AN ALTERNATIVE ALUMINO-SILICATE BINDER BASED ON THE "JUST ADD WATER METHOD"

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ABSTRACT

Alumino silicate compounds are important for alternative binders for concretes because of the reduction of CO_2 and resource conservation. Such multi-phase systems consist of a solid component, such as e.g. slag, ash, calcined clays and others, and a liquid component, in generally highly concentrated alkaline solutions (water glass or NaOH/KOH). Because of safety and other reasons, such systems exclude large-scale practical applications.

The newly developed alternative binder consists of a highly soluble silicate waste material, which is not common in geopolymere production, and a powdered Al-provider. It is called alumino silicate network (ASN) binder. Both components can be mixed dry in different ratios. Only after addition of water, a polymerization process is induced and the formation of amorphous phases, and sometimes zeolites can be observed. Mortar bars were produced and tested for different properties. The best results regarding to workability, strength development and sustainability were obtained with a molar Si/Al ratio of 2:1 in combination with fine-grained rock filler components. Investigations on the hardening process showed, that the mechanical properties varied over a wide range. Strength reserves, a ratio between compressive and tensile strength of approx. 3 and so far good resistance against acid solutions allow the use as a binder in high performance mortar and concrete structures.

Keywords: alternative binders, geopolymere, alumino silicate structures, Si/Al ratio, industrial residues

INTRODUCTION

Geopolymere binders can substitute OPC or other cementitious binders on special fields of applications. Many papers have been published on this subject in the last years [1,2,3,4,5]. However, some problems still need to be resolved, regarding to the production of such materials. More than 95% of the papers deal with the common materials such as metakaolin, slag and fly ashes [6]. An important approach is the selection of the activator [7]. In general, highly alkaline solutions (NaOH, KOH, water glass solutions) with a concentration of 8 to 10 Mol/L OH are used. Such solutions are certainly applicable without problems in the laboratory scale, but they present a considerable safety risk for the practical application [8].

Therefore, research is looking for better solutions, so called one-part geopolymers [9,10,11]. Such systems can be realized by using a Si source and sodium aluminate. Another problem is the availability of the solid materials. Blast furnace slag and metakaolin are very suitable for this, but slag is primarily being used by the cement industry to produce e.g. CEM III (EN 196) and is thus not available in large quantities. Metakaolin has to be produced in special way by burning of clays. There are numerous publications describing the mixing process, the production of test specimens and their mechanical properties [12,13]. Also recently, the use of combinations of metakaolin and slags, activated by water glass solutions has been described in the literature [14].

Papers, which deal with the formation of geopolymere structures or aluminosilicate-networks describe the mechanism as a dissolution process, what means that structures from the starting material (slag, metakaolin) have to be dissolved. At the same time, under highly alkaline conditions, aluminosilicate structures must be formed to build a network [15,16,17,18]. Based on this principle, in this work industrial residues were investigated, which can separately supply the silicon and aluminate component for the production of alumino-silicate binders. This approach is not new, as several researchers have done this before [19,20,21,22,23,24].

The new developed alternative binder consists of a highly soluble silicate waste material, which is not common in geopolymere production, and a powdered Al-provider. It is absolutely necessary to find a siliceous material which has a high solubility in alkaline solutions in a pH-range up to 14 (what means an OH-concentration of approx. 1 Mol/L). It can be reached by addition of sodium aluminate and water in different ratios to the silicate component. By reaction of both materials an aluminosilicate network (ASN) is formed, which is able to cure.

Using the commonly available materials (slag, fly ashes and metakaolin) the solubility of the starting material and the formation process of the cured aluminosilicate products cannot be differentiated. The two sub processes occur at the same time in the complicated systems. Maybe the knowledge of the dissolution behavior is not so important, because both reaction partners are jointly present [25]. For the system examined here, this is not the case, so that the dissolution behavior of Si and Al must be coordinated. This makes very high demands on the mixing and homogenizing procedure because Si and Al must be distributed very evenly into the smallest grain sizes of the starting materials. If that succeeds the “just add water” principle to produce aluminosilicate binders should be possible.

RESEARCH SIGNIFICANCE

Different solid materials and activator solutions have been investigated during the past few years. Most of the studies deal with the activation of common slags, ashes and metakaolin. These materials, which have to be balanced out, are too expensive or show fluctuations in their properties. Worldwide the search for creating the “just add water” principle is going on. The authors believe that with this study another step has been taken toward the development of cement free alternative binders for special uses but without alkaline activator solutions with concentrations of approx. 8-10 Mol/L.

EXPERIMENTAL INVESTIGATION

For the investigations, different Si-containing residues (Si 1-7) were used and different Al carriers were tried. At long last as an Al provider sodium aluminate ($NaAl_2O_4$) was selected. All materials used are in powder form. Various molar Si/Al-ratios were chosen and additionally the water content was adapted, thus, the concentration of the alkaline solution could be changed. Additionally a special manufacturing process had to be developed. Because of the special properties of some Si materials, the water must be added to the Si carrier, so that a slurry is formed which has to be added to the remaining dry components then. The mixing procedure occurs at first in a closed mixer, called Rhoen wheel mixer, and later with a stirrer. Fresh mortar properties and mechanical properties at mortar bars with a size of 1.57*1.57*6.30 in^3 (40*40*160 mm^3) were investigated. Also the durability against acids and alkaline solutions over a long period of time was determined.

Materials

In preliminary examinations different siliceous materials were tested, shown in **Table 1**. Si-1 to Si-4 are common products such as microsilica and silicon dusts from various manufacturers. Si-5 and Si-6 are waste products from the quartz glass industry, Si-7 comes from a zeolite producer. They differ in their surface area and in their content of SiO_2. Nearly all materials except Si-7 have a Si content of more than 80 %, while Si-5 and 6 consist of almost 100 wt.-% of amorphous silica. The decisive factor, however, is not the total content of SiO_2, but rather the solubility or better the solubility rate in the alkaline solution under defined conditions. **Figure 1** shows the solubility of the starting materials in 1 Mol/L NaOH solution at 40 °C. It is obvious, that the solubility rates are different, Si-5 and 6 have the largest rates, already over 90% are solved after 2 days. On the other hand, an aluminum source is needed, which contains enough soluble aluminum at the same period of time. At this point it must be inserted, that the influence of the solubility rate of siliceous materials by aluminum which is present in the solution simultaneously is not taken into account. Basic relations still need to be clarified.

At first, two waste products were investigated (see Table 1, Al-1 and Al-2), but these were not found to be suitable since the solubility rates were very low. 17 wt.-% Al_2O_3 determined is too less for use (**Fig. 2**). Finally, a pure aluminum source in the form of sodium aluminate ($NaAl_2O_4$) was selected. Sodium aluminate shows a very fast dissolution behavior of aluminum in water, comparable to Si-5 in an alkaline solution. Results are shown in **Fig. 3**. After two days more than 90 wt.-% aluminum are soluble, using a solid/solution-ratio of 1/100. The dissolution experiments with sodium aluminate were realized additionally with different solid/solution ratios (1/100, 1/10 and 1/2.2). A pH-value of approx. 14 was established for the solution with a ratio of 1/10 and approximately a little bit more than 80 % of the aluminum was in the solution. The ratio 1/2.2 corresponds to the ratio in the recipes, in this case a maximum of approx. 48 % Al_2O_3 dissolves after two to three hours. Obviously the rest precipitates as amorphous aluminum hydroxide with increasing storage time, but is still available for alumino silicate formation reactions. Based on the dissolution experiments all investigations were done using sodium aluminate as the aluminate source in combination with different Si sources.

For the mortar formulation conventional quartz sand with a grain size of 0-2 mm was used.

Specimens

Mortar prisms with a size of 1.57*1.57*6.30 in^3 (40*40*160 mm^3) were produced for strength testing. One approach consisted of 3 samples with these dimensions. **Table 2** shows the mixture proportions used for the first sample production. There were no further studies made with Si-2, Si-4 and Si-7, because preliminary investigations showed, that no hardening (Si-7), or even strong agglomeration formation were observed (Si-2, Si-4).

Parameters investigated

In the first step, the Si-carrier was prepared with sodium aluminate and water. These investigations gave a first overview of the hardening potential of the binder. In the second step, mortar prisms were produced according to EN 196-1 [26], using the Si materials Si-1, Si-3, Si-5 and Si-6. After demolding, the flexural basic vibration, using the Impulse Excitation Technique (GRINDOSONIC method), was determined continuously for 28 days to calculate the dynamic modulus of elasticity, and then the flexural and compressive strength were tested. Furthermore, large-scale tests were carried out with the best Si-material (Si-5). For this purpose the mixture proportions were adapted again. The mortar prisms were tested for strength, shrinkage and stability in the acidic and alkaline environment. For the durability tests in a wide range of pH values, the mortar prisms were shared and parts were placed into the solution pH 0, pH 2, pH 4, pH 12, pH 14 and water-solution respectively. Additionally the durability against sulphate acid was also tested in comparison to a cement prism. For this purpose, whole prisms were taken.

EXPERIMENTAL RESULTS AND DISCUSSION

Development of the binder in the lab

In a first step, preliminary investigations gave an overview on the hardening behavior of the pure binder systems. The approaches showed that not all Si-materials are suitable for binder production. Some of the mixtures, which were prepared with the different Si sources and sodium aluminate plus water without any sand components, did not harden or the hardening process took too long. **Figure 4** demonstrates the hardening process of material Si-5 in reaction with sodium aluminate and water. From left to right the Si/Al ratio increases.

The basic for first mortar bar investigations was the standard EN 196-1. According to this, mortars were produced with one part binder, three part sand and half part water. However in this case the binder consists of two parts, a silicium and an aluminum part in different ratios. The aluminate binder component and the sand component have been added very carefully to the siliceous part of the binder. In this stage of investigation, the mixing process occurs in a special laboratory mixer. The fact, that especially the materials Si-5 and Si-6 too have a large surface and therefore a very small grain size, made problems when mixing. **Table 2** gives a compilation of the Si carriers selected, of the demolding times, the water/binder ratios and of first strength results.

Especially mortar bars produced with Si-5 and a Si/Al ratio of 2/1 gave very good results. After 1 to 2 days, the prisms could be demolded. With a compressive strength of more than 60 MPa (N/mm^2) the structure formation process of both reaction components obviously leads to a very good linked network of Si and Al tetrahedra after 28 days.

Development of a manufacturing process

The conclusions based on the first investigation were, that some changes in the mixing technology were needed. For the mixing process a so called “Rhoen wheel mixer” was used, which can be seen in **Fig. 5**. This chosen technology realized, that the siliceous dust material remains in the mixer. Important is, that the use of such a mixer allows to increase the intensity of the mixing process, a necessary advance for the production of structural good formed samples. Additionally the amount of mortar material could be increased to produce mortar and concrete cubes. And in all cases a slurry technology was applied now.

For all subsequent investigations material Si-5 was selected. Si-5 has a large surface and the mixing process of the dry components Si source, sodium aluminate and sand made a lot of problems. To solve them it was necessary to produce a slurry by water addition to the Si-5 material. This water is considered in the production of the mortar prisms. Both the intensive mixing procedure by using the “Rhoen wheel mixer” and the creation of a Si containing slurry allows to produce mortar bars of a good quality.

The developed mixture proportions R1 to R4 are summarized in **Table 3**. The mixtures were realized with different Si/Al ratios and with a different water amount depending on the workability. Sometimes the addition of a certain amount of a special superplasticizer was necessary.

Structural investigation

Typical mortar samples are shown in **Fig. 6**. It can be seen: on the left side the structure on the top, on the right side the structure on the bottom (opposite to the top). The samples R1 show cracks on the surfaces and inside a very porous structure, which indicates that the structure formation process is not optimal under the chosen conditions. Better results can be obtained with R2 and R3. A closed surface and a homogenous structure inside are characteristics of these samples. R4 is a retry of R1 but with less water. The aim was to investigate the influence of the reduced water amount, which should increase the pH-value of the pore solution. However changes in this direction lead to many problems in the workability, so that the samples were very porous (see the last bar on the right side).

Strength and Dynamic modulus of elasticity

These samples were used to determine mechanical properties. **Figure 7** shows results of the compressive strength development until 8 month. How expected samples R2 and R3 reach the highest values. The compressive strength with approx. 20 to 30 MPa (N/mm^2) after 28 days is three to five times higher than samples R1 and R4. While sample R4 loses strength over time (because of the “false” Si/Al-ratio), the strength of R3 increases significantly for up to 8 months of storage. After 8 month a strength value of 50 MPa (N/mm^2) was reached.

The modulus of elasticity values (see **Fig. 8**) make the differences between the formulations even more visible. With 22,000 MPa to 27,000 MPa (N/mm^2) the values are five to six times higher than values of mixture R4. Because of the fact, that the project is worked in cooperation with a company in Germany, it is possible and it makes sense to compare the elasticity results with a reference sample. The reference is a normal mortar on the basic of limestone and cement, optimized for using as a plastering or masonry one. And this comparison also shows that ASN mortars have approx. a two and half higher elasticity than the normal mortar of the company.

Such high values in strength and modulus of elasticity allow to have a large reserve for special fields of application.

Swelling and shrinkage

Another important value is the swelling and shrinkage behavior under wet or dry conditions. **Figure 9** gives the results of the shrinkage behavior. Especially the results of sample R3 lay in a normal range of mortars also in comparison to the reference mortar material.

Durability in acid and alkaline solutions

An important part of the performed investigations was the behavior of ASN samples against acid and alkali attack. Mortar bars with the best mixture proportions (R2 or R3), but with a modified water content were produced. For these investigations, Si-5 was used as the Si source again. There are some reasons for this selection. During the investigations over a long period of time, Si-5 as an industrial waste product was always available in the same quality. The manufacturing technology could be tuned to this material and in all cases, it worked well. The mechanical properties were almost always very satisfactory.

That`s why the focus of the further investigations was on the search for special applications. A possible field can be the application as a repair mortar or a coating material in the sewage sector (see **Table 4**).

In a first series, the so produced samples with a curing age of 28 days were stored in different solutions, as it can be seen in **Fig. 10**. The pH-value was varied between 0 and 14, what means that the whole range of damaging solutions was selected. Samples in the bottles demonstrate the results after 6 month storage and no significant changes in the structural formation could be observed. In this case the chosen acid was HCl for the pH range between 0 and 7, and NaOH for the pH range until 14. During the storage, the acid solution was changed every 1-3 days to guarantee the strong acid environment.

A more practicable case is the attack of H_2SO_4, because of the combined damage by acid and sulphate ions. Such investigations were done in comparison to the similar attack on a cement prism. A pH value of 2 was chosen. After 28 days of storage (**Fig. 11a)** the cement prism shows significant changes in the surface structure. It was very easy to scratch the surface of the mortar material as it can be seen on the top of the left prism. While the cement prism indicates damages, the ASN mortar prism does not show any significant changes on the surfaces. The result after 72 days storage is even more impressive (**Fig. 11b**). The cement prism shows a clear deterioration behavior, after scratching on the surface the mass lost is very high and the surface can be characterized as a relief-like area. The behavior of the ASN prism is different from this. On the surface no weathering behavior and no significant changes could be observed, the structure remains dense and firm. However the compressive strength decreases, which is normal. In the meantime several prisms were more than 6 month in an acid solution with pH 2, but no changes could be observed.

FURTHER RESEARCH

It is desirable to test specimens over a long period of time (1 or more years of exposure) to confirm the good stability of the new developed mortar against any kind of attack (also ASR). Of course, the look for an aluminum provider, which should be a waste product indeed, has to be continued. And the third field of investigations is to improve the mixing technology in a direction, which allows producing a dry mortar consisting of the components Si source, Al provider and aggregate grains together. At this stage of investigations the production of a slurry with the Si material, in which the other components have to be distributed, is the best way to homogenize the mortar. However it must be said, that the “just add water principle” could be realized already.

CONCLUSIONS

Based on the results of this experimental investigation, the following conclusions are drawn:

1. Siliceous containing waste materials from the quartz glass industry with a high solubility of more than 95 wt.-% after 1-3 days storage in an alkaline solution are very suitable for the production of aluminosilicate network (ASN) binders.
2. Based on dissolution experiments a molar Si/Al ratio of 2/1 should be preferred, which means that the Al provider must have an analogous solubility matched to the Si carrier.
3. The manufacturing process is characterized by an intensive mixing procedure to realize that all components are homogenously distributed in the mortar. Only water has to be added to this system.
4. If this can be guaranteed in all cases, the “just add water principle” can be realized, and a very durable mortar as a repair or coating material in a sewage sector can be produced.

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TABLES AND FIGURES

Table 1–*Overview about the Si- and Al-materials*

Material	**Content SiO_2 resp. Al_2O_3** wt.-%	**Density** lbm/in³ (g/cm^3)	**Specific surface (BET)** in²/lbm (m^2/g)	**Water content** (wt.-%)
Si-1 (MS 1)	> 90	0.0864 (2.33)	12329521 (17.5)	< 1
Si-2 (MS 2)	> 90	0.0841 (2.27)	13738609 (19.5)	< 1
Si-3 (MS Sika)	> 85	0,0827 (2.23)	13879518 (19.7)	< 1
Si-4 (VA-Silica)	> 90	0,0816 (2.20)	25363586 (36.0)	< 1
Si-5 (Residue)	99.99	0,0812 (2.19)	21418139 (30.4)	< 1
Si-6 (Residue)	99.99	0,0805 (2.17)	35086294 (49.8)	< 1
Si-7 (Silica mud)	> 70	0,0831 (2.24)	243701785 (345.9)	80.0
Al-1 (Serox 1)	59 – 66	0,1001 (2.70)	-	15.0
Al-2 (Serox 2)	21	-	-	28.0
Al-3 (Red mud)	53	0,0949 (2.56)	732726 (1.04)	-

MS: Microsilica from different providers

Table 2–*Mixture Proportions for first investigations*

mixtures with Si	**Si/Al** (mol/mol)	**Demolding** (d)	**H_2O/Na_2O** (mol/mol)	**water/ binder ratio** -	**Tensile Strength** MPa (N/mm^2)	**Compressive Strength** MPa (N/mm^2)	**Dyn. Mod. E** MPa (N/mm^2)
Si-1	1:1	11	8	0.59	2.17	4.52	7696
	2:1	4	8	0.42	5.87	23.09	24091
	3:1	4	8	0.33	1.34	14.53	5772
Si-3	1:1	7	8	0.57	1.35	3.49	7509
	2:1	5	8	0.40	0.67	6.91	2796
	3:1	4	8	0.31	0.38	6.13	1754
Si-5	1:1	2	8	0.61	1.82	9.19	20921
	2:1	1	8	0.44	5.87	61.97	9647
	3:1	1	8	0.34	0.64	7.55	6919
Si-6	1:1	2	12	0.91	1.53	7.29	17690
	2:1	1	12	0.66	7.72	45.20	8174
	3:1	1	12	0.52	4.48	11.66	13037

w/b: Water/Binder-ratio
Sand: grain size 0-0.79 in (0-2 mm), convent. quartz sand, 3 times the amount of the binder (EN 196-1)
Bending tensile strength (EN 196)
Compressive Strength (EN 196)
Dyn. Mod. E: Dynamic Modulus of Elasticity (DIN 1048)

Table 3–*Formulations produced with the modified technology (the ratio aggregate/binder was always 3/1)*

Recipe No.	**Si/Al ratio**	**H_2O/Na_2O ratio**	**Fraction**	**Superplasticizer**
	mol/mol	*mol/mol*	*inch (mm)*	*wt.-%*
R1	1:1	8	0-0.08 (0-2)	**-**
R2	2:1	9.5	0-0.08 (0-2)	0.77
R3	2:1	11	0-0.08 (0-2)	0.30
R4	1:1	7	0-0.08 (0-2)	-

Table 4–*Mortar formulation for durability investigation*

Component	**Si/Al ratio**
Si source	Si-5
Si/Al ratio	2:1
H_2O/Na_2O ratio	11
Aggr./binder ratio	3:1
water/binder ratio	0.6

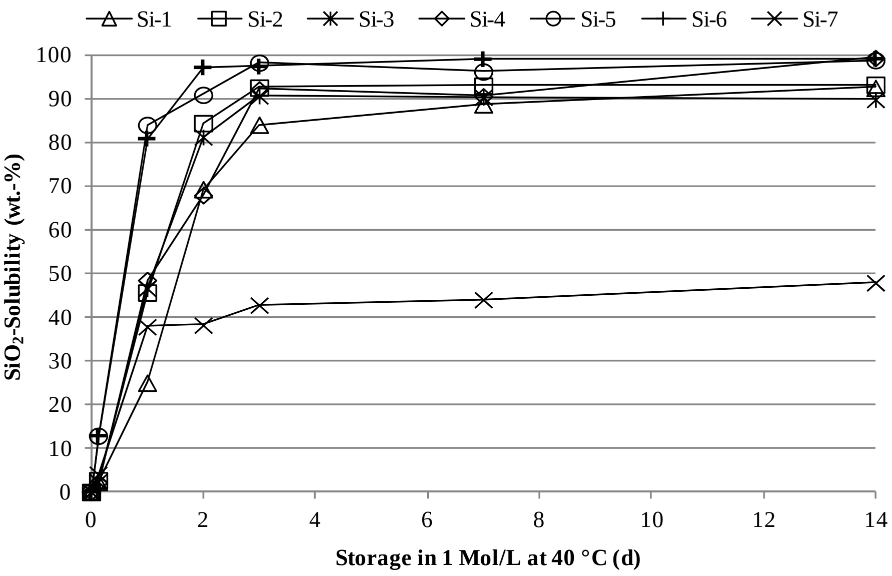

Fig. 1*–Solubility of the starting materials in 1 Mol/L NaOH at 40°C*

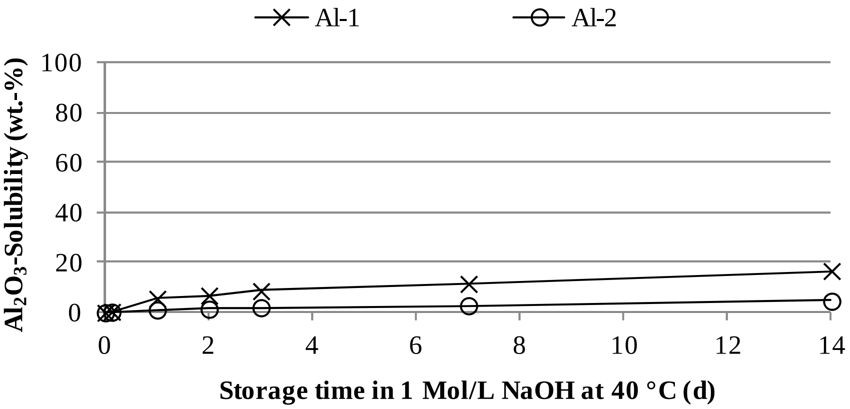

Fig. 2*–Solubility of the aluminium containing waste materials in 1 Mol/L NaOH at 40°C*

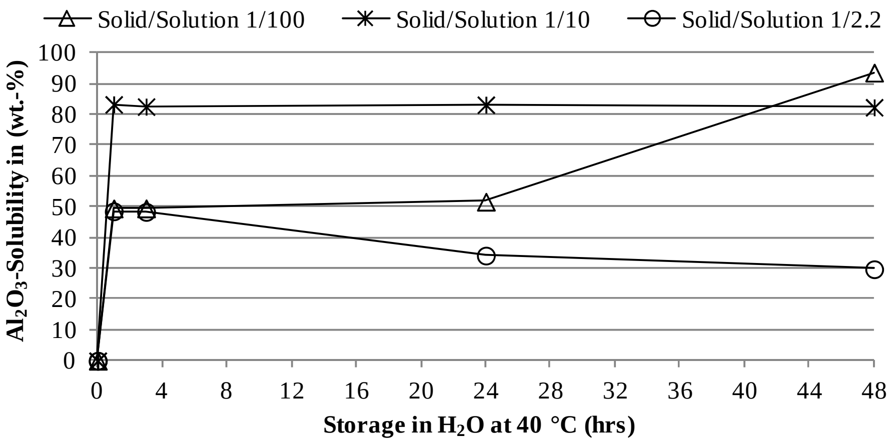

Fig. 3–*Solubility of sodium aluminate in water at 40°C*

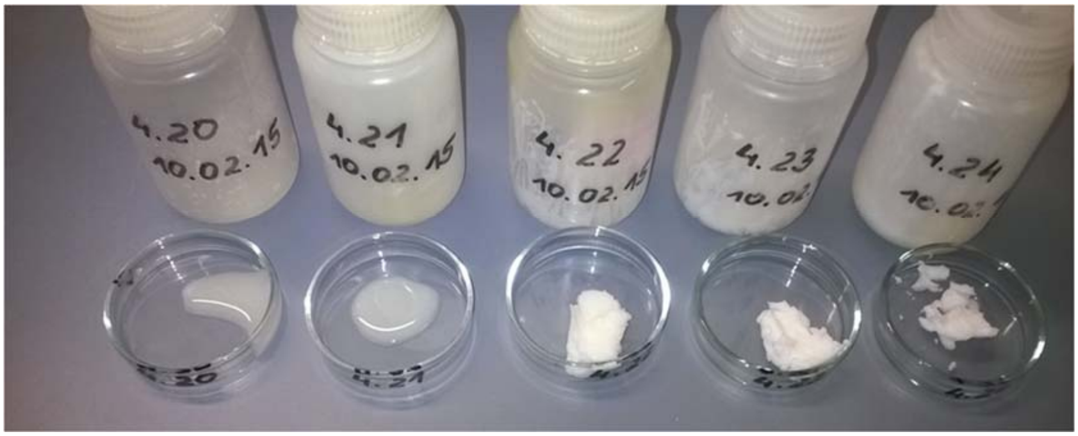

Fig. 4 *– Preliminary investigations to the hardening process*

Fig. 5–*Manufacturing process of the specimens in the technical scale*
top left: container of the Rhoen wheel mixer, top right: Rhoen wheel mixer, bottom left: dry mixture sand +sodium aluminate, bottom right: mix procedure of all components

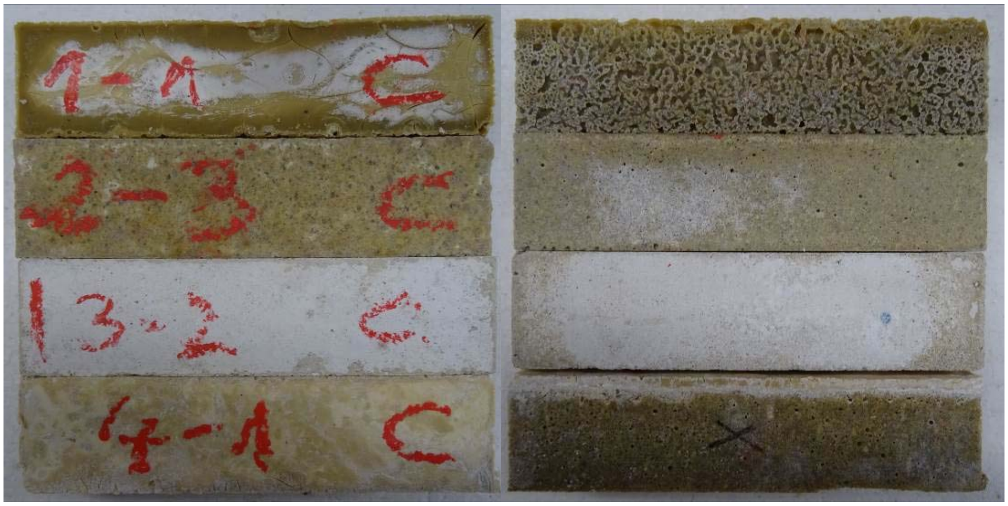

Fig. 6–*Samples R1 to R4 from the manufacturing procedure after 28 d curing*

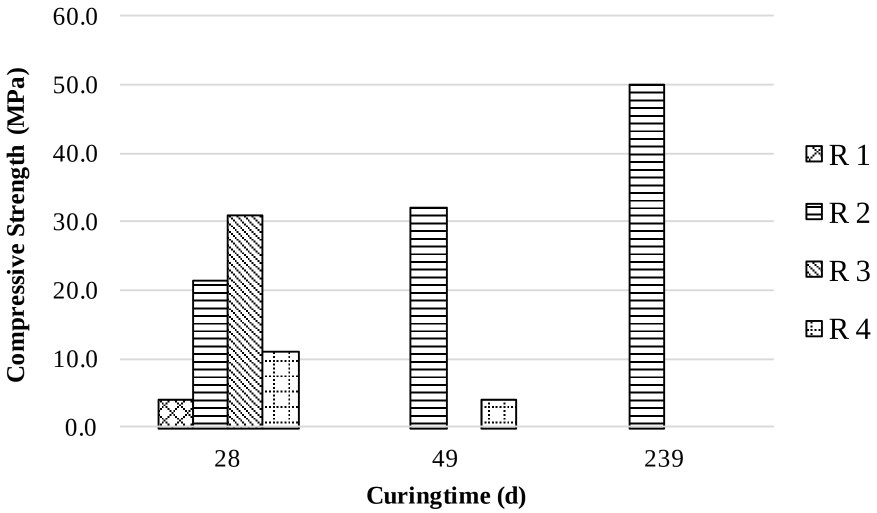

Fig. 7–*Results of the compressive strength development until 8 month*

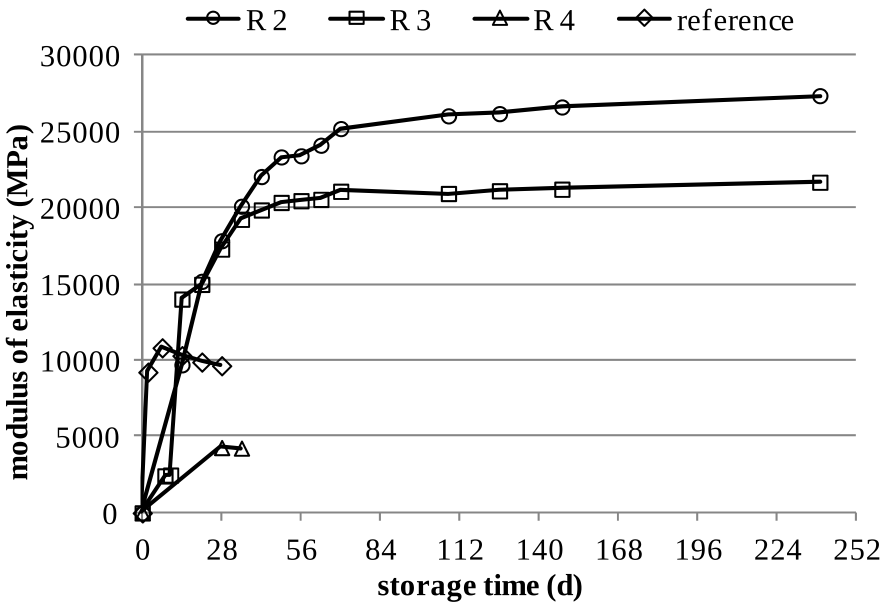

Fig. 8*–Results of the dynamic modulus of elasticity until 8 month*

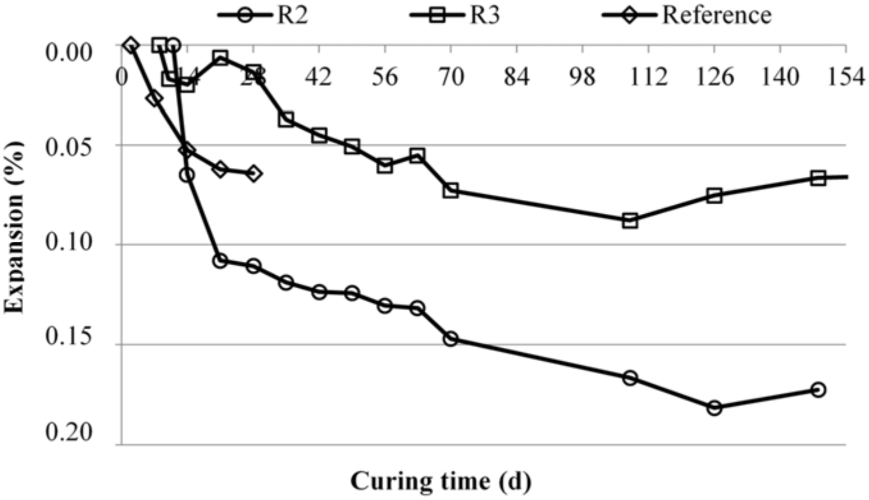

Fig. 9*–Results of the shrinkage behavior until 5 month*

Fig. 10–*Experimental setup for the durability tests in a wide pH range (from 0 to 14)*

Fig. 11a– *Mortar prisms stored in H_2SO_4 (pH 2) until 28 d (left cement, right ASN)*

Fig. 11b–*Mortar prisms stored in H_2SO_4 (pH 2) until 72 d (left cement, right ASN)*