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Article

Construction materials obtained by valorising mineral waste of red mud and mining tailings - performance and impact

ADRIANA CUCIUREANU*, BOGDAN ADRIAN STANESCU, GEORGIANA CERNICA, GHEORGHE-CRISTIAN SERBANESCU

National Research and Development Institute for Industrial Ecology – ECOIND, 57-73 Drumul Podu Dambovitei Street, district 6, 060652, Bucharest, Romania
**Corresponding author: adriana.cuciureanu@incdecoind.ro*

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Abstract
The actions undertaken by Romania, as an active participant in the sustainable policies of the European Union in the field of waste management, aim to permanently minimize the amount of waste generated, recycling, reuse and higher valorisation to ensure a sustainable management of resources and environmental protection. Increasing the degree of valorisation and obtaining secondary raw materials through the management of waste resulting from mining and metallurgical activities, recognized by very large quantities generated and stored, is the objective of extensive research activities. This paper presents the results of the research carried out in order to valorise red mud and mine tailings wastes by incorporation into construction materials with the main aim of reducing the polluting effect of the waste but also the partial substitution of the basic raw materials from the classic construction materials (cement, clay). The new construction materials (mortars, respectively ceramic materials), by substituting the silico-aluminous component from the basic raw material of the classic construction materials were obtained. The properties of the new construction materials are similarly to the classic materials. The specific pollutants presents in the wastes was retained, which leads to the reduction of the impact on the environment. The impact generated by the implementation of the optimal technological solution was analysed, compared to the impact on the environment due to the storage of the two wastes and the impact produced during the manufacture of classic materials.

Keywords: *red mud, mining tailings, valorisation, impact*

INTRODUCTION
Sustainable development has become a priority for decision-makers all over the world because the impact of humanity on the environment has been greatly accelerated in recent years, as a result of the rapid growth of the population and the concomitant reduction of natural resources [1].
One of the important objectives regarding the transition to a “cleaner and more competitive Europe” is reducing waste and increasing the recycling rate [2]. Sustainable development has a determining component related to environmental issues that represent one of the horizontal policies of the European Union [3]. The worldwide strategy is to minimize the amount of waste, respectively that of natural resources used and to maximize the recycling flow of matter and energy [4].
On spirit of to the circular economy the actions must be concentrated to reusing, refurbishing and recycling existing materials and products. "A resource-efficient Europe" is one of the flagship initiatives of the Europe 2020 strategy, which coordinates actions in many policy areas to ensure sustainable growth and job creation through better use of resources [5].
The JASPERS Final Report from 2017 [6] presents the meaning of the terms used in the specific field of waste: in Directive 2008/98/EC on waste and repealing certain Directives, article 3 paragraph (15) ‘recovery’ means any operation the principal result of which is waste serving a useful purpose by replacing other materials which would otherwise have been used to fulfil a particular function, or

waste being prepared to fulfil that function, in the plant or in the wider economy. Waste recovery brings together aspects related to eco-innovation, ecological responsibility and profitability. By creating new products, it opens up the opportunity to tap into new and emerging markets, create incremental revenue, or reduce the company's operating costs [7].
These were part of the global trends of adopting appropriate waste management solutions in an integrated system, in the spirit of sustainable development. Optimizing the management of waste flows implies the creation of added value. The demand for simple and viable methods of waste management is constantly growing, at an extraordinarily high speed [8÷10]. On the last years the important steps to reducing, recycling wastes from mining and metallurgical industries were crossing, in circular economy context. In Romania, the mining and metallurgical industries are the sectors with the greatest impact attributed to inadequate waste management, generated and stored. Hence the fact that the reduction of adverse effects arising from the development of mining and metallurgical activities requires a holistic approach to waste management.
In this context, the current research on the creation of new secondary raw materials from the processing of industrial waste from the manufacture of alumina (red mud) - a distinct and relevant category for the non-ferrous metallurgy industry and from the processing of metalliferous ores (mining tailings) was included.
The optimal solutions for valorizing these mineral wastes by incorporating them into construction materials were analyzed both from the point of view of the performance of the newly obtained materials (mortar, ceramic materials) and from the point of view of the impact on the environment.

EXPERIMENTAL PART

The red mud has a high content of oxides of the major elements Fe_2O_3 (41%) and Al_2O_3 (25%) and the mining tailings SiO_2 (62%) and Al_2O_3 (19%). Their composition confers increased reactivity on the waste surface [11÷13].
These metal oxides act as activators of the strengthening process of construction materials.
Starting from these considerations, the experimental program aimed at substituting a quantity of the basic raw material - cement or clay - with these mineral wastes. The wastes were constituted as silico-aluminous components in the new construction materials obtained: mortars, respectively ceramic materials.
In a first stage, the two mineral wastes were subjected to preliminary processing, as was presented in our previous study [14]. The wastes were dried in a thin layer in plastic trays for 72 hours at an ambient temperature of 25÷30^{0}C and then were grinded in a vibrating mill with RS 200 discs, at 900 rpm, for 8 seconds, followed by sieving to a material size below 20 µm.
In the second stage, the wastes were characterized. The metals were determined by Energy Dispersive X-ray Fluorescence (EDXRF) using a Rigaku CG analyzer (Japan), equipped with a 50W X-ray source with a Pd anode and Al, Mo, Cu, RX9 secondary targets. The other indicators, (such as: pH, sulphate, chloride, DOC, TDS) were analyzed using electrochemical, gravimetric, and GC analytical techniques.

Experiments for obtaining mortars and testing

The materials (cement and red mud) were weighted on an analytical balance, in different variants, according to preliminary tests: cement (MC); cement substituted with 10% red mud + 10% mining tailings (P1); cement substituted with 20% red mud + 10% mining tailings (P2); cement substituted with 10% red mud + 20% mining tailings (P3); cement substituted with 20% red mud + 20% mining tailings (P4).
The same amount of sand and water was added to each sample. The next step, was to homogenize the materials in a dedicated laboratory mixer. The mixture was added into prismatic molds (40x40x160mm), which was vibrated on a special table to obtain uniform mixtures. The final step was to unwrap the samples for 24 hours after casting and keeping in water at 20^{o}C until the testing time.
In the figure 1 are presented the steps for obtaining the mortars, when the cement was used.

The mortars were tested in terms of consistency of the fresh mortars using the "spreading table" and in terms of bending resistance and compression strength at 28 days ± 8h, using a mechanical press for the bending test, respectively a compression device placed in a compression testing machine [15].

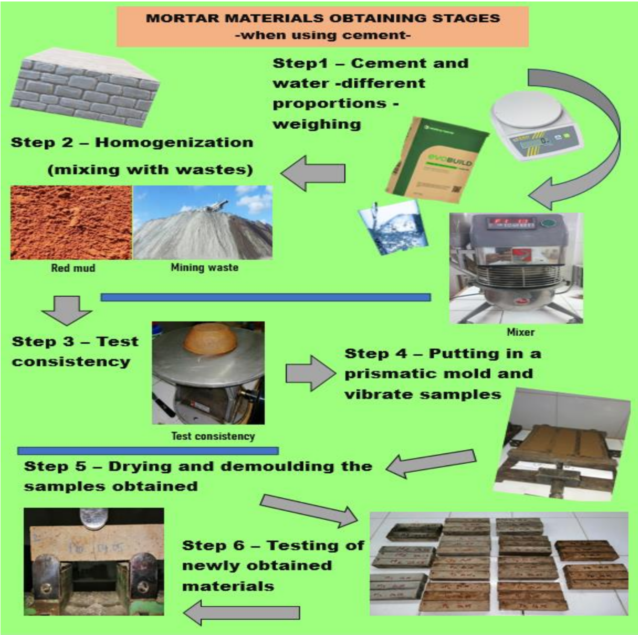

Fig. 1. Experimental steps for obtaining mortar material

Experiments for obtaining ceramic materials and testing

The ceramic materials were weighted in different variants, according to preliminary tests: clay (MA); clay substituted with 5% red mud+5% mining tailings (A1); clay substituted with 15% red mud+15% mining tailings (A2); clay substituted with 5% red mud+5% mining tailings+3% sand (A3); clay substituted with 15% red mud+15% mining tailings+3% sand (A4).

The materials were homogenized manually until reaching an acceptable degree of plasticity and homogeneity. Then, the molds were obtained into a cubical mold (40 mm x 40 mm). The mixture was pressed and removed the excess material. The molds were dryied on air, then in an oven at 60°C and finally at 105°C. Calcination of the samples were performed in an oven at 1000°C for 2 hours.

In figure 2 is presented the diagram for obtaining ceramic materials.

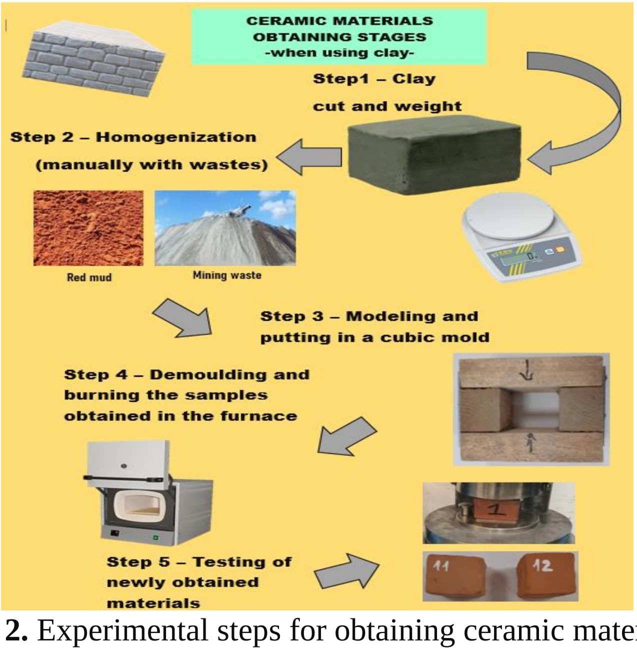

Fig. 2. Experimental steps for obtaining ceramic materials

Testing the polluting potential of the mortars and ceramic materials

The new obtained construction materials, mortars and ceramic types, were tested regarding leaching tests in order to establish the specific pollutants retained in the material matrices. The tests were carried out in a batch system according with a standardized method [16], and the results were compared with maximum values from Order 95/2005 [17].

The eluates resulting from the leaching tests, after filtration, were characterized for metal content using an Agilent 7900 Technologies ICP-MS equipment. The inorganic indicators (chlorine, DOC, TDS) were analyzed using current analytical methods (volumetric, gravimetric).

The impact produced on the environment

The impact generated by the implementation of the optimal technological solution was analyzed, compared to the impact on the environment due to the storage of the two wastes and the impact produced during the manufacture of classic materials.

The methodology considered in the impact analysis was the ”Rapid impact assessment matrix (RIAM)” [18÷21].

The general principle is to assign grades (values) for criteria from group A (A1-importance of the environmental component, A2 - magnitude of change/effect) and from group B (B1 - permanent, B2 - reversibility, B3 - cumulative).

Applying the calculation procedure, it was determined the environmental scores ES, which is the average score for the analyzed factor. ES were converted into impact categories according to data from table 1.

Table 1. Impact categories

Environmental scores (ES)	Categories	Category description
+72 to +108	+E	Major positive changes/impact
+36 to +71	+D	Significant positive changes/impact
+19 to +35	+C	Changes/moderate positive impact
+10 to +18	+B	Changes/positive impact
+1 to +9	+A	Changes/slightly positive impact
0	N	No change/status quo/does not apply
-1 to -9	-A	Insignificant slight negative changes/impact *It does not require specific mitigation measures*
-10 to -18	-B	Changes/negative impact *It requires general and specific reduction measures*
-19 to -35	-C	Changes/moderate negative impact *It requires specific reduction measures*
-36 to -71	-D	Changes/significant negative impact *It requires compensatory measures*
-72 to -108	-E	Changes/major negative impact *It requires compensatory measures*

RESULTS AND DISCUSSION

The mortars obtained in the tested experimental variants are presented in Figure 3.

Fig. 3. The produced mortar materials

The main chemical characteristics of the materials used in the experiments for obtaining the mortars and ceramic materials are presented in table 2 and highlight the following: red mud, cement and clay have an alkaline pH and mining tailings have a slightly acidic pH; Si, Fe, Al, Ca, Na were identified in the highest concentrations (over 10 g/kg dry matter (d.m.)).

Table 2. The main chemical characteristics of the materials used for mortar and ceramic materials

Characteristics	Measure unit	Red mud	Mining tailings	Cement	Clay
pH	pH unit	10.90	4.20	12.99	9.44
Chlorine	mg/kg d.m.	36	43	157	-
Sulphates	mg/kg d.m.	125	126	224	-
Aluminum	mg/kg d.m.	131100	98290	39220	95050
Silicon	mg/kg d.m.	64310	288450	112070	267230
Iron	mg/kg d.m.	286960	31690	27580	46090
Titanium	mg/kg d.m.	13550	4620	2070	4980
Calcium	mg/kg d.m.	43850	1850	381720	60620
Sodium	mg/kg d.m.	72760	6590	23730	9330
Magnesium	mg/kg d.m.	<25	5680	9810	15870
Potassium	mg/kg d.m.	196	68070	10680	27260
Arsenic	mg/kg d.m.	10	217	6	14
Chromium	mg/kg d.m.	1030	128	203	125
Nickel	mg/kg d.m.	35	<25	38	80
Copper	mg/kg d.m.	111	74	163	75
Zinc	mg/kg d.m.	57	1160	840	130
Lead	mg/kg d.m.	34	2650	116	33
Barium	mg/kg d.m.	<25	990	258	306

The variation of the consistency of the mortars obtained in a fresh phase, situated in the range 100 mm to 103 mm (figure 4), does not highlight big differences, regardless of the composition of the mortars. The values are close to those usually found in construction materials: for plaster mortars executed by mechanical processes, the consistency fall in the range 100 mm to120 mm; for solid brick masonry mortars and light concrete blocks, the consistency is situated between 80 mm and 130 mm.

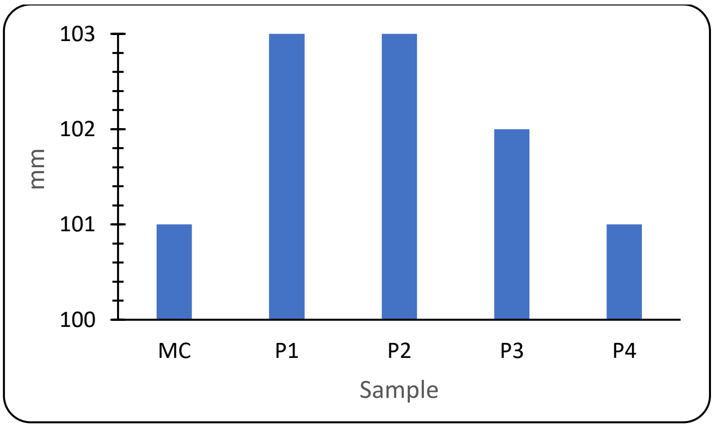

Fig. 4. The consistency of the mortars obtained in a fresh phase (mm)

The results of the strength tests of the new obtained materials, in terms of bending strength and compression strength, are graphically represented in figures 5 and 6.

A good correlation it was observed between the bending strength and the compression strength.

All mortars produced, regardless of the content of mining tailings and red mud introduced, had a compression resistance corresponding to class CS IV (compression resistance $\geq$6 N/mm^2) of plastering mortars according to specific standards.

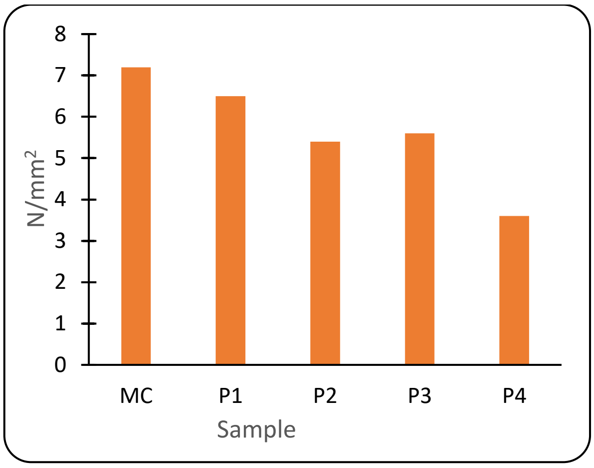

Fig. 5. Bending strength (N/mm^2)

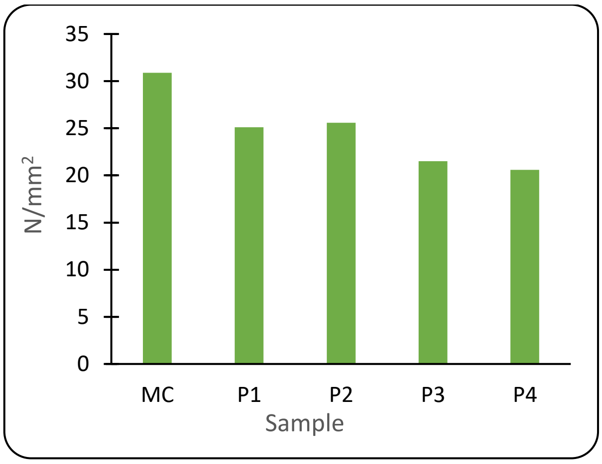

Fig. 6. Compression strength (N/mm^2)

For use as a masonry mortar, depending on the compressive strength, the mortars can be classified as M10 (compressive strength ≥10 N/mm^2) or even higher: M15 (compressive strength ≥15 N/mm^2), M20 (compressive strength ≥20 N/mm^2).
In conclusion, all the obtained and tested mortars can be used for this purpose.
The ceramic materials obtained in the tested experimental variants can be seen in figure 7.

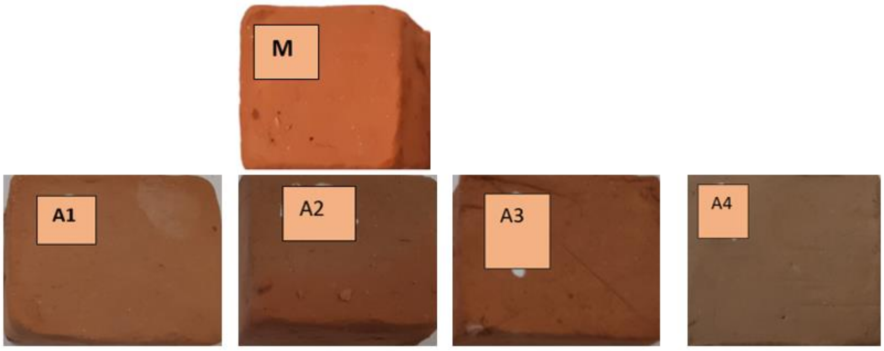

Fig.7. The produced ceramic materials

The results of the compression strength tests of the new ceramic materials obtained, are graphically represented in figure 8.

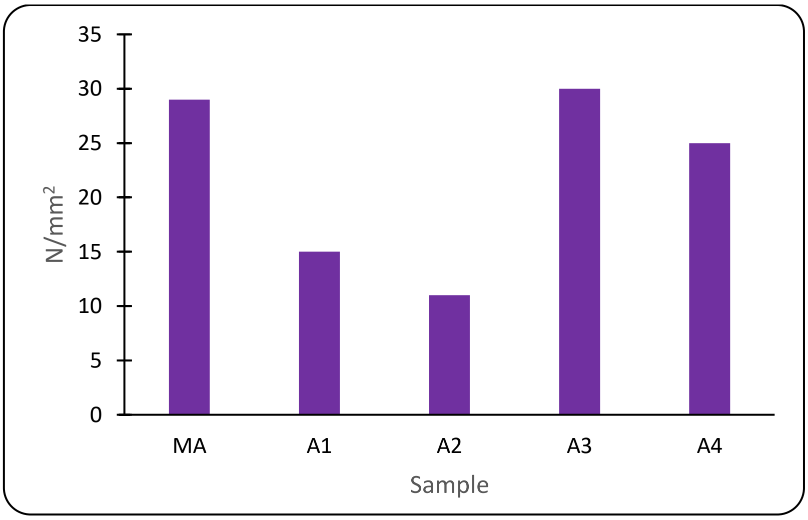

Fig. 8 Compression strength **(**N/mm^2**)**

In figure 8 it can be seen that the addition of sand in the materials component, regardless of the mineral waste concentrations introduced, leads to the doubling of the compressive strength values of the obtained materials.
For all samples of newly obtained ceramic materials, the compressive strength values were above the average values declared acceptable by the manufacturers for masonry elements made of fired clay (minimum 10 N/mm^2).
The tested ceramic masses have values of resistance to appropriate use in high density (HD) and low density (LD) ceramic masonry blocks according to specific standards.

Tests for pollution potential
The characterization of the eluates does not highlight the polluting potential of the new construction materials. The most of the metals were determined in concentrations below the quantification limit of the applied methods. Only chromium presented values in the range of 0.07÷0.4 mg/kg d.m. and copper in the range of 0.1÷0.45 mg/kg d.m. The mineral load expressed as TDS showed values below 3000 mg/kg d.m.
The characteristics of the eluates resulting from the leaching tests carried out on the new created materials highlighted the fact that the specific pollutants of mineral waste were retained very well in the material matrix. This is good advantage concerning environmental point of view.

Quantifying global impact (RIAM) matrix
Based on the quantification of the impact for the relevant factors analyzed, the global impact on the environment is presented.
a) production of materials
The final environmental score is -9 → General impact category (-A): slight negative insignificant impact
b) waste storage
The final environmental score is -7 → General impact category (-A): slight negative insignificant impact
c) the technological solution for waste recovery
The final environmental score is +11 → General impact category (+B): positive impact

As a result of the evaluation carried out, it was found that the proposed technological solution of introducing the two mineral wastes as substitutes for a quantity of the classic construction materials leads to a change in the general impact category from (-A): slight negative insignificant impact (in the current conditions of the impact produced during the manufacture of materials and the storage of waste) to (+B): positive impact (in the conditions of the recovery of waste into new construction materials (figure 9).

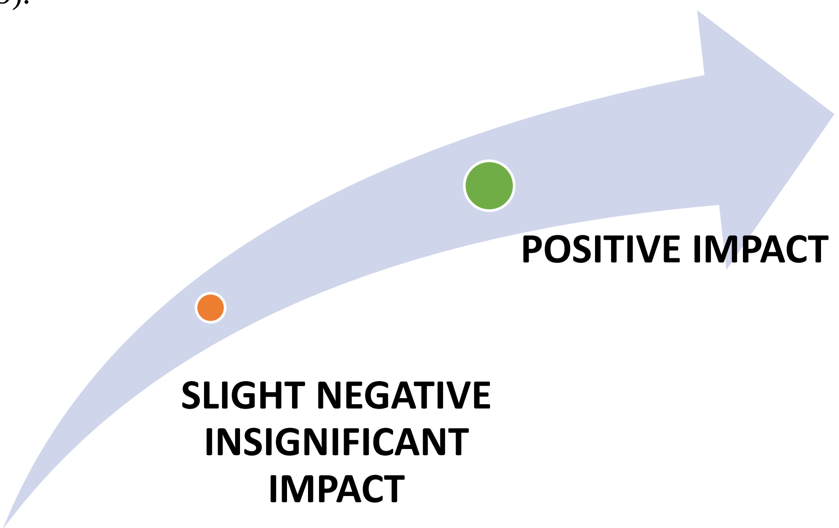

Fig. 9. Change of the environmental impact of wastes / wastes incorporated construction materials

The schematic representation of the analyzed environmental impact regarding materials production, waste storage and waste recovery is presented in Figure 10.

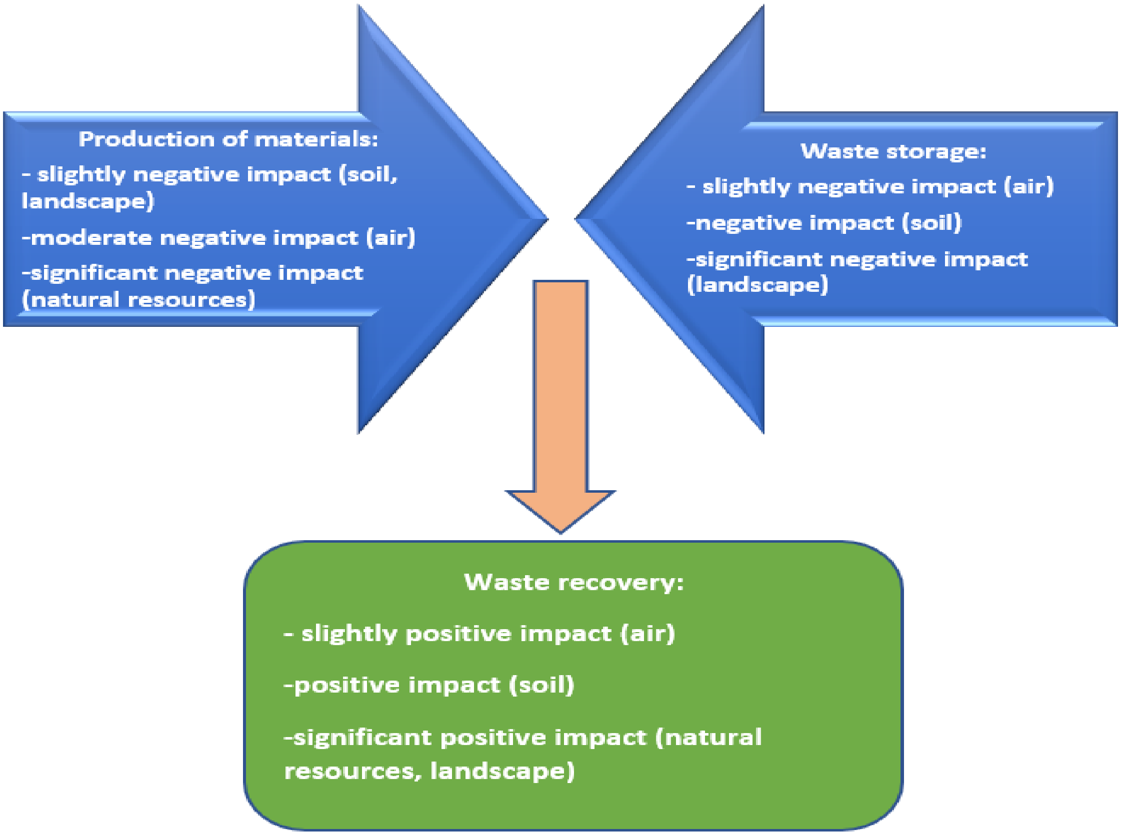

Fig. 10 Scheme of the analyzed environmental impact

CONCLUSIONS

The new construction materials, with the addition of the two mineral wastes, red mud and mine tailings, were presented resistance characteristics close to those made from the classic materials (cement and clay).

The studied mineral wastes could be a good substitute for materials / composite used in classical construction materials, reducing the quantities required to be produced. Implicitly, there will be a reduction in emissions with negative impact on the environment.

The pollutants presented in the mineral wastes were retained in the matrix of the obtained materials.

The impact generated on the environment of the proposed technological solution of recycling red mud waste and mine tailings by embedding in construction materials is beneficial for the environment and constitutes an important step in increasing the level of waste recycling in Romania.

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