completely new ways of processing and recycling building materials must be found [2–4].

One possible approach is the production of lightweight aggregates. For this purpose, material sources that come into question as raw materials are first finely ground, granulated and then sintered in a rotary kiln [2,5]. Due to the material composition, gases are produced inside the granulates, which leads to an increase in volume and a decrease in density. In addition to the recycling of residual materials originally destined for landfill, high-quality raw materials are thus produced which can be used in the construction sector both as crushed and uncrushed lightweight aggregates.

Due to the positive properties of lightweight aggregates in construction, such as weight reduction, thermal and acoustic insulation and their use in geotechnical applications, the demand for lightweight aggregates in the construction sector is expected to continue to increase in the future [2,6].

2. Theory of Bloating

The production of lightweight aggregates requires early sintering of raw materials, which undergo an increase in volume during rapid heating in the temperature range of 1100–1250 °C [7]. The feedstocks must fulfill two basic requirements: they must soften during heating and form a dense, pyroplastic sintered shell. At the same time, gas formation must take place inside the body, which inflates the body and forms a pore system. Therefore, the raw materials used must contain both flux- and gas-forming substances. The viscosity behaviour of the raw materials plays an important role [8]. A viscosity that is too low allows the gas to escape too easily, while a viscosity that is too high hinders the gas pressure and does not allow any bloating effect. The suitability of a raw material for bloating can be determined with the help of a bloating curve, which is created by heating microscopic examinations. The shrinkage–elongation behaviour of the raw material during the heating process and its change in shape are recorded [9].

Accordingly, the influence of the mineralogical and chemical composition of the raw material on the melting and sintering temperature can be checked. Characteristic thermal points are observed and recorded photographically as a shadow image. With increasing temperature, combined with a slight shrinkage of the material, the sintering of the outer shell begins due to crystal water evaporation and mineral transformations [10,11]. In the case of multiphase mixtures of materials, sintering takes place with the participation of partial melts. Sintering is a physical process characterised by the fact that the loose grain contacts between the individual particles of a powdery substance are closed by mass transfer when the temperature rises. This results in the formation of a spatial solidification. The process of sintering can basically be described in 3 steps, as shown in Figure 1 and described below [9,10,12,13].

The melting of the matter inside the granules, the softening of the outer skin and the reaching of the pyroplastic state are depicted as point A, followed by the formation of bloating gas and an increase in volume of the granulate. After reaching the bloating maximum, point B, the volume is reduced due to further increasing viscosity. With the transition to the hemisphere point, the resulting pores fill with melt, and the bloating gases escape to the outside, point C. Further heating causes the hemispherical mass to melt completely, and the floating point is reached [7,9,12,14,15].

The assessment of whether a raw material is suitable for the production of lightweight aggregates is usually made based on RILEY's investigations into the bloating behaviour of clays. The representation takes place in a ternary diagram which was extended by a zone for optimal bloating properties, Figure 2 [16,17]. The diagram is formed from the mass fractions of the main oxides SiO_2, Al_2O_3 and flux (FM) normalised to 100% [17]. In addition to RILEY, the range of good bloating properties has been reviewed and extended by various authors [12]. Leismann [10], for example, used literature data to determine the phase proportions of natural clays that are favourable for bloating behaviour, as shown in Table 1.

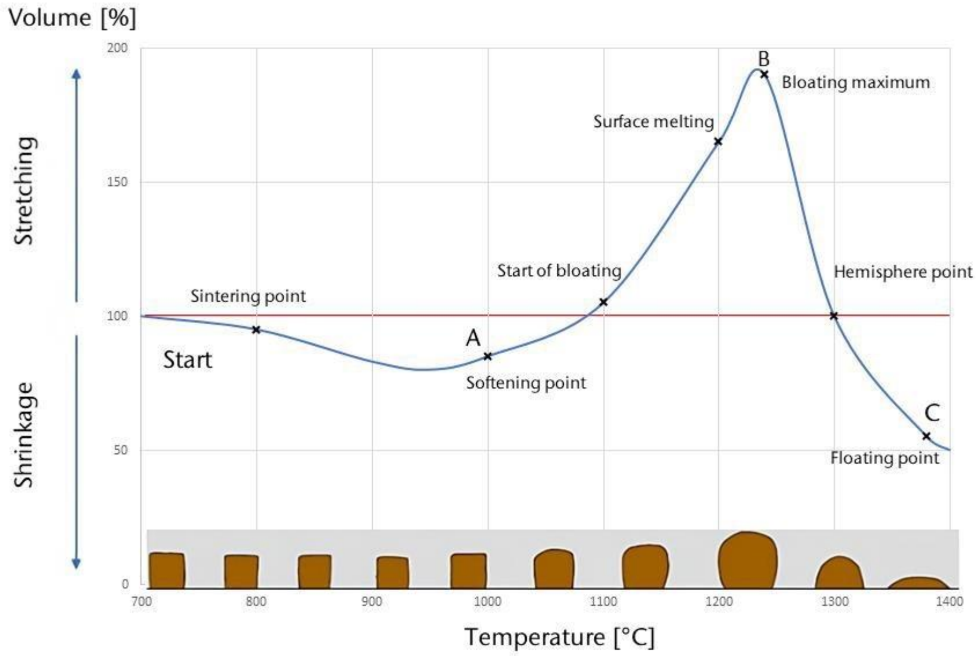

Figure 1. Example of a bloating curve and sample behaviour during the heating process; highlighted with letters A to C are the softening point, bloating maximum and hemisphere point as well as the initial volume of the granules as a red line own illustration according to Diettrich and Kraus [9,12].

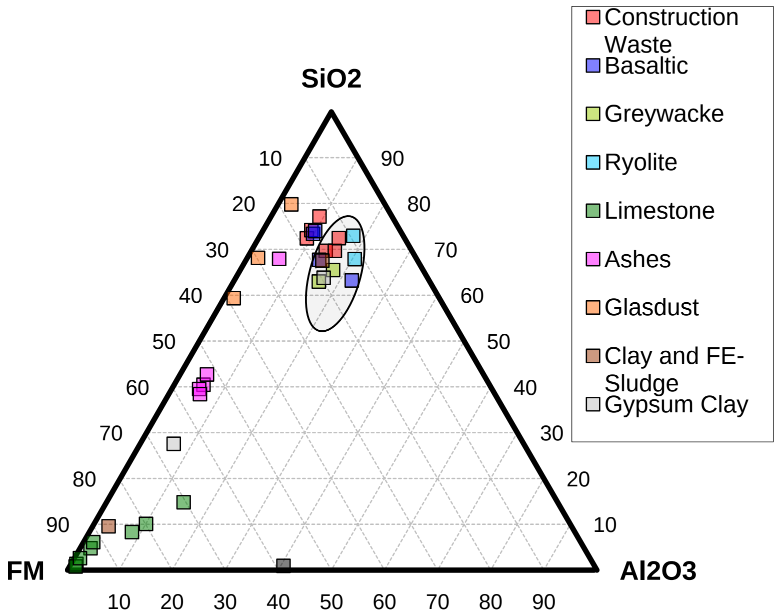

Figure 2. Classification of residual and waste materials in the ternary diagram.

Table 1. Phase components favourable for bloating behaviour according to Leismann [10].

Phase	Share [%]
SiO_2	50–78
Al_2O_3	12–25
FM	8–25
Organic carbon	0.6–5
Fe_2O_3	5–10 (or higher)
CaO	<5
FeS_2	a lot (fine-grained)

The most important gases produced during bloating are released between 850 and 1150 °C from the decomposition of the flux components Fe_2O_3, $CaCO_3$, $CaMg(CO_3)_2$, $CaSO_4$ and FeS_2 [18,19]. These bloating gases are O_2, CO_2, CO, H_2O, N_2, SO_2 and H_2. According to Hoffmann [14], the organic carbon content also has an important influence on the bloating capacity [19]. The carbon not only acts as a reducing agent but also as a regulator of the atmospheric environment in the granulate interior. Ideal carbon contents at the beginning of the pyroplastic melting phase are according to Hoffmann 0.3–0.5% but can vary depending on the material [14,19].

A reducing atmosphere prevails inside the granules due to the bloating gases. This atmosphere is stable due to the gas pressure directed outwards and the continuous generation of further bloating gases. At the same time, the reducing atmosphere leads to a reduction in the firing temperature required for the formation of the bloating gas. For the reduction in iron(III) to iron(II) oxide, for example, this means a decrease in temperature from 1400 °C to 1180 °C. Since this is one of the most frequently encountered gas formation reactions, it is assumed that the oxygen released in the process contributes significantly to the gasification of the carbon and thus to the formation of the reducing environment.

3. Experimental Section

As part of the research that was conducted, 35 residual and waste materials were examined for their potential suitability for the production of lightweight aggregates. Based on the chemical analyses and the classification in the ternary diagram according to Riley, 9 substances were classified as basically suitable. The suitability of these 9 substances was investigated experimentally. Decisive criteria were the granulability and the bloating behaviour in the heating microscope (EMI) and the muffle oven. It was shown that shaping by means of build-up agglomeration on the granulation plate is possible for all materials, although the results vary greatly with the selected parameters.

Furthermore, it was shown in the granulation tests with a dolomite–quicklime mixture that this can be used as an additional binder and noticeably increases the strength of the uncalcined raw granulates.

3.1. EMI Tests

Within the framework of tests with a heating microscope (EMI 301-M17/S—Hesse Instruments) at the Institute for Applied Building Research (IAB) Weimar, the bloating properties of various mixtures could be investigated in advance of the oven tests.

The results showed that the greywacke and rhyolite dusts from natural stone quarrying have basically good bloating properties with a bloating value of 1.4 compared to the initial value. The bloating values of other analysed pure materials are shown in Table 2. In addition to pure materials, a variety of mixtures was tested.

Table 2. Overview of the bloating values of the pure substances in the EMI.

Phase	Share [%]
Greywacke	1.4
Rhyolite	1.4
Broken bricks 0–8 mm	1.1
Tuff	1.1
Rogalith	0

The use of magnesium oxides and dried iron sludge increased the proportion of flux, which led to a deterioration of the bloating properties. The use of greywacke and tuff as a mixture, on the other hand, led to an improvement of the bloating value, compared to pure tuff, to 1.4. The addition of dolomite leads to a deterioration of the bloating properties due to the magnesium content. When using 5% dolomite in the mixture, the reduction in the bloating value is about 10%; doubling the dolomite content to 10% is accompanied by a doubling of the bloating value reduction to 20%.

The use of iron sludge led to a reduction in the bloating temperature. However, since iron slurry is to be understood as a flux, the lower viscosity in the melt also leads to a reduction in the bloating effect. Here, too, the effect is still comparatively small with additive quantities of 5% but becomes clearly noticeable with an increase to 10%. The material system "Rogalith", which served as a reference, did not show any natural bloating properties.

Figure 3 shows the corresponding curves in the measurement diagram. The curves represent the relative change of the resulting and recorded shadow area of the specimen and start with 100% at the beginning of the measurement.

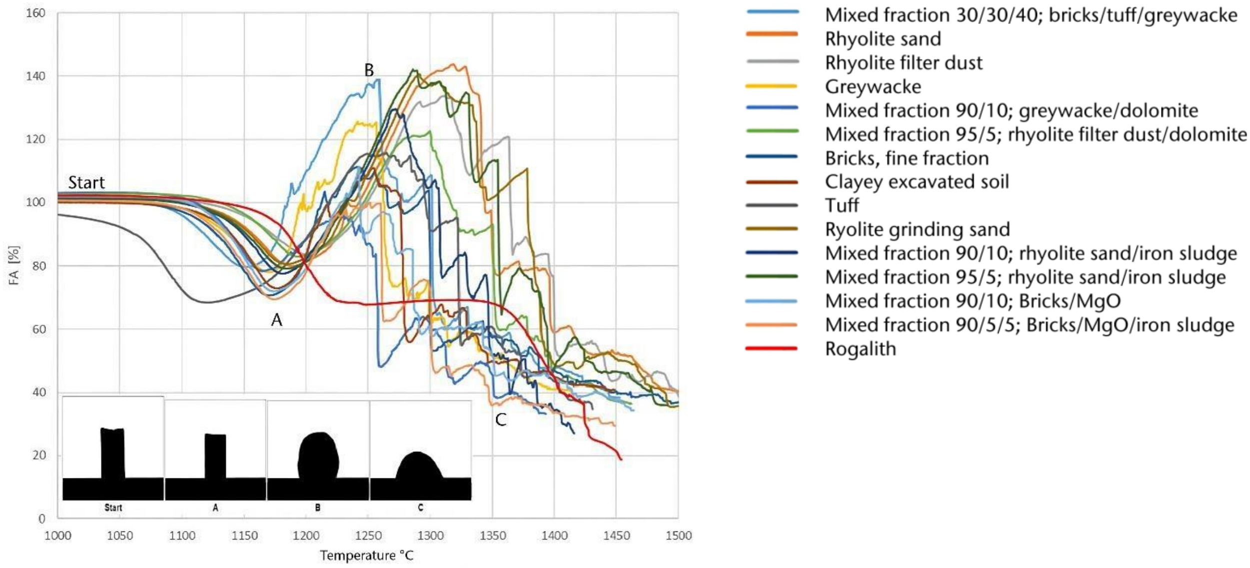

Figure 3. Comparison of EMI results, including EMI silhouettes of rhyolite, highlighted with letters A to C are the softening point, bloating maximum and hemisphere point.

3.2. Oven Tests

The firing tests in the oven were carried out in an electrically heated high-temperature oven from the company Nabertherm GmbH. The granulates were placed at 20 °C in refractory pans lined with kaolin as a separating agent and placed in the oven. Appropriate heating rates and holding times were determined via the oven control. First, heating to 100 °C was carried out within 3 min, with a holding time of 5 min at 100 °C. The temperature was then raised to 500 °C in 5 min.

Then, the oven was heated up to 500 °C in 5 min with a subsequent 5 min holding time. Subsequently, the heating process was continued to maximum temperature (depending on the experiment, 1000 °C, 1050 °C, 1100 °C, 1150 °C or 1200 °C) with a heating rate of 50 °C/min. When the maximum temperature was reached, the samples were kept at the corresponding temperature for 5 min. The subsequent cooling rate was 50 °C/min. At 800 °C, the samples were removed from the oven and cooled further at room temperature.

In the firing tests in the muffle kiln, the pure rhyolite delivers the best results. After calcination in the oven, a bloating value of 1.9 and a bulk density of the granulates of 0.9 g/cm^3 could be achieved. A reference batch with brick dust only achieved a bloating value of 1.5 without the use of a bloating agent. Due to the way the test was carried out in the muffle oven, the granulates are more bloated towards the upper side. The reason for this is the stationary position and the lack of circulation that prevails in rotary kilns. Further investigations in a laboratory rotary kiln are necessary to confirm the results. The results of the firing tests with granulates of rhyolite confirm the results from the heating microscope and the statement made by RILEY on the suitability of magmatic rocks for the production of lightweight aggregates without the addition of expanding agents such as silicon carbide.

Figure 4 shows the calcination results of the rhyolite. At 1000 °C, only a slight sintering of the surface is visible. At 1050 °C, the superficially visible degree of sintering increases, and at this point, the volume of the granules initially decreases. At 1100 °C, the aggregate starts bloating, and bloating values of 1.1 with a density of 1.6 g/cm^3 are reached. At 1150 °C, the surface begins to sinter completely. When the temperature step of 1200 °C is reached, the degree of bloating is 1.9 compared to the uncalcinated aggregate. The density at this point is 0.9 g/cm^3. However, it can be seen from some small pores on the surface that bloating gases have already escaped. For a better overview, a summary of the rhyolite results of the oven tests can be found in Table 3.

Figure 4. Comparison of rhyolite lightweight aggregates, calcinated at temperatures between 1000 °C and 1200 °C. Different aggregates with varying starting sizes.

Table 3. Overview of the results of the oven tests with rhyolite.

Temperature °C	Density [g/cm^3]	Share [%]
1000	2.1	0.9
1050	1.8	0.8
1100	1.6	1.1
1150	1.1	1.5
1200	0.9	1.9

A detailed microscope image of the rhyolite is shown in Figure 5. In the upper region of the aggregates, coarse pores with a diameter of more than 2000 µm are visible. These pores represent larger voids in the material. The formation of these oversized pores can be attributed to the calcination process in the muffle oven. During calcination, gases are released at high temperatures. These gases tend to rise upwards and escape from the material. Since the aggregates rest statically, the coarse pores collect preferentially in the upper region of the aggregate. In contrast, smaller pores are present in the lower support region of the aggregates. This could be due to the fact that, in this area, the aggregates are more compressed or have been compressed by the weight of the overlying layers.

In comparison, the pore distribution in the rotary kiln process is more uniform than in the muffle oven. This is due to the rotation of the aggregates in the rotary kiln over the entire length of the kiln. In the muffle oven, on the other hand, the granules lie statically, causing the bloating gases to rise to the top. Uneven pore size distribution and oversized pores can have a negative impact on the aggregate's stability. The rotary kiln process enables a more homogeneous pore distribution and thus improves the stability of the aggregates. However, for a mere assessment of the bloating effect, tests in the muffle oven are completely sufficient.

The suitability of iron sludge as a possible bloating agent could not be shown within the scope of this work; both the granulation and calcination results deteriorated.

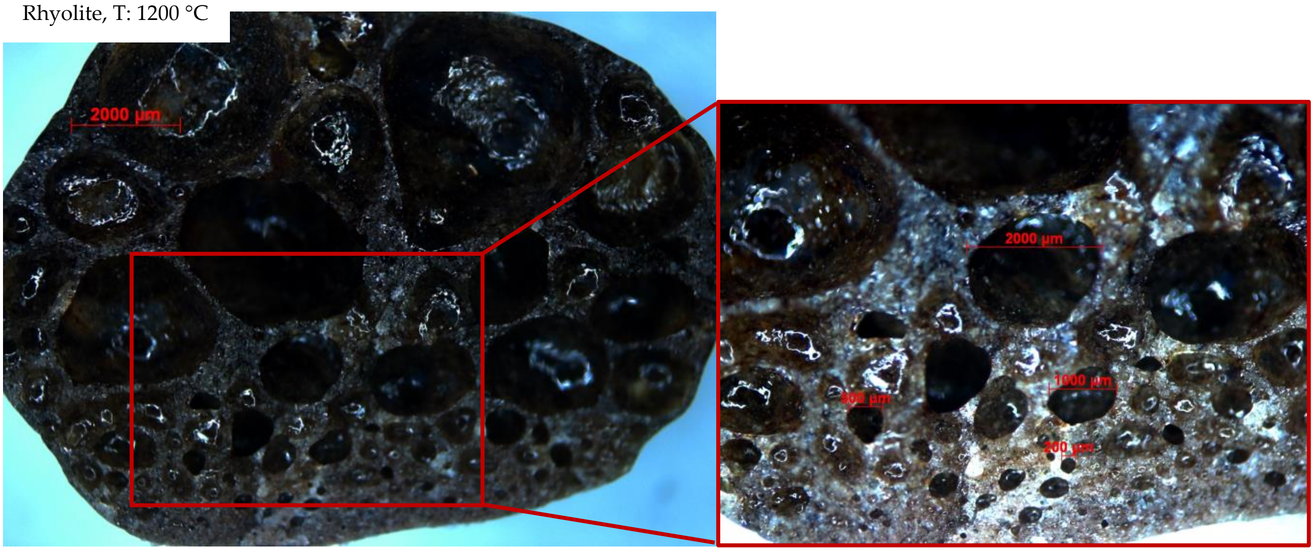

Figure 5. Rhyolite lightweight aggregate, calcinated at 1200 °C.

4. Discussion

Within the scope of the present study, a total of 35 residual and waste materials were examined for their suitability as lightweight aggregates. Of these 35 materials, 9 were classified as promising and subjected to a more detailed investigation. In the practical tests, both the fractions from rhyolite and from greywacke were able to demonstrate the best bloating properties. The bloating values achieved in the heating microscope were 1.4 of the initial aggregate, while in the tests in the high-temperature oven, bloating values of up to 1.9 could be achieved with densities of 0.9 g/cm^3 without the addition of bloating agents. Interestingly, the bloating properties of rhyolite and greywacke as pure materials were better than those of the reference samples based on crushed brick and concrete; these only achieved bloating values of 1.1 in the heating microscope and 1.5 in the oven.

Mixtures with various concentrations of additives were only successful to a limited extent. As a rule, these led to an excessively high flux content and a resulting melting of the aggregates. Despite the negative effect of the melting of the aggregates, a reduction in the firing temperature could be observed by the addition of iron sludge. This could be a promising approach for subsequent investigations to reduce the required kiln temperatures.

The tests also showed that the addition of fluxing agents in quantities of less than 10% by mass had only a minor effect on the overall performance of the aggregates. However, at higher additions of 10% or more, the test results deteriorated noticeably. Overall, the materials rhyolite and greywacke, as residues from primary raw material extraction, were identified as promising for use in the production of expanded granulate. The process-related presence of the fractions as already finely ground filter dust and crushed sand fractions has a positive effect on the preparation required for granulation. This means that corresponding comminution stages can be saved. However, improving the properties through additives is a complex task and requires further analysis.

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