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Modifying the properties of finely ground limestone by tumbling granulation

Oliver Macho[1], Maroš Eckert[1], Barbora Tomášová[1], Peter Peciar[1], Martin Ščasný[2], Roman Fekete[1], Marián Peciar[1]

Slovak University of Technology in Bratislava
Faculty of Mechanical Engineering, [1]Institute of Process Engineering, [2]Institute of Manufacturing Systems, Environmental Technology and Quality Management
e-mail: oliver.macho@stuba.sk, maros.eckert@stuba.sk, barbora.tomasova@stuba.sk, peter.peciar@stuba.sk, martin.scasny@stuba.sk, roman.fekete@stuba.sk, marian.peciar@stuba.sk

Abstract

Calcium carbonate in the form of finely ground limestone is a material that has found its application in a wide range of industries, in the chemical, rubber, agricultural, and paper industries, is used for desulfurization of boilers and other. In civil engineering, ground limestone is used for the production of building materials, plaster and mortar mixtures, as a filler in concrete mixtures, in road construction, and as an essential component of mastic asphalt. This paper deals with examining the modification of the properties of finely ground limestone by the tumbling agglomeration method. It has been shown that the components of concrete with a round grain have a positive effect on the pumping of concrete in comparison with an elongated grain or the rough surface of crushed stone. The experiments will be carried out on a granulation plate using a variety of granulation liquid. The agglomerates and their properties were compared with untreated finely ground limestone, with a focus on detecting changes in compressibility, density and particle size. The output of this paper is a description and graphical representation of the changes in the properties of ground limestone before and after the agglomeration process.

Key words: ground limestone, agglomeration, compressibility, flow properties

1 Introduction

Tumbling granulation is a process in which powder particles perform a rolling movement and due to the addition of a granulating liquid are bound into larger units. Products formed thusly are called agglomerates (granules). At the points of contact of the particles are formed liquid bridges that connect the individual powder particles into agglomerates. The number of agglomerates formed is lower than the number of primary particles of the powder.

By the agglomeration is achieved improved flow properties of powder materials, better dosing, reduced output blockages of hoppers, and not least, dust reduction. A granular material is a complex system which exhibits non-trivial transitions between the static, the quasi-static and the dynamical states. Indeed, an assembly of grains can behave like a solid or a fluid, according to the applied stress. In between solid and fluid granular states, very slow dynamics are observed. When a complete macroscopic characterization of a powder is needed, all these granular states have to be precisely analyzed [3]. To control and to optimize processing methods, these materials have to be precisely characterized. The characterization methods are related either to the properties of the grains or to the behavior of the powder bulk [6].

Ground limestone has applications in a wide range of industries. In civil engineering ground limestone is used primarily in the production of building materials, plaster and mortar mixtures, as a filler of concrete mixtures and as an additive to Portland cement. Portland cements with limestone show consistently stable properties. The presence of limestone decreases the porosity of the cement, therefore the diffusion coefficient of chloride ions is reduced, resulting in greater resistance to chloride corrosion. Ground limestone has a positive effect on improving adaptability, reduces water separation, and stabilizes the color of the concrete. This paper deals with modifications of the properties of ground limestone by tumbling agglomeration on a granulating disc, using both distilled and utility water.

2 Experimental material

As an experimental material was used very finely ground limestone (Calmit) pursuant to STN EN 13 043. The manufacturer indicates the density of the material at 2700 kg/m^3 and the content of carbonate at more than 95 %. The relative humidity of the incoming material was 0.84 %. The characteristic dimensions of particles (Table 1) and their distribution (Figure 1) were analyzed by a Morphologi G3 microscope (Malvern Instruments), permitting the measurement of particle size on the basis of static image analysis. Evaluation of particle size was based on their volume, and a series of measurements of three equal sized samples (13 mm^3) was performed.

Table 1: Characteristic dimensions of experimental material

Dimension	**Sample 1**	**Sample 2**	**Sample 3**	**Mean value**	**Standard deviation**
Dv10 (μm)	9.6	9.5	9.6	9.6	0.06
Dv50 (μm)	26.4	25.2	24.2	25.2	1.10
Dv90 (μm)	92.2	97.6	94.7	94.8	2.71

Image analysis captures a two-dimensional image of particles, from which the various parameters of particle size and shape are then calculated. CE (Circle equivalent) Diameter is the diameter of a circle with the same area as the 2D image of the particle. D_{v10} is the diameter at which 10 % of a sample's volume is comprised of smaller particles, as with Dv_{50} and Dv_{90}.

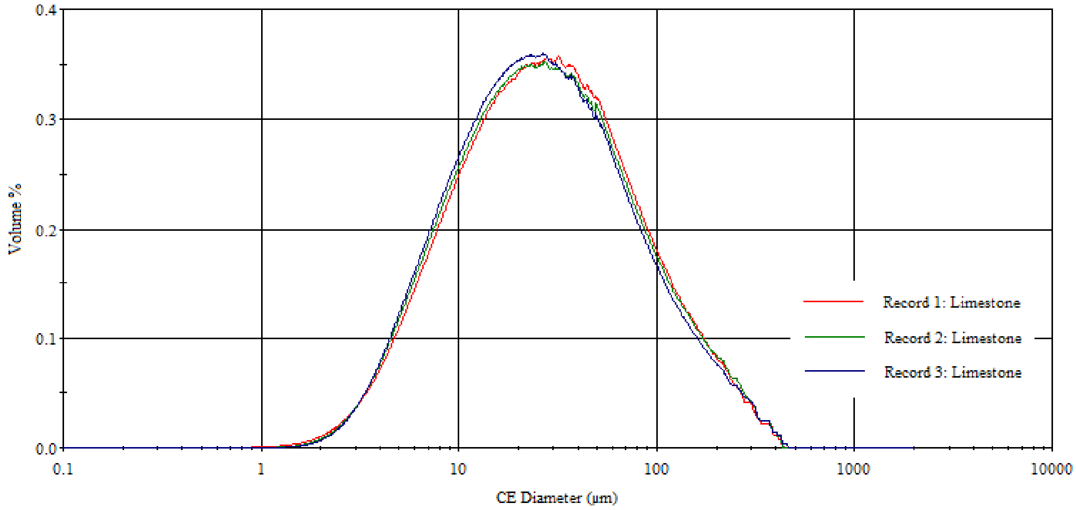


Figure 1: Particle size distribution of the experimental material

3 Tumbling granulation

The granulation of finely ground limestone was carried out on a tumbling granulation plate. The diameter of the granulation plate was 400 mm, the height of the plate 70 mm, and rotation speed, 480 rpm. The granulating disc was inclined at 45° to the vertical axis. Distilled water (DW) was used as the granulating liquid in the first series of experiments, and utility water (UW) in the second series. The relative mass concentration of the granulating liquid to the incoming material was 3 % and 10 %. The agglomerates are produced by a batch process, with a total of 900 grams of product produced for each sample. The duration of the granulation process was 3 min. The agglomerates were dried in the open air at laboratory 22 °C, while changes in the properties of the product according to the times of hardening were also investigated.

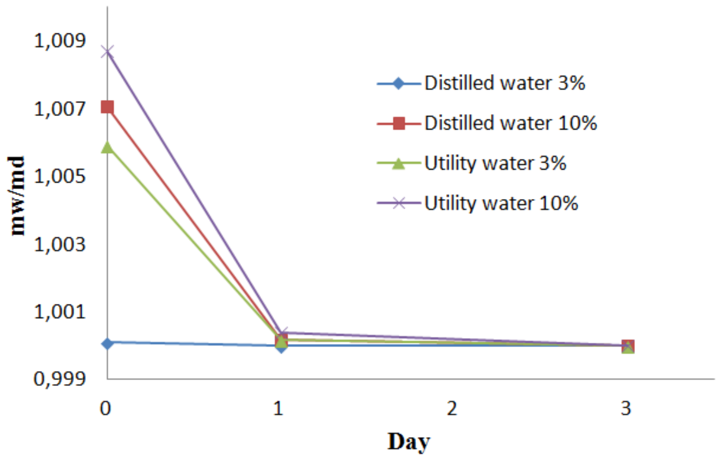


Figure 2: Measurement of the moisture of agglomerates: mw – weight of wet material, md – weight of dry material

Agglomerate humidity is expressed as the weight ratio of wet (mw) and dry (md) materials (Figure 2). After three days from the production of the agglomerates the humidity did not change and the agglomerates' properties changed only by the hardening the agglomerates.

4 Granule size distribution

Granule size distribution (GSD) was determined by an image analyzer of dry particles, PartAn 3D (Microtrac). The particles falling from the vibrating hoppers were recorded using a high-speed camera that captured images of the falling particles. Based on the series of images an overall 3D picture of the individual agglomerates was created. For comparison with the primary particles a cumulative distribution curve of granule size (Figure 3) was made on the basis of their volume. The characteristic sizes of the agglomerates are found in Table 2.

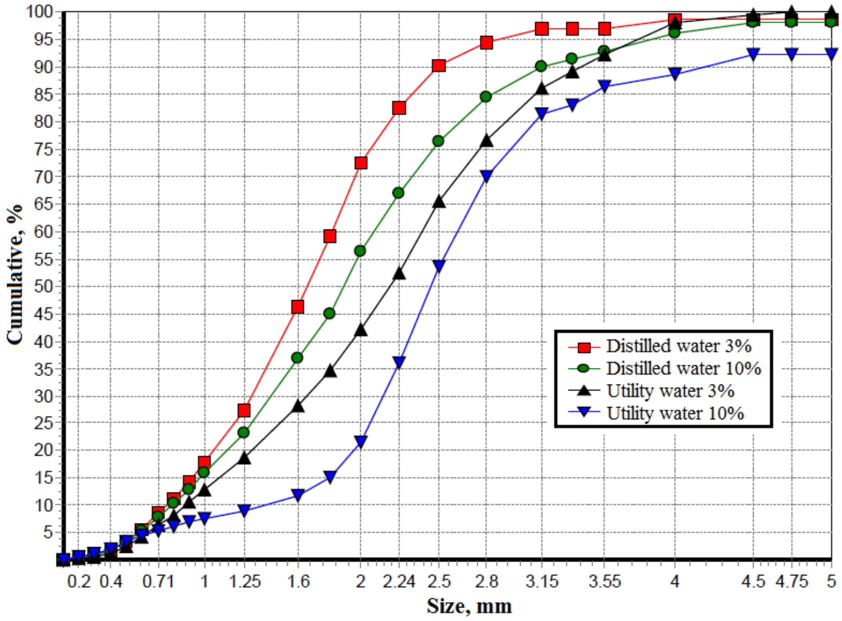


Figure 3: Distribution curves of agglomerates

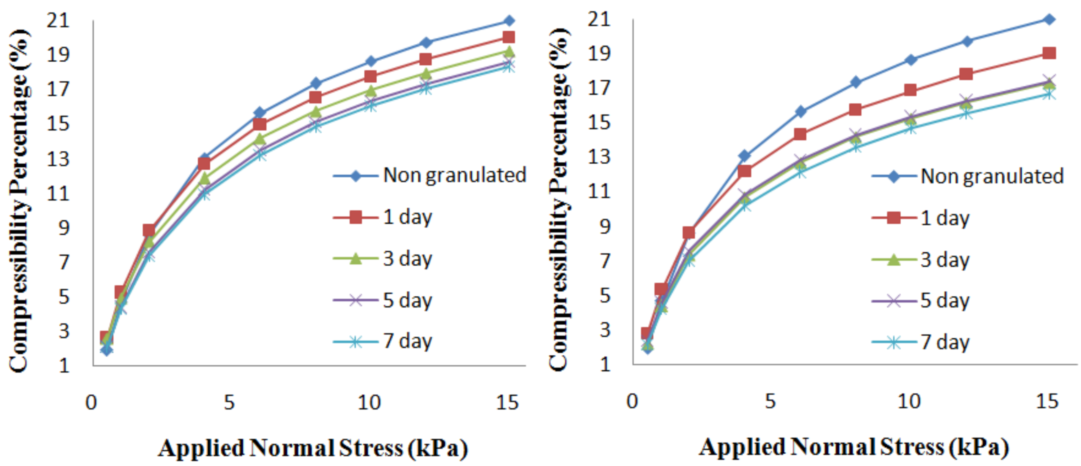
Table 2: Characteristic dimensions of granules

Dimension	DW 3 %	DW 10 %	UW 3 %	UW 10 %
Dv50 (mm)	1.66	1.9	2.19	2.43

5 The measurement of compressibility

The compressibility and compactibility of a powder are influenced by the flow properties and, in the microscale, by the adhesion forces between particles. Compressibility is the ability to reduce the volume under pressure [2]. Measuring of compressibility and density were performed on a Freeman FT4 (Freeman Technology) Rheometer of powder materials (Figure 9). The FT4 Powder Rheometer is designed to characterize powders under various conditions in ways that resemble large-scale production environments [8].

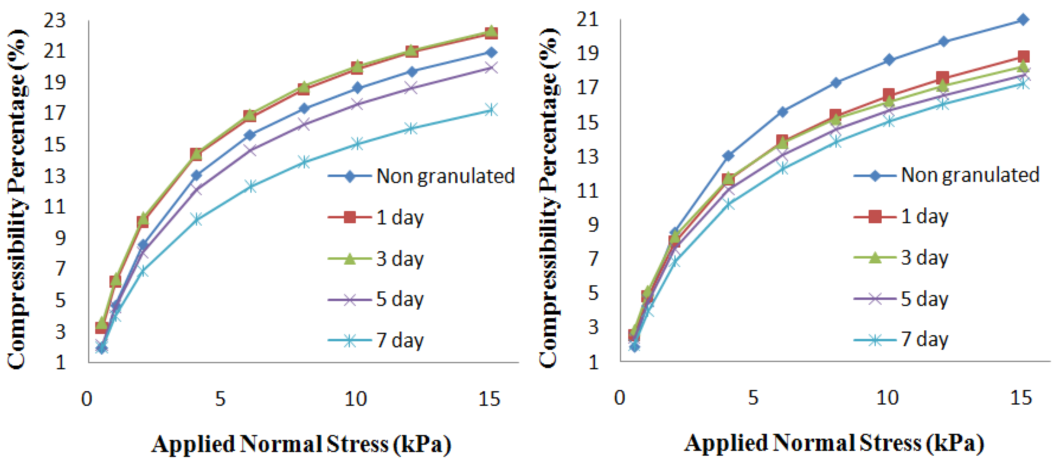
Individual samples were pre-prepared in a step called conditioning. It is essential to produce a standardized packing condition as a preliminary to each test cycle. Conditioning involves gentle displacement of the whole powder sample in order to loosen and slightly aerate the powder. This process removes any packaging history such as pre-consolidation or excess air [1]. The sample composed of the experimental material and the formed granules was placed in a system of two concentric cylinders with a diameter of 50 mm and a volume of 85 ml. A blade with a diameter of 48 mm was used to advance the preparation of the sample. In the actual measurement of the compressibility the blade was replaced with a vented piston, which applied to the sample normal stress in a range from 0.5 to 15 kPa. Series of five measurements were carried out, into account being taken mean value. Compressibility deviation was not greater than two percent (Figures 4, 5).



a) b)

Figure 4: Compressibility of granules depending on normal stress:

a) distilled water 3 % mass; b) distilled water 10 % mass



a) b)

Figure 5: Compressibility of granules depending on normal stress:

a) utility water 3 % mass; b) utility water 10 % mass

A series of measurements was conducted with the goal of finding the changes in compressibility and bulk density over time. The measurements were carried out after the first, third, fifth and seventh day following the production of the agglomerates. Compressibility equals the percentage change in the volume of the material brought about by compression. Due to the action of normal stress, all the examined samples showed the features of very cohesive powders.

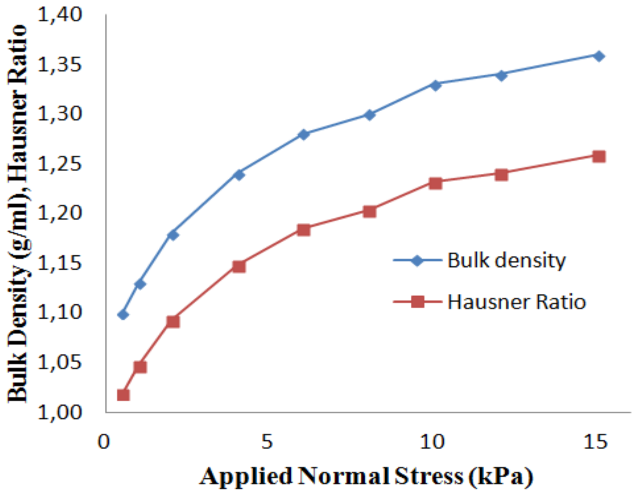
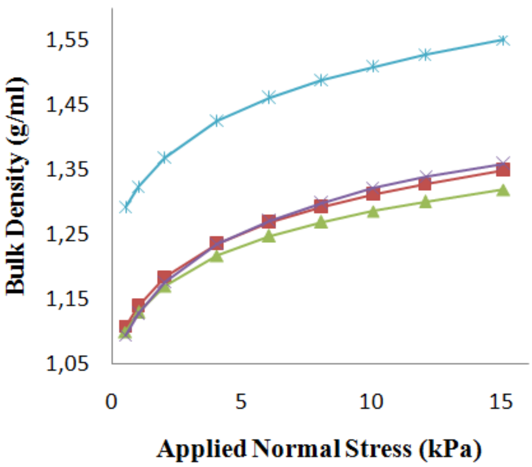
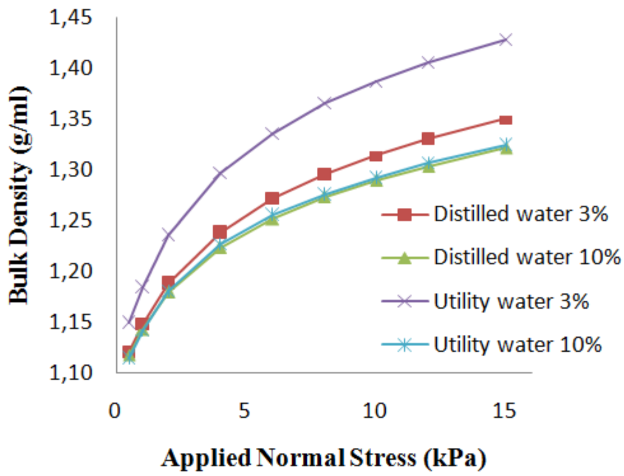


Figure 6: Bulk density and Hausner Ratio of non granulated limestone depending on normal stress



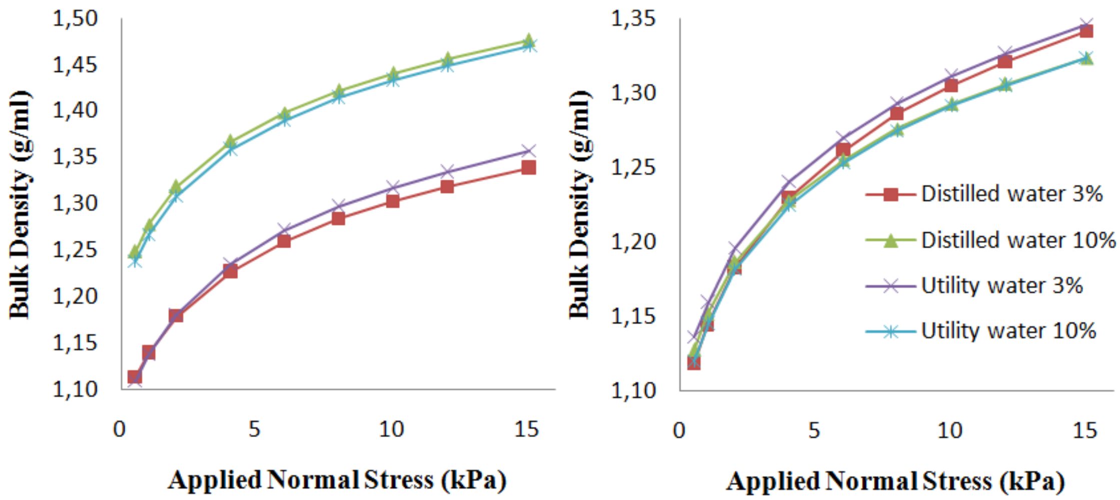
a)



b)

Figure 7: Changes of bulk density depending on normal stress:

a) after 1 day; b) after 3 days



a) b)

Figure 8: Changes of bulk density depending on normal stress:

a) after 5 days; b) after 7 days

Compressibility of powder materials can be described by the Hauser ratio (1), which gives to the ratio the bulk densities of the compressed and uncompressed material.

$$HR = \frac{\rho_{TAP}}{\rho_{BULK}} \tag{1}$$

In our case we customized the relationship to a modified form

$$HR_m = \frac{\rho_{under\ stress}}{\rho_{CBD}} \tag{2}$$

where represents bulk density under loading of normal stress, and the bulk density of the sample measured after the conditioning cycle. Greater Hausner ratio values mean higher strength in the examined material. Hausner ratio is also an indicator of flow properties of powder materials (Table 3). Several authors deals with the problems of the flow properties of powders [4,5,7].

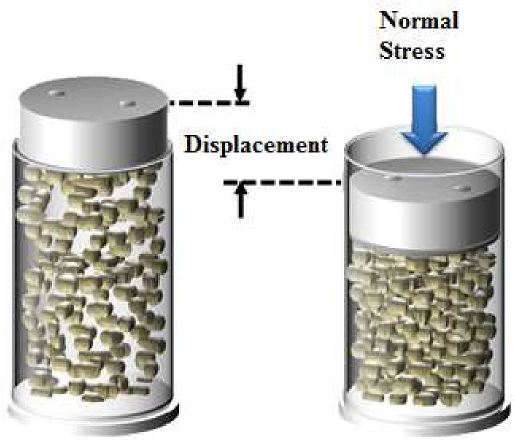


Figure 9: Measurement of compressibility

Table 3: Scale of flowability by Hausner ratio

Flow property	**HR** (1)
Excellent	1,00 - 1,11
Good	1,12 - 1,18
Fair	1,19 - 1,25
Passable	1,26 - 1,34
Poor	1,35 - 1,45
Very poor	1,46 - 1,59
Very, very poor	>1,60

6 Results and discussion

Evaluation of the experiments was designed to determine the distribution characteristics of the agglomerates and the changes in compressibility and flow characteristics depending on the duration of hardening time. The distribution curves of the agglomerates produced by distilled water had greater monodispersal fractional composition than the agglomerates produced by utility water. The particle size medians of the granules produced by the utility water were higher than those with the distilled water. Larger quantities of liquid caused more significant bundling of the powder material, and for this reason the agglomerates with a 10 % concentration of granulation liquid were larger. Compressibility of the agglomerates made from distilled water and from utility water achieved approximately the same values. The granules formed with a 3 % concentration of granulating liquid showed higher compressibility values than with a 10 % concentration.

The wetter agglomerates had lower strength but the hardening time of the granules had a more significant influence on the strength of the agglomerates, which was reflected in the increased values of bulk density found (Figures 7, 8). Over time, the liquid bridges connecting the individual particles changed to bridges of a solid character that eventually gained higher strength. The impact of the duration of hardening is also visible on the values of Hausner ratios (Figures 10, 11), which had a decreasing trend.

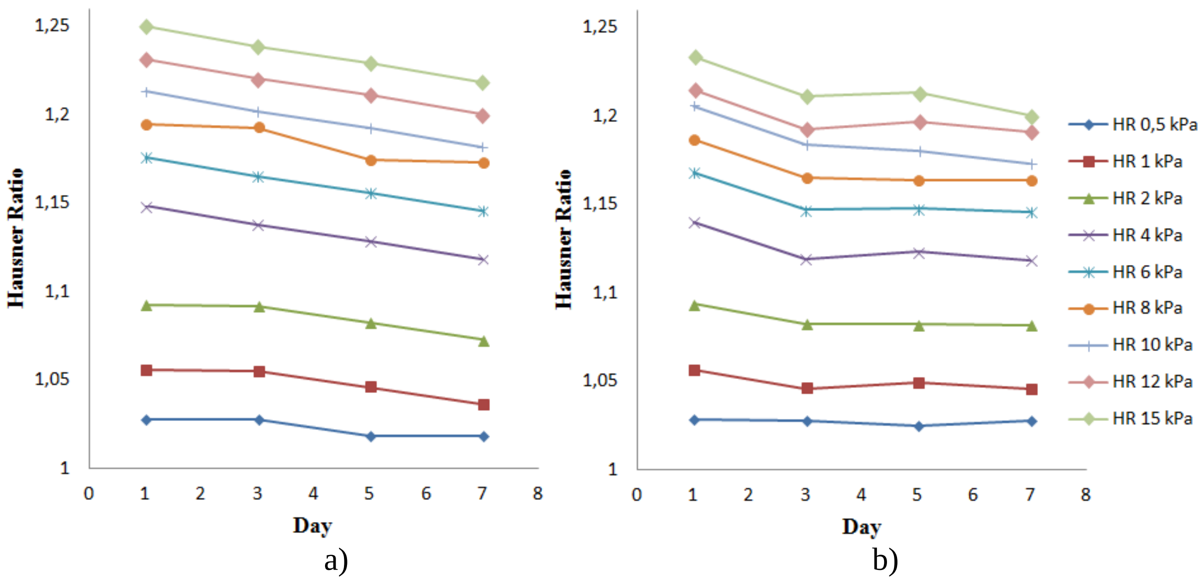
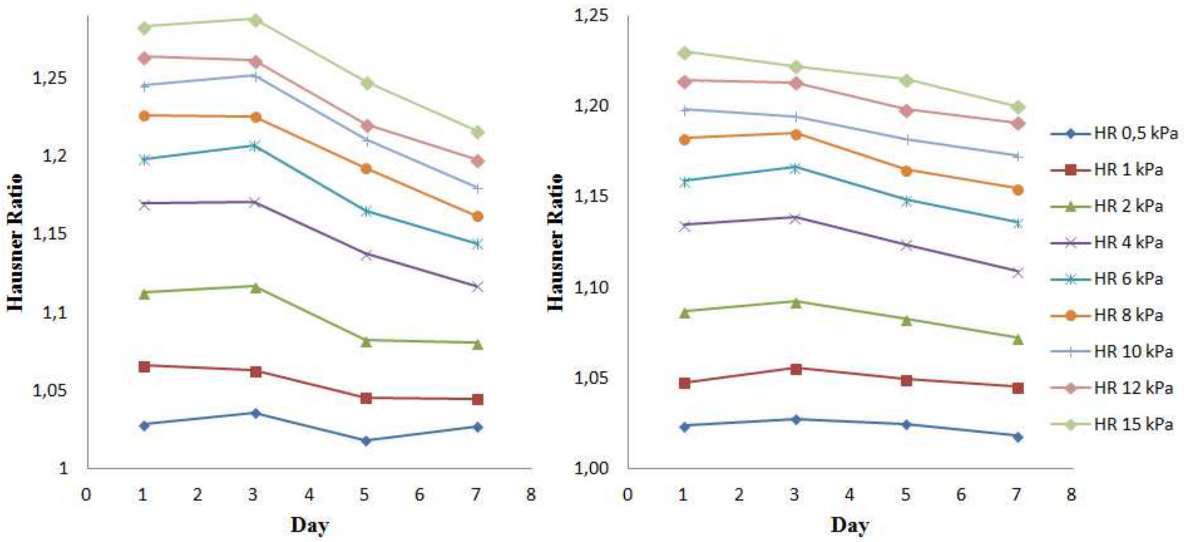


Figure 10: Changes of Hausner Ratio over time:

a) distilled water 3 % mass; b) distilled water 10 % mass



a) b)

Figure 11: Changes of Hausner Ratio over time:

a) utility water 3 % mass; b) utility water 10 % mass

Hausner ratio values for the produced agglomerates after stressing to a normal load of 2 kPa showed excellent flow properties (Hausner Ratio < 1.11). The agglomeration process did not reduce the flowability of the limestone, but rather eliminated the dustiness of the product, which showed to be well granulated by tumbling agglomeration. Dust reduction of the raw material resulted in a limitation of the loss of the dust fractions of the processed raw materials, whether during dosing, transport or further processing.

7 Conclusion

Measuring the compressibility of agglomerates of finely ground limestone created by using distilled and utility water did not show significant differences in the values of percentual compressibility. Agglomerates of limestone and utility water however had larger characteristic dimensions. The experiments have shown the impact of duration time on the hardening of the created agglomerates. Increased strength was visible in the created agglomerates' values of percentual compressibility, bulk density and Hausner ratios. In subsequent experiments it will be possible to track changes in the compressibility of the agglomerates over longer durations of hardening. The tumbling agglomeration process has proven to be a suitable process for reducing the dustiness of ground limestone without affecting its flowability while on the other hand preventing losses of dust fractions.

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