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RECYCLING OF STEEL FURNACE SLAGS (SFS) BY EFFICIENT INTEGRATION IN CONSTRUCTION MATERIALS AS AGGREGATE PARTIAL REPLACEMENT

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Abstract

Slags, mixtures of mainly metal oxides and silicon dioxide, represent by-products or wastes generated by the ore smelting processes. There are several types of slags, but for construction applications as recycling possibilities, there are generally used slags generated by iron and steel making industry. The present study is focused on electric arc furnace slags (EAF), produced, and stored in the western part of Romania, Caransebeș city. The current slag deposit has been operating on this site since 1771, with the establishment of the furnaces to produce cast iron in Reșita. This paper presents a preliminary experimental study on the possibilities of using the Reșița SFS slags as a partial substitute for aggregate in cementitious materials for the construction industry. The opportunity and necessity of the proposed research direction cover several purposes: waste management implementation, environmental protection and natural resources saving, for the Circular Economy (CE) implementation in the Romanian industry. The initial results emphasise the concept's viability applied to the Reșita slag landfill, encouraging further exploration of this environmental engineering topic.

***Key words**: Circular Economy, construction eco-materials, mineral addition, steel furnace slag (SFS), waste recycling.*

INTRODUCTION

As a result of the diverse smelting processes applied to metallic ores generate slags, valuable by-products emerging as a potentially valuable resource for various civil infrastructure applications. The current study focuses on using slags generated from different steelmaking procedures. In the steel production process, the primary refinement takes place at a blast furnace, where iron ore undergoes processing to produce pig iron. The resulting slag from this process, known as blast furnace slag, has applications such as being employed as an aggregate in the form of air-cooled blast furnace slag (ACBFS). However, it is more commonly utilized as a supplementary cementitious material (SCM) in the form of ground granulated blast furnace slag (GGBFS) (Shi & Qian, 2000; Snellings et al., 2012; Özbay et al., 2016; Wang, 2016; Buddhdev & Timani, 2020). Following the blast furnace, pig iron undergoes refinement to produce crude steel. Additionally, other forms of crude steel are created through the refining of recycled and scrap steel. Various processes can be employed for this purpose, and the resulting slag from these processes is collectively referred to as steel furnace slag (SFS). The steel furnace slag (SFS) includes: a) the electric arc furnace (EAF) slag; b) the basic oxygen furnace (BOF) slag; c) the ladle metallurgy furnace (LMF) slag; and d) the argon oxygen decarburization (AOD) slag (Brand & Fanijo, 2020). (The effective utilization of SFS worldwide is estimated to be around 80% (Branca et al, 2020). Over time various research studies investigated the different types of SFS materials and their impact on the properties of cementitious composites when used as an aggregate and/or supplementary cementitious material (SCM). The general conclusions indicate that not all SFS additions are identical and consequently, they should be categorized or qualified based on their intended purpose or use, especially when considering applications such as concrete, mortar, alkali-activated material, or soil stabilization (Brand & Fanijo, 2020).

The steel furnace slag (SFS) aggregate can significantly impact the properties of cementitious composites, even when the mixture volumetrics remain constant (Giergiczny, 2014; 2019). For instance, concrete incorporating recycled concrete aggregates (RCAs) typically exhibits strength reductions compared to virgin aggregates, but the extent of this reduction varies widely, depending on several influencing factors, such as microstructure development, adhered mortar, moisture absorption and content, RCA heterogeneity, and overall quality (Li & Herbert, 2012; Snoeck, 2015; Teixeira et al., 2016; Hemalatha & Ramaswamy, 2017; Li, 2008). In the case of SFS aggregates, even when considering the same type of SFS, research indicates that concrete strength can either increase, decrease, or remain similar to that of concrete with virgin aggregates.

Slag possesses a complex chemical composition characterized by elements such as calcium oxide, silicon oxide, aluminium oxide, and others, which determines the physical, mechanical and durability characteristics of materials where used. Slag concretes can develop compressive tensile properties comparable to classic mixes, sometimes even superior. At the same time, the durability performance of slag cement-based materials offers better durability behaviour (Brand & Fanijo, 2020).

Aggregates (sand, gravel and coarse particles), play a pivotal role regarding the mix design of both concrete and mortar, constituting a substantial proportion of their and also determining the future material performance in terms of physical, mechanical and durability. Nevertheless, aggregates are a finite and non-renewable resource sourced from rivers or quarries, and their excessive exploitation, particularly in Romania, poses a considerable threat to the environment. This overuse can lead to adverse consequences such as shoreline erosion, diminished groundwater levels, destruction of local flora and fauna, landslides, and accidents. The existing legislative framework in Romania is deemed inadequate as it permits excessive and detrimental extraction practices. Furthermore, Romania faces a scarcity of sand, particularly in the fine-grain form, intensifying the urgency for viable solutions to address these pressing issues (Hemalatha & Ramaswamy, 2017).

Valorisation of mineral additions (by-products/residues of industrial flows) in construction materials via innovative inclusion processes, in the context of Circular Economy (CE) strategies in Romania

The current research is applied to steel furnace slags (SFS), specifically electric arc furnace slags (EAF), produced and stored on this site, in the western part of Romania, in Reșita, Caraș-Severin County, since 1771. This paper presents a targeted, theoretical and experimental study on the possibilities of using the identified SFS slags of EAF type, as a partial substitute for aggregate in cementitious materials for the construction industry. The intended major outcome of the study is the increase of the industrial by-product added value concerning the current use. The opportunity and necessity of the proposed research direction were identified by the use of strategic management specific methods, within the research project PN 23 35 04 01 of Nucleu Programme of the National Research Development and Innovation Plan 2022-2027, "ECODIGICONS", supported by the Ministry of Research, Innovation and Digitalization, and it is covering several purposes associated to the project's objectives: waste management implementation, environmental protection and natural resources saving, for the Circular Economy (CE) implementing in the Romanian industry.

The presented research is conducted with respect to the first Axis, the *Development of innovative engineering solutions for eco-intelligent construction products by an efficient capitalization of additions generated by local industries*, of the two fundamental research axes of the PN 23 35 04 The project's objectives include identifying scientific, technical and applied solutions for the national construction infrastructure, in accordance with the National Strategy regarding the Circular Economy (SNEC 2022), National Strategy for Research, Innovation and Smart Specialization (SNCISI 2022-2027), and the Recovery and Resilience Plan for Romania (PNRR).

The first Axis of the project is concerned with identifying recovery solutions for mineral wastes or by-products as results of local or regional industrial processes, inert, latent hydraulic or with pozzolanic activity materials, with potential applicability in construction materials or products (ADD-S). Targeted mineral additions, in the form of powders, coarse granules, slurries, or mixtures in suspension, are evaluated from this valorising perspective as partial or complete replacement of traditional constituents: aggregates as "skeleton" of concrete or cement as binder.

The research currently aims to identify ADD-S type materials produced by the local industry (Timisoara, Timiș county, the Western region or even Romania) and to assess their potential valorisation in cementitious materials, geopolymers, mixtures asphalt, etc., with diverse applicability areas for the innovative engineering development by the means of eco-intelligent construction products, with advanced functionality (materials, elements and structures, models and technologies, circular eco-design algorithms and directions, etc.), in the context of SNCISI 2022-2027 regarding the transition to EC and SNEC 2022.

Figure 1 presents the Eco-CP Flowchart, namely the logical scheme for the structural implementation of specific project objectives associated with the first Axis of the project. The considered abbreviations are the following:

- ADD-S: Additions Derived from Waste and Industrial Byproducts (inert/hydraulically latent/pozzolanic, etc.), and/or derived from demolition and/or decommissioning of buildings;
- AdT: Target Additions (AdT), with high potential for capitalization in circular design for eco-intelligent construction products;
- Eco-CCM: Eco-intelligent Composite Materials for Constructions, with advanced functionality (Eco-Composite Materials for Constructions);
- Eco-CP: Eco-intelligent Construction Products, with advanced functionality, made by using Eco-CCM (Eco-Construction Products).

One of the identified AdT directions is represented by the Steel slag, SFS (EAF and/or BOF), waste/by-product, specifically electric arc furnace slags (EAF), produced and stored in the western part of Romania, in Reșita, Caraș-Severin County.

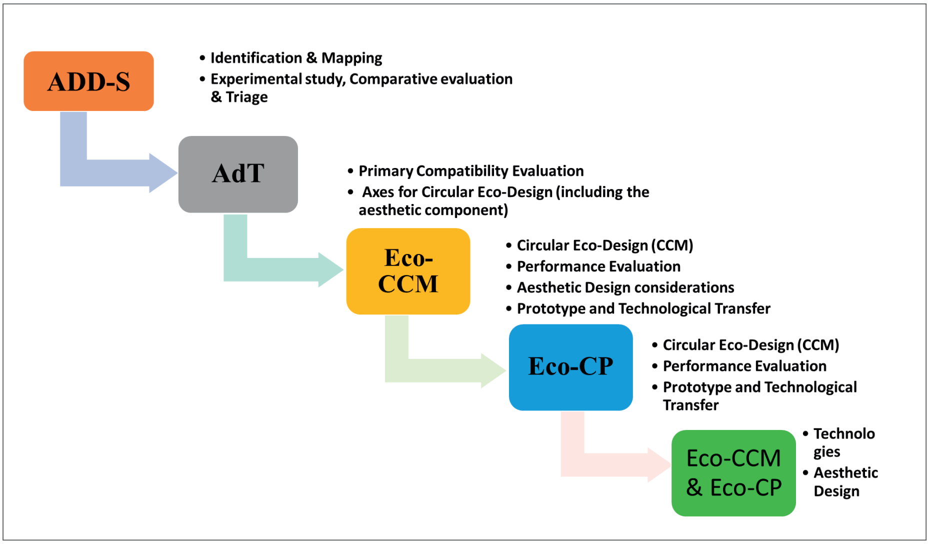

Figure 1. Eco-CP Flowchart: The logical scheme for structural implementation of specific project objectives associated with the first Axis of NIRD URBAN-INCERC research project PN 23 35 04 01

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MATERIALS AND METHODS

The present preliminary research is performed to evaluate the opportunity to capitalize on the targeted addition (AdT), namely the EAF slag derived from local industries from the Romanian Western Region (Caraș-Severin County) in cement-based materials for construction. The mineral waste particularly used for the study is abbreviated as RS1, representing the Reșita EAF slag of granular class 0/4 (Figure 2, f). The applied procedure consists in developing of regular mortar mixes, References (R) and also test mortars, RS1 mortars, developed by using RS1 addition. The References (R) mortars represent usual cementitious mixes (mortars) of aggregate (sand), binder (cement) and water. The RS1 test mortars are produced by considering the RS1 addition as a partial substitute for sand in the reference (R), aiming to fast evaluate the elementary compatibility of the RS1 material with the cementitious matrix and thus, the viability of the research direction. The specific comparative analyses of the RS1 mortars with respect to the References (R) are performed in terms of physical and mechanical performance. The control mixes (R1 and R2) represent usual mortars, and the test mixes (RS1 mortars) are developed by considering the RS1 addition as a partial substitute for sand in the reference (R): 25% and 50% substitution percentage by mass, in accordance with the classical approach of previous studies.

The sequence and duration of the mixing operations are established following the EN 196-1 specifications. The procedure proves viable and is maintained without adjustments throughout the present experimental stage.

Raw materials selective analysis is also performed. The developed mixes (R and RS1) are physically and mechanically evaluated, considering both fresh and hardened state: visual analysis of the mixes, consistency, density, and early age flexural and compressive strength, as critical parameters for further development of the study.

Raw materials

The R and RS1 mixes were produced with locally available raw materials (Figure 2):

- Natural aggregates of granular class 0/4 (sand, S), (Figure 2, a and b);
- Portland Cement, CEM II/A-LL 42.5 R (C), Carpatcement (Figure 2, c and d);
- AdT: EAF slag derived from local industries from the Romanian Western Region (Reșița, Caraș-Severin County), RS1 (Figure 2, e and f).
- Water (tap water).

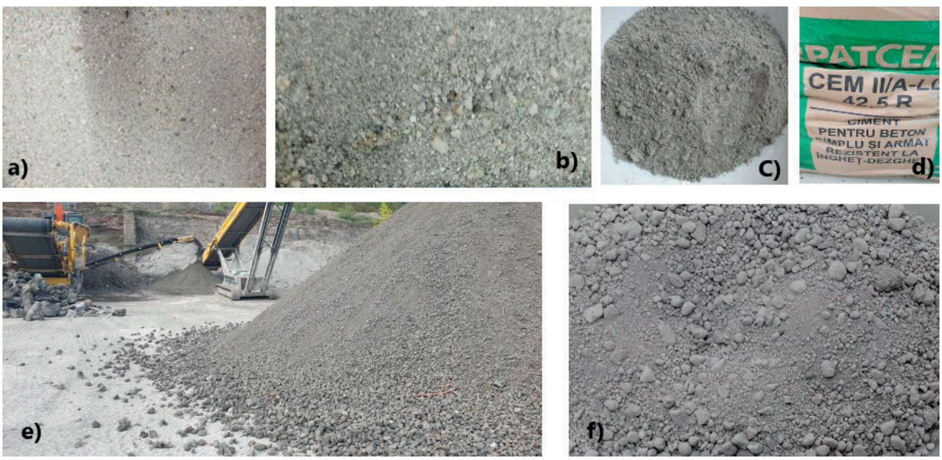

Figure 2. Raw materials for cement-based mortar mix design: a) and b) Natural sand 0/4 (S 0/4); c) and d) Binder - Carpatcement Portland Cement CEM II/A-LL 42.5 R; e) AdT: EOF (RS) derived from the aggregate crushing line, Caraș-Severin County; f) selected AdT: RS1

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Preliminary identification procedures for the RS 1 addition

Experimental testing is carried out in order to analyse the RS1 addition inert additions from a physical, mechanical, chemical, mineralogical, etc. aspect, to obtain a preliminary, identification assessment, along with the preliminary compositional integration stage. The EOF crushed aggregate, RS1, was subjected to SEM imaging preliminary determinations, performed by the ISIM Timisoara laboratory. The analysis was done in terms of visual analysis of granule shape and also of chemical analysis performed by energy dispersive spectroscopy (EDS) technique. Table 1 and Figure 3 present the chemical composition of RS1 particles and Figure 4 shows the visual characteristics of the RS1 particles.

The experimental sieving analysis of the samples is performed according to EN 933-1 method, and the results are presented in Figure 5. Granulometric analysis by sieving is carried out both, for the usual sand 0/4 (S), for the substitution by-product RS1 and additionally for the S + RS1 mixtures, in the considered substitution proportions: RS1 25% and RS1 50%.

Table 1. SEM analysis: Chemical analysis of RS1

CaSiO			
Element	Signal Type	Wt %	Wt % Sigma
O	EDS	48.78	0.71
Mg	EDS	3.65	0.17
Al	EDS	2.63	0.14
Si	EDS	10.36	0.23
S	EDS	1.71	0.10
Ca	EDS	30.19	0.46
Mn	EDS	0.36	0.11
Fe	EDS	2.31	0.16
Total		100.00	

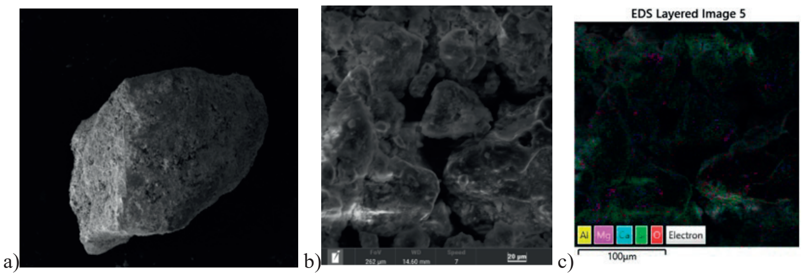

Figure 3. SEM analysis: Visual aspect of ADD-S RS1 grains

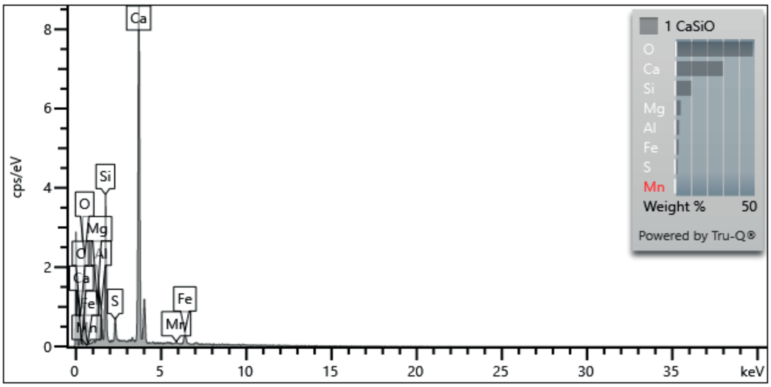

Figure 4. SEM analysis: Chemical analysis of RS1

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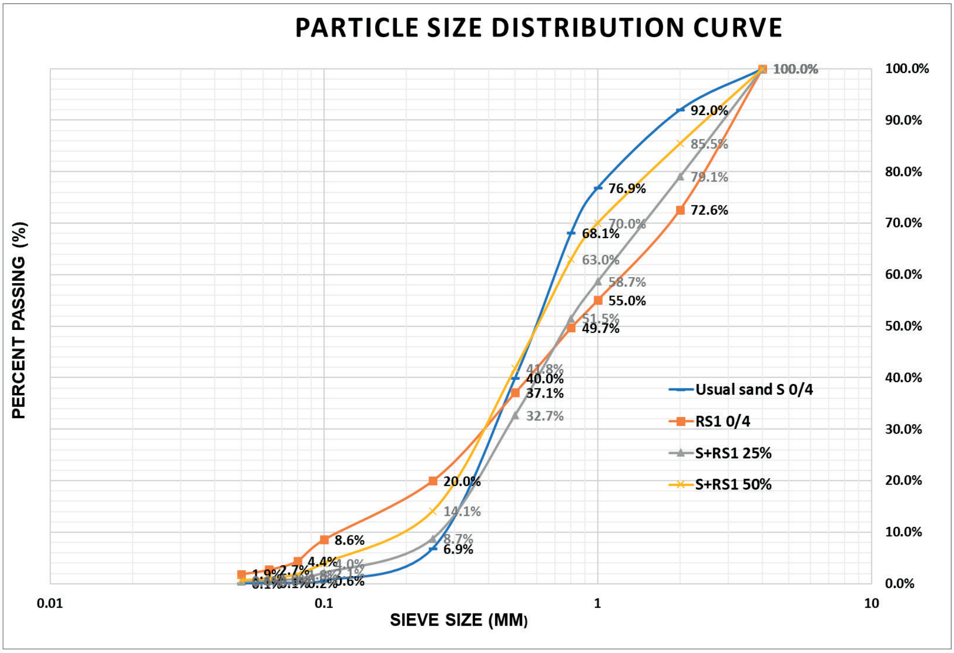

Figure 5. Aggregate grading: Natural sand 0/4 (S), RS1 and aggregate mixes according to the proposed proportions

Mix proportion and specimen preparation

The reference mortar mixes, with classic raw materials: regular sand 0/4 (S), cement and water are produced. The initial reference mix R1, was produced, characterised by the water-to-cement ratio (W/C) 0.56. Further on, considering the dynamics of the research, R2 (W/C = 0.61) was developed. Development of the RS1 mortar mixes implied incremental replacement, by mass, of the usual aggregate (S), with RS1 additions. The considered substitution proportions are RS1 25% and RS1 50%.

The fresh state characteristics of the RS1 mixes show an alteration of workability, especially when using the 50% RS1 substitution, which determined the use of R2 (W/C = 0.61) suitable for relevant specific evaluation. Table 2 presents the mix proportions (established with respect to the cement content, C = 1.0) of all the mixtures used for the preliminary study: the References (R1 and R2) and the RS1 mortars (RS1-25%-0.56; RS1-25%-0.61; RS1-50%-0.61).

The low-volume batch mixing procedure was designed according to EN 196-1 specifications.

Table 2. Mix proportions of the considered mortar mixtures: References and RS1 mixtures

Components / Mixtures	C	S 0/4	RS1	W/C	A/C
R1	1	3	-	0.56	3
R2	1	3	-	0.61	3
RS1-25%-0.56	1	2.25	0.75	0.56	3
RS1-25%-0.61	1	2.25	0.75	0.61	3
RS1-50%-0.61	1	1.5	1.5	0.61	3

Fresh state evaluation of the preliminary mixes

The mortars were evaluated during and after specific mixture sequences regarding the fresh state mixing behaviour (Figure 6). Further on, the fresh state characteristics are determined: the fresh state density (Figure 7, a and b) and the consistency of the materials (Figure 7, c to l). The fresh state density was determined according to EN 1015-6, by weighing the freshly poured material in a container with a predetermined volume and referring to its volume, respectively 1 l.

The consistency of the fresh mortars was determined by the reference method specified by EN 1015-3, namely the spreading mass.

After mixing and fresh state evaluation (Figure 6), the mortars were casted into the 40 x 40 x 160 (mm) prismatic, metallic molds (Figure 8, a), cured at the temperature T (20±1) °C and relative humidity RH (90±5) % for 24 h. The hardened specimens (Figure 8, b) were removed from the molds after 24 hours, visually evaluated and placed in water at the temperature T (20±1)°C, until the considered testing age, respectively 7 days.

Hardened state evaluation of the preliminary mixes

The physical-mechanical performance of the mortars is preliminary evaluated by the bending tensile strength, determined on the 40 x 40 x 160 mm prismatic specimens, at an early age, namely 7 days. The flexural tensile strength and the compressive strength were determined in accordance with EN 1015-11 and EN 196-1. The flexural strength used the three-point bending (3PB) test (Figure 8, c) and the compression was performed on the resulted halve-prisms specimens (Figure 8, d).

RESULTS AND DISCUSSIONS

The visual evaluation of the compositions developed with the RS1 addition indicates an alteration of the workability of the mortar when using the RS1 as sand substitution in the mix.
The workability of the fresh mortar alters considerably when increasing the RS1 substitution percent from 25% to 50%.

Figure 6. Fresh state aspect of the mortars: a) R1; b) R2; c) RS1-25%-0.56; d) RS1-25%-0.61; e) RS1-50%-0.61

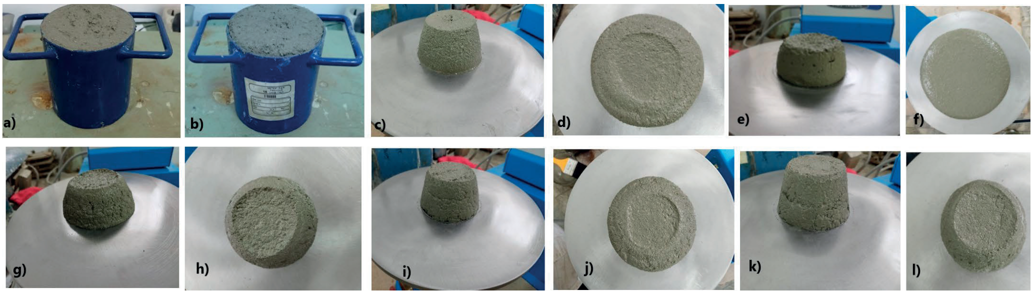

Figure 7. Fresh state characteristics of the mortars: a) and b) determination of fresh state density for RS1-25%-0.56 and RS1-50%-0.61; c) to l) mortar consistency (before and after the shocks): c) and d) R1; e) and f) R2; g) and h) RS1-25%-0.56; i) and j) RS1-25%-0.6; k) and l) RS1-50%-0.61

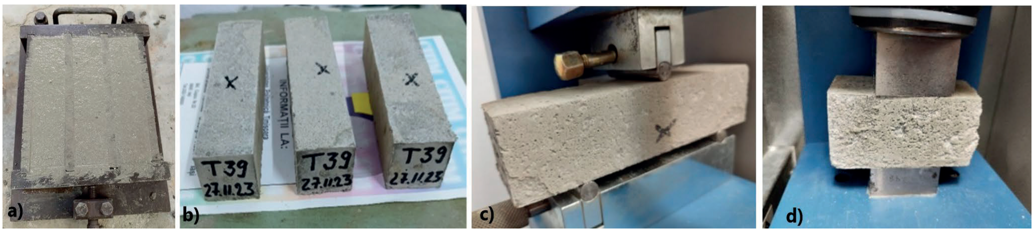

Figure 8. Hardened state characteristics of the mortars: a) and b) specimens' production; c) flexural strength determination by 3PB; d) compressive strength determination on halve-prism specimens

The stiff aspect of the composition RS1-25%-0.56, (A/C ratio = 0.56), similar to the reference composition R1, determines the development of the composition RS1-25%-0.61, with supplementary water addition, respectively A/C ratio = 0.61, and implicitly development of reference R2 with similar A/C ratio, for pertinent performance evaluation. Reference R2, however, presents an extremely fluid consistency (Figure 6, b and Figure 7, e and f), showing early traces of compositional segregation. The composition with high RS1 addition, 50%, was developed with a similar A/C ratio, of 0.61, showing extremely low workability and difficult handling when placed in the prismatic molds (Figure 6, e).

The fresh state determinations lead to the conclusion that the targeted addition, AdT RS1 induces, along with the integration in cementitious compositions, a consistent requirement for mix water increase, in order to regulate the workability. This fact is considered to lead to the deterioration of the mechanical resistances. Therefore, the need to evaluate the RS1 addition, in the field of real densities and masses, as well as the water absorption coefficient, prior to compositional optimization determinations through specific procedures, including addition, is identified.

The apparent fresh state density is determined (Figure 7, a and b) and the results are presented in Table 3.

Table 3. The fresh state apparent density: References and RS1 mixtures

Mixtures	The fresh state apparent density (kg/m^3)
R1	2150
R2	2140
RS1-25%-0.56	2130
RS1-25%-0.61	2120
RS1-50%-0.61	2160

The physical-mechanical performance is evaluated at the early age of 7 days, in order to perform a fast preliminary compositional analysis, namely the RS1 to the cementitious matrix primary compatibility. Also, identifying further research directions and establishing possible compositional adjustments, as the case may be, are targeted objectives of the early-age evaluation.

The results obtained are presented in Table 5, respectively Figure 9 and Figure 10.

The results of consistency determination for the mortars are shown in Table 4 and representative aspects are shown in Figure 7, c to l. The results are confirming the previous conclusions, namely the workability alteration of the mix when introducing/increasing the RS1 addition as sand partial replacement.

Table 4. Fresh state consistency of the mortars: References and SG mixtures

Mixtures	The fresh state consistency (kg/m^3)
R1	170
R2	210
RS1-25%-0.56	120
RS1-25%-0.61	160
RS1-25%-0.61	130

Table 5. Mechanical performances of RS1 compositions at young ages: flexural strength and compressive strength (7 days)

Mixtures	7-day 3PB Flexural resistance		7- day Compression resistance	
	Strength (MPa)	Strength gain/loss vs R1 (%)	Strength (MPa)	Strength gain/loss vs R1 (%)
R1	**7.0**	-14.6	**23.8**	-7.8
R2	5.9	-	22.0	-
RS1-25%-0.56	7.2	3.4	28.5	19.5
RS1-25%-0.61	7.1	1.7	22.8	-4.2
RS1-50%-0.61	6.9	-0.6	22.7	-4.8

The physical and mechanical performances at early ages (7 days), the tensile strength by bending (3PB) and respectively the compressive strength, represent the essential parameters in evaluating the primary compatibility of the considered mineral addition, RS1, to the cementitious matrix. The comparative evaluation of the material performance indicates that the substitution does not induce a strength alteration, but even a slight increase.

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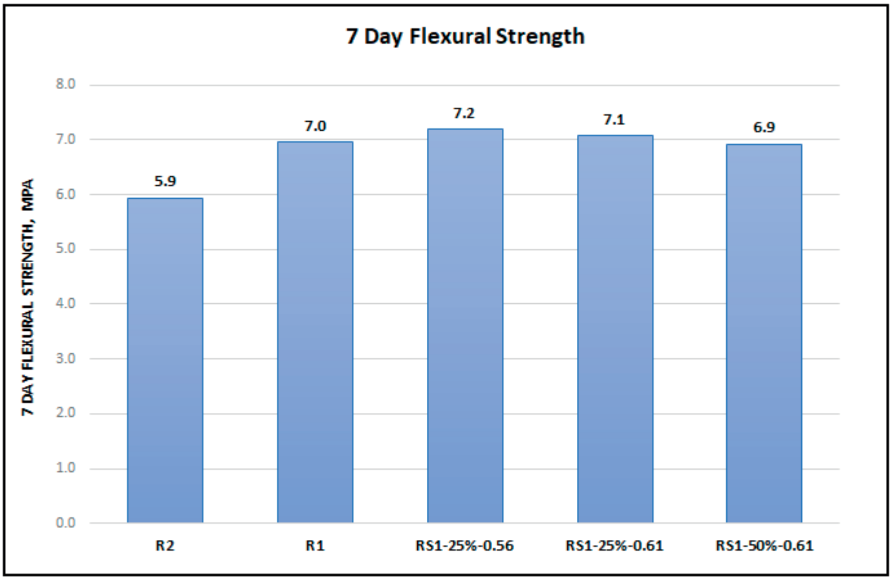
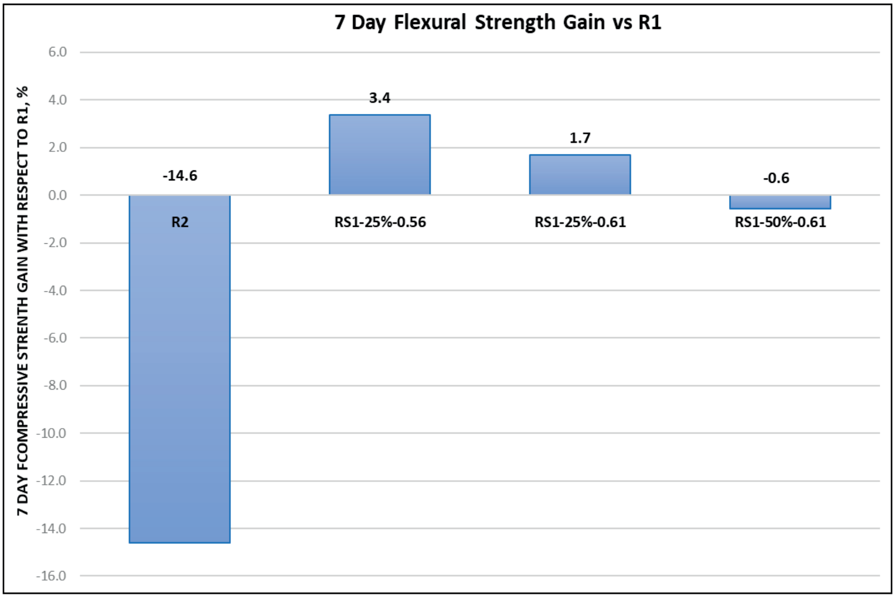
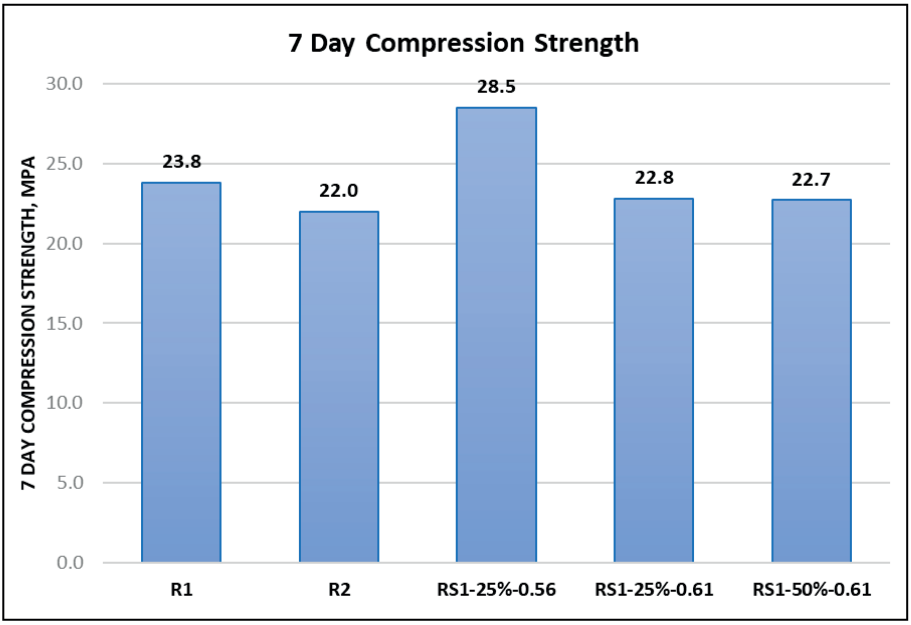


a)

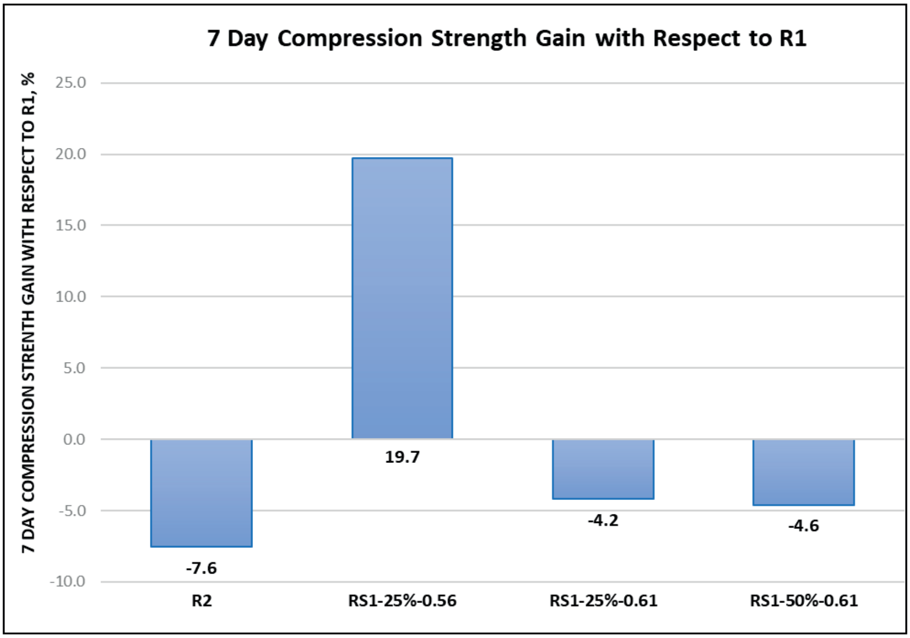


b)

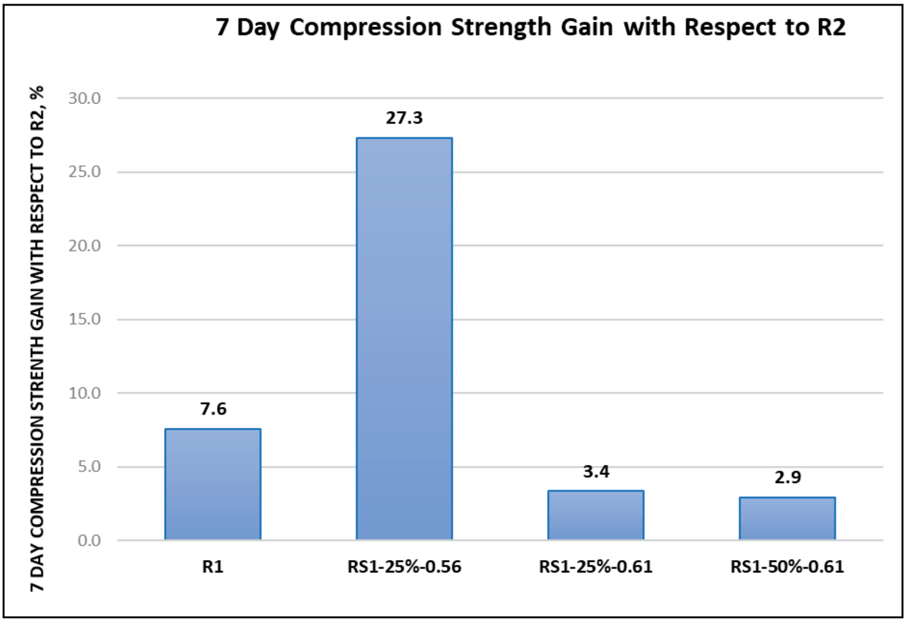
Figure 9. Early age mechanical strength: a) 7-day flexural strength;
b) 7-day flexural strength gain/loss, with respect to R1 (A/C = 0.56)


7 Day Compression Strength
7 DAY COMPRESSION STRENGTH, MPA
30.0
25.0
20.0
15.0
10.0
5.0
0.0
23.8
22.0
28.5
22.8
22.7
R1
R2
RS1-25%-0.56
RS1-25%-0.61
RS1-50%-0.61

a)


7 Day Compression Strength Gain with Respect to R1
7 DAY COMPRESSION STRENTH GAIN WITH RESPECT TO R1, %
25.0
20.0
15.0
10.0
5.0
0.0
-5.0
-10.0
-7.6
19.7
-4.2
-4.6
R2
RS1-25%-0.56
RS1-25%-0.61
RS1-50%-0.61

b)



c)

Figure 10. Early age mechanical strength: a) 7-day compression strength; b) 7-day compression strength gain/loss, with respect to R1 (A/C = 0.56); c) 7-day compression strength gain/loss, with respect to R2 (A/C = 0.61)

CONCLUSIONS

The preliminary results obtained both in the field of flexural tensile strength, as well as in the field of compressive strength, at the age of 7 days, indicate the possible viability of the proposed substitution.

The comparative analysis of the results allows the following synthetic conclusions to be drawn:

- The increase in the water content, respectively the A/C ratio from 0.56 to 0.61 induces, predictably, the alteration of the mechanical performances, both in the control area (R2 vs. R1) and in the case of the compositions with the addition: RS1-25%-0.56 vs. RS1-25%-0.61. The results after 7 days are conclusive in this regard.
- The compression strength results at 7 days indicate the promising potential of the compositional integration of the RS1 material, namely the electric arc furnace slags (EAF), produced and stored in the western part of Romania, in Reșita, Caraș-Severin County, in cementitious materials.
- Additional tests, to confirm/refute this conclusion, are necessary and they represent ongoing research.

Preliminary testing and corresponding results provide encouraging conclusions regarding the viability of the concept. RS1 waste can be positively integrated into cement-based materials, as it proves to be compatible with the cementitious binder system and also with ordinary sand. The initial physical and chemical testing of RS1 as a potential substitute for ordinary 0/4 local sand, together with the physical and mechanical performance of the proposed initial eco-mortars obtained in this preliminary experimental investigation partially confirms the conclusions of recent studies in the subject and also the potential of the Reșita electric arc furnace slags (EAF) for integration into cement-based construction materials. The compositional behaviour developed with an RS1 sample supports the compatibility of the addition with the usual components of cementitious composites, with a high potential for use in construction products.

At the same time, relevant characteristics of mechanical performance and durability of cement-based composites, together with specific tests regarding the evaluation of contamination with potentially harmful materials for the health of the population, of RS1 additions taken from several locations of the Reşita dump, are critical considerations in the evolution of the subsequent scientific approach, in direct correlation with the applicability guidelines for the valorisation of the electric arc furnace slags (EAF) for valorisation in the circular design for eco-intelligent construction products.

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