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PMM for Surface Treatment of Concrete with the Utilization of By-Products

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Abstract : The disadvantage of PMM's (polymer-modified mortars) that are currently on the market is the utilization of expensive polymer additives and also the cost of the other components. One of the possibilities how to decrease this price is the effective utilization of waste materials which are very inexpensive in spite of their good properties. The combinations of different degree in polymer adhesiveness and waste secondary raw materials – fly ash – are experimentally verified in the paper. The use of fly ash in adhesive materials for ceramic tiles is limited by unsatisfactory initial adhesiveness to sintered ceramic sherd as a result of a running pozzolanic reaction that lowers the efficiency of polymer additives. On the other hand, the use of adhesive and backfill coating materials for gluing ETICS board insulation materials has brought very good results.

Keywords: polymer-modified mortar, by-products, external thermal insulation composite systems, adhesiveness

1. Introduction

Human activities towards the environment can be characterised as energy-materialistic. Mankind has been constantly and steadily increasing the amount of waste materials in the environment by its activities, which results in disruption of the fragile ecosystem. The term waste materials covers everything that has not been used from the natural resources during manufacturing or any other human activity even after further processing. With regard to utility, waste materials can be classified as exploitable and unexploitable. Exploitable waste materials can be divided into used and non-used ones. Exploitable waste materials can be suitably used as secondary raw materials for further processing either directly or after necessary treatment.

Materials with various properties are necessary for various kinds of concrete structures and their parts. The most important properties are the ones that ensure durability. It can be said that polymer-cement materials have been used more and more extensively recently precisely due to their long-term durability. They are cement bound composite materials utilizing polymer additives improving properties such as deformation, adhesiveness, strength and waterproofing. These materials are called two-component when a polymer component in the form of dispersion is added into classic material during mixing at a building site. As regards to PMM's, recently there has been a new trend: the polymer component is added into the classic material during its production in the form of dry additives called redispersable polymers. These compounds are then classified as single-component polymers of modified cement composition. Classic dispersions are usually acrylates, styrene-acrylates or styrene-butadiene dispersions; mainly alkaline resistant copolymers of ethylene and vinyl acetate (EVA) are added into single-component materials. EVA is copolymer used worldwide for the production of polymer-cement adhesive materials.

Polymer-modified mortar is the material used very frequently for renovations, repairs or generally for upkeeping and maintaining the surface of concrete structures, both for common re-profiling of the surface and for spreading other layers on the structure. Adhesiveness must be a dominant property of such materials because even the highest quality material that will not be firmly anchored or glued to the surface will never be able to fulfil its function. Because there is a full range of PMM's, all the results that we dealt with in our research of PMM's cannot be mentioned here. That is why we have chosen some information from the field of adhesive materials for this article. The Czech Republic has seen a boom in innovations of adhesive materials.

2. Objectives

2.1 Adhesive materials

Adhesive materials are liquid or solid substances that have high inner cohesiveness and adhesiveness to the surface of glued materials. Due to these properties they have the ability of joining firmly the surfaces of the same or different materials by cohesive and adhesive forces.[1] One advantage of gluing can be seen in the possibility of joining the same or different materials without considering their thickness. The properties of glued assembly required for

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its use make an important factor when selecting an adhesive and gluing technology. Such requirements include mechanical strength, thermal endurance, chemical stability, watertightness of the glued joint, resistance to the weather etc. With regard to carried out innovations, we will deal with adhesive materials for ceramic tiling and External Thermal Insulation Composite Systems.

2.2 Adhesive materials for ceramic tiling

Ceramic tiling is a typical multilayer element where joining of several materials of various properties can be found and where mutual co-operation is affected especially by thermal and moisture extensibility. The flexibility module, expressing the ability of material to deform flexibly or vivid under external loads (material flexibility), is another important parameter.

Adhesive materials are the cause of defects in about 20% of cases and it is primarily their wrong selection. One of the conditions of durability of ceramic tiling or paving is quality and stable anchorage of the tiling to its sublayer which is affected by the condition of said sublayer, the character of the tiling back face, the type of adhesive material and especially by external conditions[2] such as low tensile strength, unsatisfactory preparation and generally low ability to fix tiling multilayer for a long time. According to long-time experience the sources of defects are in this area in more than 65% of cases - see Fig. 1.

Other defects of glues dwell especially in imperfect mixing of adhesive materials or mortars as well as in unsuitable selection of an adhesive material for a given environment. This type of defects can be found where adhesive materials, but especially cement mortars, are mixed or made directly at a building site. In such cases not only wrong dosage of the individual components can be met but also the absence of some adhesives etc. On the other hand, the use of industrially produced adhesive materials shows, especially in cement based materials, defects caused by failure in observance of precise water dosage.

2.3 Adhesive and high-build coating materials for External Thermal Insulation Composite Systems (ETICS)

Thermal insulation of buildings in the Czech Republic is a very topical problem given constantly rising costs of fuels and energy. This applies to new buildings as well as the existing ones. Non-transparent structures (external walls, staircase walls, roofs etc.) cause about 40~60% of total heat losses. Average climatic conditions in the Czech Republic can be characterised by mean external temperature during heating season of +3.8°C and the number of 242 heating days, i.e. about 8 months a year.

The basic requirement for thermal insulation is to provide a compact thermal insulating casing of a building. Reasons for thermal insulation of buildings are not only of economical but also technical nature. The technical reasons include: elimination of water vapour condensate on the internal surface of external structures, improvement of thermal comfort, reduction of building overheating in summer months, reduction of loads of heating systems, possibilities of new architectural solutions and, due to saving energy, positive effect on the environment.

The first thermal insulation of a residential house by means of a contact thermal insulation system was implemented in Berlin in 1957. This technology has spread very fast in the Czech Republic as well since then, which can be documented by thousands of insulated facades of prefabbed buildings, family houses and other buildings. The arrival of important world producers has brought a shift in the quality of materials used and systems and home producers have adapted to this trend too.

The basic structure of ETICS consists of a background structure, adhesive material, dowels, lathes, reinforcing layer (consisting of a filling material and reinforcing meshwork), surface treatment and additional elements (set of products for adaptations of corners, dilatation etc.) – see Fig. 2.

3. Waste materials

Waste materials used as secondary raw materials differ from natural raw materials mainly in the following parameters:

- The amount of waste materials is limited by the location and capacity of the waste originator and often by uneven amount of their origin.
- Chemical composition and some physical properties are not frequently stable; they can show big difference in properties.
- Storage and handling waste materials are not carried out with sufficient care; soiling, admixing of other substances may occur; sufficient selectivity of waste material dumps is not ensured. This all hampers the full utilization of waste materials.
- Waste materials often contain various additives that even in the slightest amount may adversely affect the final properties of products made from them.

Power plant fly ash is a mineral residue left after incinerating fossil fuels and obtained by catching it from gaseous combustion products in separators. Annual production of fly ash by power

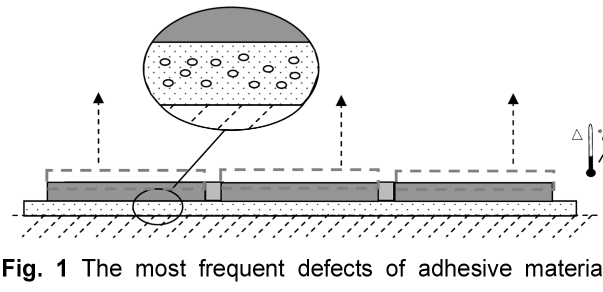


Fig. 1 The most frequent defects of adhesive materials.

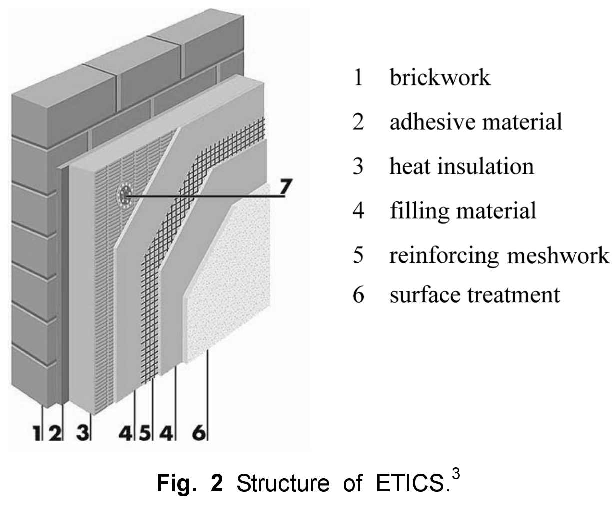


Fig. 2 Structure of ETICS.[3]

plants in the Czech Republic is about 10 Mt and it creates the greatest amount of waste materials in industry and power engineering. More than 70% of electricity in the Czech Republic is produced in thermal power plants incinerating coal.

According to the type of a separator, fly ash can be classified as large-grained, obtained from mechanical separators, and fine-grained, obtained from electric filters.

Two various technologies are generally used for incinerating coal. The first one is classical incinerating under high temperatures. The other method, that seems to be more ecologically and economically efficient, is fluid incinerating of fuels. This means that ground fuel with an admixture of limestone or dolomite is incinerated in a circulating layer. In the course of a dissociative process the released SO_2 binds with CaO producing $CaSO_4$.

Power plant fly ash is very similar to trass, natural pozzolan, in its properties and composition.[4] The industrial utilization of power plant fly ash represents only about 10 % and due to high production of fly ash in the Czech Republic it is important to ensure its efficient use in building industry where mainly its pozzolanic properties can be utilized.[5] That means the use of the reaction of silicate material with calcium hydrate [$Ca(OH)_2$] because substances similar to ones created during setting and hardening of Portland cement are formed:

$$x\, Ca(OH)_2 + y\, SiO_2 + z\, H_2O \rightarrow x\, CaO \,.\, y\, SiO_2 \,.\, z\, H_2O \qquad (1)$$

Particles of individual kinds of fly ash partly differ from each other but it can be generally said that fly ash particles, if they were formed of sufficiently liquid melting, are of a round shape with 1 to 100 μm in diameter. The little balls are either filled or hollow and their enamel is usually greyish or yellowish.

Physical properties such as bulk density, specific surface area and specific density are the basic physical characteristics of fly ash.[6]

- Specific surface area - it usually falls between 170 to 300 m^2/kg according to Blain.
- Specific density - is roughly between 2,090 to 2,670 kg/m^3 with the average of 2,300 kg/m^3.
- Bulk density - the values are between 500 to 910 kg/m^3 for loose fly ash and from720 to 1,320 kg/m^3 for settled fly ash with the average of 990 $kg/m.^3$

Fly ash can be divided into three types according to particle shape:

- Most of fly ash contains round enamelled particles of varying sizes. Fly ash from a power plant used by us falls into this type.
- Round enamelled particles are eroded and corroded.
- Apart from a share of enamelled particles there are also particles cased in a highly porous layer.

Mineralogical composition of fly ash depends primarily on inorganic constituents of the incinerated coal and the conditions of combustion process. Fly ash mainly contains non-transparent grains formed by silicious glass, lodestone, gehlenite and periclase. The transparent are represented by quartz, muscovite, carbonates, and sulphates.

Mineralogical composition of fly ash:

- Hydraulically active components (aluminosilicate minerals, Ca-aluminates, Ca-silicates, Ca-ferrites, glasses)
- Non-hydraulic minerals sometimes acting as hydraulicity exciters (anhydrite, CaO, MgO)
- Initiatory (exciting) components of hydraulicity (sulphides, alkaline salts)
- Inactive substances (crystalline quartz, hematite, lodestone, mullite, acid silicates, newly created carbonates)

Fly ash can be divided into three groups according to its chemical composition:[7]

- Basic fly ash with a high content of CaO (about 30%)
- Acid fly ash with a content of SiO_2 over 50% and mean content of $Al_2O_3 + Fe_2O_3$ to 10%
- Aluminosilicate fly ash with a content of about 50% of SiO_2 and about 30 % of Al_2O_3

Power plant fly ash from a power plant in the Czech Republic was used for this work. The power plant incinerates lignite and uses classical technologies of incinerating solid fuels. Annual production of fly ash is about 300,000 tons and only about 33,000 tons of the fly ash is utilized in the building industry. The rest is in the form of stabiliser (a mixture of fly ash and limestone) and is used for artificial countryside adaptation, specifically as filling a quarry after finished mining. Combustion is done under high temperatures (1,400~1,600°C). Power plant fly ash from classical incinerating frequently contains, apart from β-quartz and other crystalline modifications of SiO_2 cristobalite and tridymite, mullite is some amount, usually more than 50 % of vitreous basis and iron oxides (α-Fe_2O_3–hematite and γ-Fe_2O_3 – maghemite).

Chemical composition of fly ash from power sources using coal as their fuel is roughly as follows: 40~50% of SiO_2, 15~35% of Al_2O_3, 4~10% of Fe_2O_3, 0.5~5% of TiO_2, 2~8% of CaO, 1~3% of MgO, 1~5% of $K_2O + Na_2O$. Power plant fly ash shows the higher reactivity, the higher is its content of SiO_2 present in the enamel and simultaneously the lower is the content of mullite.[8]

The set objectives of this work can be summarised as follows:

- To monitor changes in properties of polymer-cement adhesive and backfill coating materials by applying selected types of waste materials as a replacement of part of filler content.
- To find maximal possible limit of filling with individual waste raw materials.

4. Results and discussion

Basic physical-mechanical and physical-chemical tests were carried out for input raw materials. The tests provided a survey on the composition and structure of the waste materials. The tests included especially granulometry, chemical analyses, DTA and X-ray analyses, specific densities were determined.

4.1 Properties of power plant fly ash

Fly ash from a power plant incinerating lignite was used for this work. Properties and composition of the used fly ash are listed in Tables 1 and 2. The results of mesh analysis of the fly ash are in graphic form in Fig. 3.

Mineralogical composition of the used fly ash: β quartz, mullite, hematite

Effectiveness index of the used fly ash (according to ČSN[a] EN 450):

after 28 days of maturing 84%

after 90 days of maturing 93%

[a]Czech national Standard

4.2 Influence of fly ash admixture on the properties of adhesive materials for ceramic tiling

Formulas of reference mixtures, i.e. mixtures without added power plant fly ash, are listed in Table 3.

Within our experiments a gradual replacement of the filler with fly ash was carried out in the amounts of 20, 40, 60 and 80 %. Fly ash contains a high proportion of fine particles and that is why ground limestone was used as well. The resulting values of adhesiveness according to ČSN EN 1348 can be found in diagram in Fig. 4.

The diagram makes it obvious that none of the tested mixtures reached the initial adhesiveness value of the reference mixture according to ČSN EN 1348 even with the lowest content of fly ash of 20%.

On the other hand, another test according to ČSN EN 1346 showed that the standard requirement for open time determined by means of tensile adhesiveness using the mixture with maximum fly ash content of 60% was reached in mixtures SA and SB; the mixture with 80% of fly ash in the SC mixtures fulfils the requirement too.

The results obtained during the determination of open time by means of tensile adhesiveness according to ČSN EN 1346 were markedly better in the mixtures with fly ash than the results of initial adhesiveness according to ČSN EN 1348. With regard to inconsistent results their cause had to be found.

Elimination of concrete board absorbability was carried out in both cases by means of concrete surface penetration. When a tiling element is placed on the layer of adhesive material that has been spread over the penetrated board, sucking off water either to the background or the tiling element does not take place. When determining initial adhesiveness according to ČSN EN 1348, the standard prescribes the use of ceramic tiling elements of AI type (according to EN 176) with absorbability of only weight 0.2 %. Ceramic tiling of 0.17% absorbability was used in our experiments.

The cause of the marked decrease in initial absorbability in the

Table 1 Specific density, specific surface area and absorbability of the used fly ash.

Monitored parameter	Found out value
Specific density (kg/m^3)	2,040
Specific surface area (m^2/kg)	242.6
Absorbability (%)	75.1

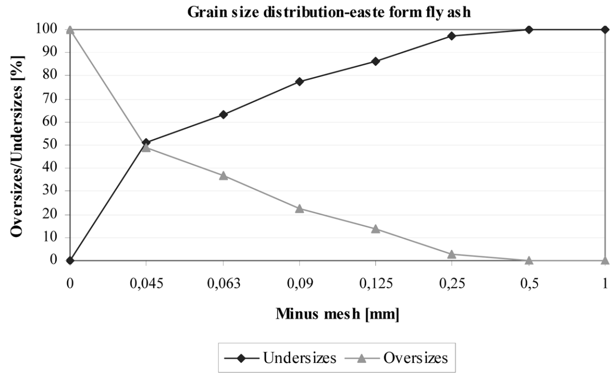


Fig. 3 Fly ash total residue/sifting on screens.

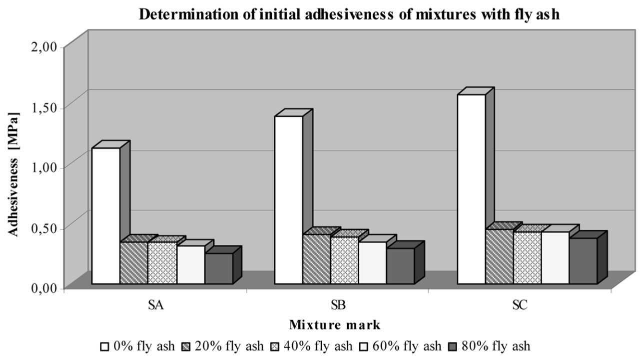


Fig. 4 Determination of initial adhesiveness of mixtures with fly ash according to ČSN EN 1348.

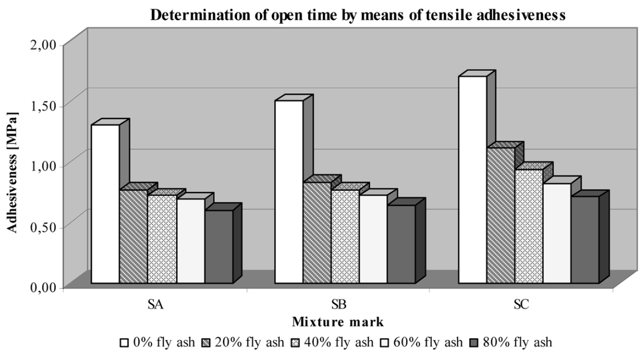


Fig. 5 Determination of open time by means of tensile adhesiveness of mixtures with fly ash according to ČSN EN 1346.

Table 2 Chemical composition of the used fly ash.

CaO (%)	MnO (%)	Al_2O_3 (%)	Fe_2O_3 (%)	Sulphates (%)	SiO_2 (%)	Na_2O (%)	MgO (%)	K_2O (%)	TiO_2 (%)	ZZ (%)
1.79	0.03	28.93	6.08	0.2	56.82	0.32	1.31	1.79	2.02	0.74

Table 3 Composition of reference mixtures for gluing ceramic tiling.

Component of adhesive mixture for gluing ceramic tiling		Composition of reference mixtures (%)		
		SA/R	SB/R	SC/R
Binder	Cement CEM 52,5 R	36.00	36.00	36.00
Filler	Fine ground limestone	43.55	43.20	40.60
	Ground limestone 0.2 ~ 0.5 mm	18.60	17.15	17.30
Additives	Copolymer EVA, foam remover, cellulose ether	1.85	3.65	6.10

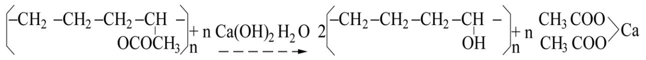
SA/R – common adhesive material

SB/R – adhesive material with increased adhesiveness

SC/R – adhesive material with increased adhesiveness and joint flexibility mixtures with fly ash can therefore be seen in a more intensive pozzolanic reaction of fly ash, which was confirmed by the study of microstructure (see chapter 4.4). Due to a relative sufficiency of water a greater amount of $Ca(OH)_2$ that can take part in the pozzolanic reaction creates at the beginning. The pozzolanic reaction can be expressed as follows:

$$x\,Ca(OH)_2 + y\,SiO_2 + z\,H_2O \rightarrow x\,CaO\,.\,y\,SiO_2\,.\,z\,H_2O \qquad (2)$$

But then a marked reduction of $Ca(OH)_2$ takes place, which is caused by the running pozzolanic reaction. This fact may limit cross-linking of the polymer additive and also the interaction with Ca^{2+} ions that can be expressed as follows:



A sufficient amount of $Ca(OH)_2$ solution is necessary for the interaction of the polymer additive with Ca^{2+} ions. The ion can be bound from the solution, which is not apparently possible in the case of a running pozzolanic reaction.

A present pozzolanic reaction is positively indicated by a carried out thermic analysis and RDA where the sample containing fly ash showed a higher content of C-S-H phase than it was in the reference sample (chapter 4.4).

Lowered values of initial adhesiveness in the mixtures containing fly ash as a filling component can be ascribed to estimated synergic effect of both cross-linkage of the polymer component, which leads to the creation of insufficient polymer film that is one of the factors affecting adhesiveness to a ceramic element, and limiting the interaction of the polymer additive with Ca^{2+} ions.

In case of the determination of open time by means of tensile adhesiveness according to ČSN EN 1346 the use of a ceramic tiling element of the BIII type with 15 ± 3% absorbability (ceramic tiling elements with 12.7% absorbability were used) is required. Therefore it can be assumed that, in case of the determination of open time by means of tensile adhesiveness, part of water was drained by the ceramic element, which limited the course of the pozzolanic reaction and that adhesiveness was increased also due to partial mechanical anchorage of the adhesive material in a porous sherd.

4.3 Influence of fly ash admixture on the properties of adhesive and high-build coating materials for External Thermal Insulation Composite Systems (ETICS)

Formulas of reference mixtures for ETICS, i.e. mixtures without added power plant fly ash, are listed in Table 4.

In this part of experimental work too ground limestone was

Table 4 Composition of reference mixtures for ETICS.

Component of mixture for ETICS		Composition of reference mixtures (%)		
		SD/R	SE/R	SF/R
Binder	Cement CEM I 52,5 R	38.00	38.00	37.00
Filler	Siliceous sand	0.00	16.60	16.20
	Ground limestone 0.2~0.5 mm	57.60	40.40	40.10
Additives	Copolymer EVA, foam remover, cellulose ether	4.40	5.00	6.70

SD/R – adhesive and backfill coating material with higher frost resistance
SE/R – common adhesive and backfill coating material
SF/R – adhesive and backfill coating material with higher frost resistance and joint flexibility

Table 5 Composition and properties of mixtures with fly ash.

Sample mark	Proportion of the used filler (%)		Adhesiveness		
	Ground limestone and sand	Fly ash	To concrete	To concrete after 25 FC**	To a thermal insulant: - from polystyrene - from polystyrene after immersion in water - from polystyrene after cycles of immersion in water and drying
			MPa	MPa	MPa
SD/R	100	0	1.82	2.24	0.15*
SD/FA2	80	20	2.03	2.15	0.15*
SD/FA4	60	40	1.65	2.09	0.15*
SD/FA6	40	60	1.36	1.85	0.15*
SD/FA8	20	80	1.16	1.41	0.15*
SE/R	100	0	1.87	1.93	0.15*
SE/FA2	80	20	1.48	1.74	0.15*
SE/FA4	60	40	1.66	1.72	0.15*
SE/FA6	40	60	2.05	2.14	0.15*
SE/FA8	20	80	1.55	1.61	0.15*
SF/R	100	0	2.12	2.14	0.15*
SF/FA2	80	20	1.99	2.26	0.15*
SF/FA4	60	40	1.97	2.05	0.15*
SF/FA6	40	60	1.78	1.90	0.15*
SF/FA8	20	80	1.70	1.79	0.15*

* Defects were found in the thermal insulant in all the cases
** FC – freezing cycle

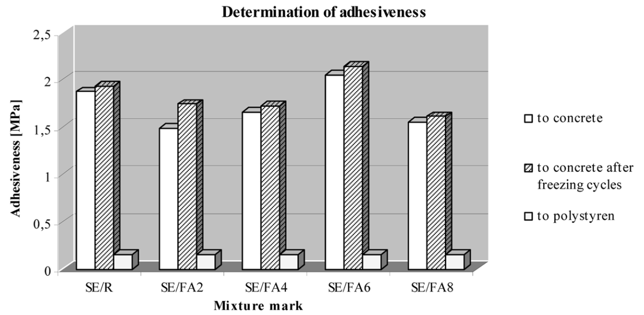


Fig. 6 Determination of adhesiveness according to ČSN EN 1542 of SE mixtures with fly ash.

replaced by fly ash in the amounts of 20, 40, 60 and 80%. The results obtained when working on this part are presented in Table 5 and shown in a graphic form in Fig. 6.

As is apparent from the graph, a very positive result was obtained in SE/FA6 mixture containing 60% of fly ash. This mixture showed adhesiveness values higher than the reference mixture both after 28 days and after freezing cycles. The effect of the polymer additive was checked, the filler granularity curve was modified in a suitable way and the above described pozzolanic reaction of the fly ash contributed to an improvement of the monitored physical and mechanical parameters.

4.4 Study of microstructure

Two samples were selected for studying the effect of a pozzolanic reaction of fly ash. One sample contained SC/FA6 fly ash – content of 60% of fly ash as a filling component and it was compared to the reference sample (SC/R). Thermograms of the reference SC/R and the SC/FA6 mixture, containing 60% of fly ash the replaces part of the filler, are shown in Fig. 7.

The sample containing SC/FA6 fly ash shows an increase in weight loss in the area of 260°C caused by a higher content of crystalline C-S-H phase and conversely at 510°C by lower content of portlandite or secondary calcite too at around 850°C.

Figs. 9 and 10 present RDA records of the reference mixture and a mixture containing fly ash. A raised background is obvious in the picture with the sample with added fly ash, which is caused, apart from others, by a higher content of C-S-H phase. A higher amount of portlandite and calcite created from it was identified in the reference sample. These results are in harmony with the analysis using REM (Fig. 9) where pseudomorphs of portlandite were identified with an electronic probe in the first photograph. Portlandite transformed to minerals of gyrolite and aphwillite type, which is documented by the second photograph. The third photograph shows residuary relicts of portlandite that has not morphed to C-S-H gels and crystals or calcite.

The following minerals were identified:

- The S1/R mixture:calcite (K), portlandite (P), ettringite (E), quartz (Q), β dicalcium silicate (B).
- The S1/P6 mixture: calcite (K), portlandite (P), ettringite (E), quartz (Q), β dicalcium silicate (B), mullite (M).

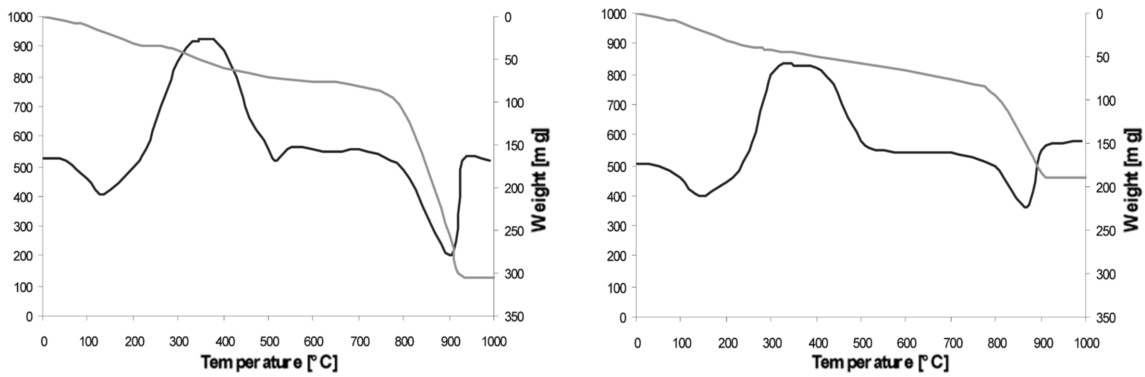


Fig. 7 Thermograms of the SC/R and SC/FA6 samples.

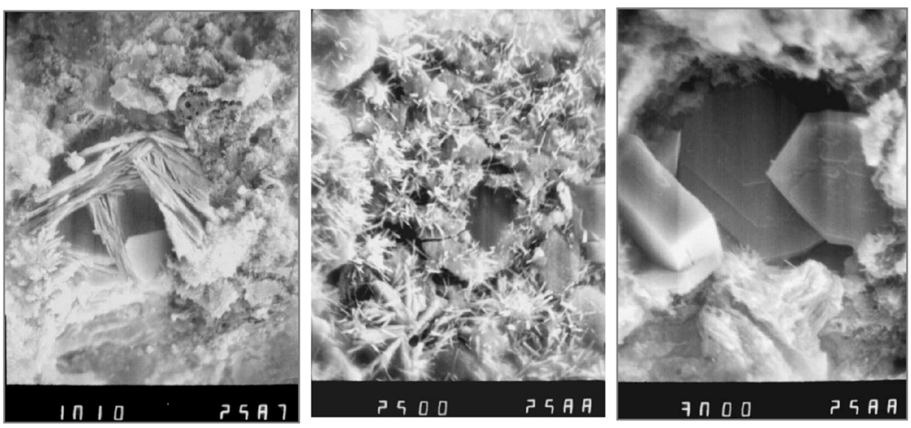


Fig. 8 Photos from REM of the SC/FA6 mixture containing 60% of fly ash as filling component, magnification of 1,010, 2,500 and 3,000 times.

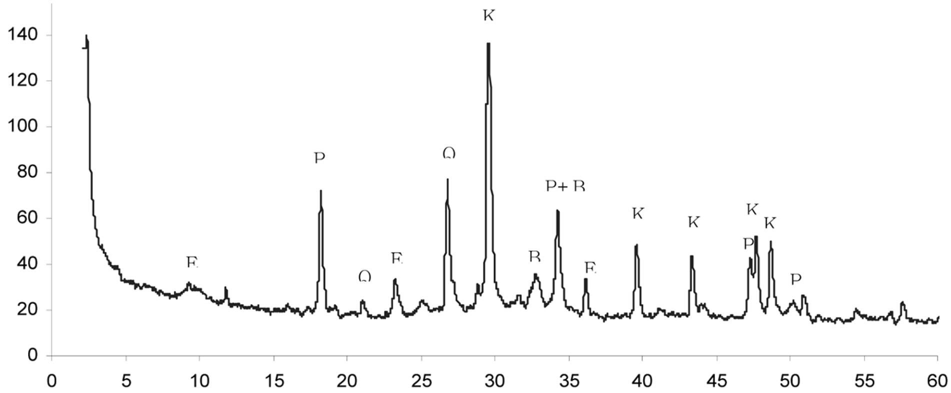


Fig. 9 RDA record of the SC/R mixture.

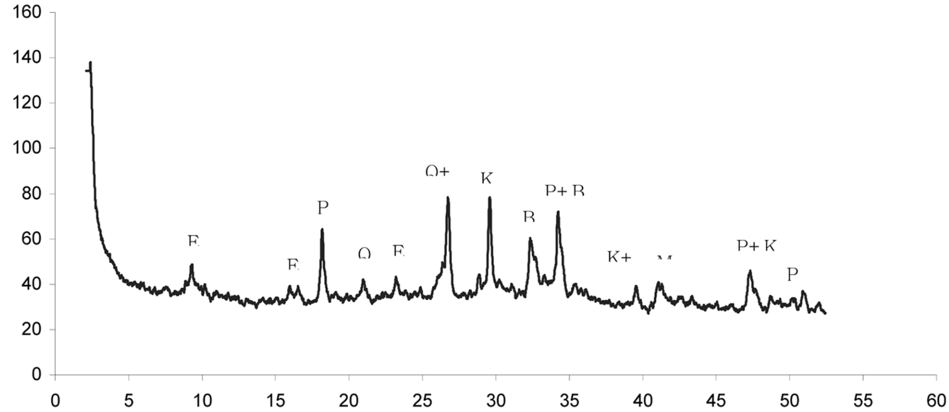


Fig. 10 RDA record of the SC/FA6 mixture.

5. Conclusions

When observing the modification of polymer-cement mixtures by power plant fly ash it was interesting that fly ash increased strength characteristics and frost resistance of the tested mixtures. Its use in adhesive materials for ceramic tiling is however limited by insufficient adhesiveness to sintered ceramic sherd due to a more intensive pozzolanic reaction and creation of crystalline C-S-H phase that decreases the efficiency of polymer additives. In case of adhesive and high-build coating materials for ETICS very good results were obtained and application of fly ash can lead to the increase in adhesiveness values after subject to frost as well as tensile and bending strength. There was positive effect on frost resistance and absorbability of mixtures containing this waste material, which makes it predestined for use especially as backfill coating materials where higher frost resistance is required.

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