

Effective Finishing Composites Using Non-Traditional Natural Raw Materials

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Abstract—One of the current areas of building materials science is the development and implementation of highly efficient facing materials, which can significantly improve the architectural expressiveness of human settlements. Used in modern construction autoclave silicate materials have rather low decorative properties. However, the high whiteness of such wall materials allows them to be dyed with the introduction of colored pigments. It was established that deposits of the initial stage of clay formation processes can be used as energy-saving raw materials for the production of silicate materials of hydrothermal hardening. With the use of the studied materials, it is possible to obtain effective painted high-hollow silicate bricks and stones, as well as cellular concrete, the use of which in construction will improve the comfort of human living.

Keywords— *finishing materials, aluminosilicate raw materials, energy-saving raw materials, comfortable living environment, clay rocks, hydrothermal processing, silicate materials.*

I. INTRODUCTION

For modern construction, the most urgent task is to create a comfortable living environment for humans, which will not only meet housing needs, but also ensure a high quality of life in general. The system «human – material – habitat» is a complex system, the study of which requires the involvement of scientists of various specialties. This line of research is the most important in the 21st century. The creation of a comfortable environment requires the interaction of a whole complex of sciences, starting with building materials science, medicine, architecture, etc., i.e. a transdisciplinary approach.

The system “human-material-habitat” is a very complex open system, the study of which is possible using transdisciplinary science - geonics [1]. Transdisciplinary research is based on the large-scale use and transfer of knowledge, patterns, cognitive schemes from one discipline to another with obtaining emergent properties - properties that individual links (disciplines) do not possess, but they are a result of the integrity of the system, which is especially important for building materials science and creating materials of a new generation [2-4].

One of the important characteristics of building materials are their decorative qualities. These include color, texture of the front surface, and straightness of the faces from the outside. Using various raw materials and additives, you can get wall silicate materials of various colors (white, light cream, red, etc.). Over time, traditional unpainted silicate brick becomes gray because of environmental exposure, alternate moistening and drying, carbonation, ultraviolet, whiteness is lost. Considering all these factors, various pigments are used in the production of silicate bricks; they give the resulting product a clear and saturated color that does not fade in time. An important condition for the use of pigments should be their ability to retain their color at pH > 7 (alkaline environment), also to withstand the conditions of autoclave treatment, resistance to ultraviolet radiation and other external factors.

Of great interest is the use of industrial waste or rock types as coloring pigments. Such approaches for dyeing silicate materials will make it possible to impart the desired color to

the product, and to obtain a material with high performance characteristics. Breeds that were studied in this work have a light brown color. Energy-saving wall materials for green construction based on this raw material are brown and light brown in color.

Considering that natural processes have completed part of the work on the disintegration of rock, it is possible the process of interaction of rock-forming minerals with a binder component not only at high pressure, but also at atmospheric pressure at temperatures up to 100 °C.

II. METHODS AND MATERIALS

For studies, clay rocks, including non-traditional ones, from the Kursk Magnetic Anomaly region were used. We also used quartz sand with a modulus of particle size 1.52 and a SiO₂ content of 94.2 wt. % Burnt crushed calcium lime was used as a lime component.

To obtain materials of dense structure, the raw material mixture was prepared using lime-sand-clay binder, obtained by joint grinding of rock and lime to the required specific surface of 350 m² / kg. Samples were obtained by the method of semi-dry molding at a specific pressing pressure of 20 MPa. The autoclave treatment was carried out at a vapor pressure of 1 MPa according to the mode of 1.5 + 6 + 1.5 h. 2 days after autoclaving, the average density ρ , compressive strength R_{comp} and water absorption W were determined. X-ray studied the phase composition of the neoplasms and thermographic analyzes. According to the non-autoclave technology, the samples were steamed in a steam chamber at a temperature of 95 ° C for 12 hours.

To determine the particle size distribution of the materials, the MicroSizer 201 was used, which allows determining particles with sizes from 0.2 to 600 microns. For the study of the mineralogical composition of raw materials and synthesized tumors used the method of x-ray phase analysis. Studies were performed on the X-ray diffractometer model - ARL X'TRA. Thermo Fisher Scientific.

In addition to X-ray phase analysis, Differential-Thermal Analysis was used to identify the products of tumors and the mineral composition. Studies were performed on a device - Derivatograph Q - 1500 D. For scanning electron microscopy (SEM), a MIRA 3 LM microscope was used.

III. RESULTS

One of the main directions of development of science now is energy saving, rational environmental management, the development of innovative technologies for the production of green composites, which is especially important for building materials [5-7].

The energy intensity of production of the most common wall materials in Russia - ceramic and silicate bricks, expanded clay concrete blocks, etc., is significantly higher than their foreign counterparts.

An urgent task is to reduce the energy intensity of the production of building materials using unconventional,

including man-made materials, whose rock-forming minerals are thermodynamically unstable compounds.

Analysis of the data on the study of the raw material base of autoclaved hardening allowed theoretically substantiating and experimentally confirming the possibility of using the clay phase of the unfinished phase of mineral formation processes instead of sand. Such clay rocks are widespread, as well as in large quantities simultaneously extracted during the mining of ore minerals. The specificity of these rocks is the presence of thermodynamically unstable compounds, such as the imperfect structure of hydromica mixed-layer minerals, finely dispersed weakly rolled quartz, as well as a small amount of montmorillonite and kaolin. The use of such raw materials will allow you to control the synthesis of tumors to produce materials with desired properties [8-9].

Clay rocks have a very diverse mineral composition and properties. In recent decades, using modern research methods (electron microscopy, X-ray diffraction analysis, and infrared spectroscopy), the structures of clay minerals and their properties have been studied in detail. It was found that the elementary layers and the spaces between them in the clay system are nanoscale and have a highly developed active surface [10-12]. If the nanocrystals are separated from each other by physical or chemical means, then a universal modifier is obtained, the distance between the plates of which is about 1 nm [13-15].

The introduction of clay into a lime-sprouts mixture allows one to obtain volume-colored samples. Color intensity increases with increasing clay dosage. With the content of the latter in an amount of 30-40%, the color of the samples reaches the saturation of the color tone of the original clay, and the silicate brick is colored from yellowish to creamy in color.

The obtained experimental data showed (Fig. 1, 2) that with an increase in the content of the studied clays, the tensile strength at compression of the samples first decreases and then increases. This dependence is characteristic of both types of clay and is most clearly manifested in samples containing 8% CaO.

The optimal addition of montmorillonite-hydromica-quartz clay (Fig. 1), corresponding to the maximum strength of the samples, is 40 and 50% with a content of 8 and 4% CaO, respectively. The strength of samples containing 4% CaOact increases from 7.5 to 30 MPa (four times), and samples containing 8% CaOact increases from 20 to 42.8 MPa (twice).

Consequently, the effect achieved with the addition of clay, the higher, and the less lime in the original mixture. Sandy montmorillonite-kaolinite clay increases the strength of silicate materials to a lesser extent than montmorillonite-hydromica-quartz (Fig. 2). The optimal additive corresponding to maximum strength is 20 and 40% with a content of 8 and 4% CaO, respectively.

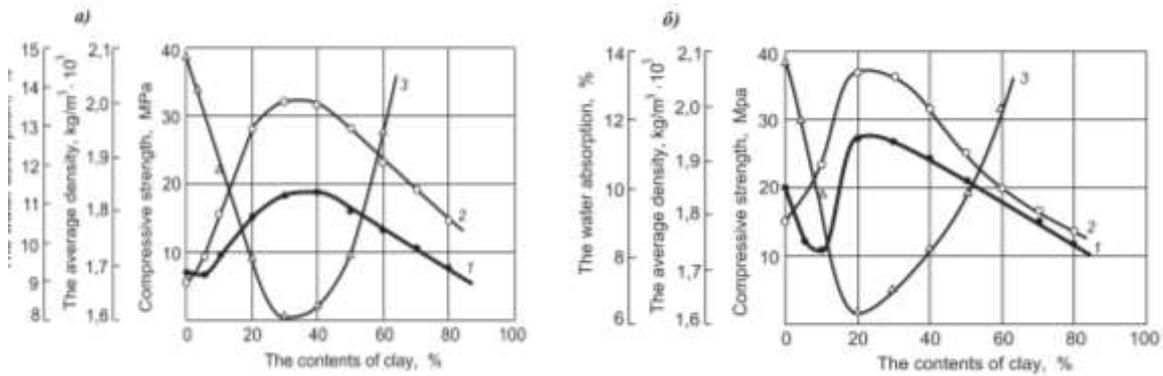


Fig. 1. Physical and mechanical properties of silicate materials, depending on the content of montmorillonite-hydro-mica-quartz clay, the content of active CaO: a; 4%; 6 – 8%; 1 – compressive strength; 2 – average density; 3 – water absorption

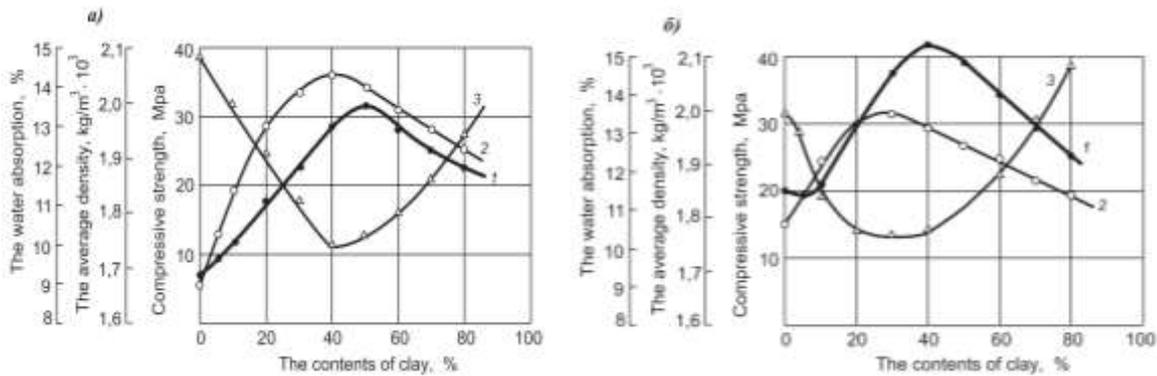


Fig. 2. Physical and mechanical properties of silicate materials, depending on the content of montmorillonite-hydro-mica-quartz clay, the content of active CaO: a; 4%; 6 – 8%; 1 – compressive strength; 2 – average density; 3 – water absorption Physical and mechanical properties of silicate materials depending on the content of sandy montmorillonite-kaolinite clay content of active CaO: a – 4 %; 6 – 8 %; 1 – compressive strength; 2 – average density; 3 – water absorption

The cementing compound of the control samples is represented by calcium hydrosilicates CSH (B) (exo-effect at 835 ° C). In samples with clay additives, along with calcium hydrosilicates, hydrogarnets are formed, the amount of which increases with increasing clay content. The optimal amount of hydrogarnet in samples based on montmorillonite-hydromica-quartz clay corresponds to the maximum increase in strength. With an increase in the content of hydrogranates in samples with sandy kaolin-montmorillonite clay, the strength increases to a lesser extent.

In samples with a content of 5 and 10% of the studied clays (8% CaO), free calcium hydroxide remains (endo-effect at 520 ° C on the thermogram, reflex 2.62 on the roentgenogram), which is associated with an insufficient amount of clay for interaction. As a result, the number of neoplasms decreases and the strength of the samples with montmorillonite-kaolinite sand decreases. With montmorillonite-hydromica-quartz clay in this case, the strength is not reduced.

In mixtures with active CaO 4 and 8% unreacted clay appears in samples with a content of sandy montmorillonite-kaolinite 20 and 30%, montmorillonite-hydromica-quartz 30 and 50%. Therefore, the required clay additive for interaction

with calcium hydroxide is 10% (4% CaO) and 20% (8% CaO) for sandy montmorillonite-kaolinite, 20% (4% CaO) and 40% (8% CaO) for montmorillonite-hydromica-quartz. % CaO).

Samples with 8% CaO have maximum strength with an optimum amount of clay for interaction with CaO. For sandy montmorillonite-kaolinite clay in this case, the highest average density and minimum water absorption of the samples are also achieved.

For materials containing 4% CaO, the greatest strength is achieved with a significantly greater clay additive than is necessary for full interaction with lime. This can probably be explained by the fact that the stoichiometric clay content in this case does not yet provide the densest packing of the material, as well as the formation of the optimal microstructure of the cementing compound.

In Table 1 shows the results of tests for frost resistance of samples based on montmorillonite-hydromica-quartz clay.

TABLE I. FROST RESISTANCE OF SILICATE MATERIALS ON THE BASIS OF MONTMORILLONITE-HYDROSOLISTO-QUARTZ CLAY

№	Composition, wt. %		Loss of strength, % after alternate freezing and thawing, cycles		
	CaO	Clay	15	25	35
1.	4	–	Destroyed		
2.	8	–	25.0	33.56	–
3.	4	30	–	18.47	32.60
4.	4	50	–	17.71	21.77
5.	8	40	–	17.01	23.53

As can be seen from the above data, the frost resistance of samples based on montmorillonite-hydromica-quartz clay is 35 cycles. As mentioned above, in samples with a clay content of 50% (4% CaO), intact clay minerals remain. However, the cold resistance of these samples is higher than that containing 30% clay. This is probably due to lower water absorption and, consequently, less damaging effects of alternate freezing and thawing.

Approbation of the results obtained in the factory confirmed the possibility of volume staining and reducing the energy intensity of the production of silicate brick using clay rocks of the sedimentogenesis zone and the initial stage of lithogenesis.

According to the results of the research, an experimental batch of a volume-colored silicate brick was produced based on lime-clay binder from a lime-sand mass, which is used at the plant and with the addition of clay from sand screenings. Clay in the amount of 20% wt. was added in the form of a lime-clay binder.

The test results showed that the strength of raw materials increased from 0.71 to 1.04 MPa, which will allow producing high-hollow silicate bricks. The compressive strength of brick samples of standard sizes, steamed at isothermal holding time of 4 and 8 hours, increased from 19.2 to 29.4 MPa and 29.1 to 30.4 MPa, respectively. An increase in the steaming time led to a significant increase in the strength of the control bricks (1.5 times), and a slight one based on lime-clay binder (1.04 times).

Thus, on the territory of the Kursk magnetic anomaly, raw materials were discovered, the use of which will allow producing effective high-hollow silicate bricks and facing plates at operating plants. The use of the studied materials will allow at the same time to reduce the time of isothermal exposure or pressure during autoclaving.

To obtain wall materials using a non-autoclave technology, three sandy-argillaceous rocks from deposits of the Kursk Magnetic Anomaly region were used (Table 2).

TABLE II. GRANULOMETRIC COMPOSITION OF SANDY-CLAY ROCKS

Rock	The content of fractions, wt. % size sit, mm					
	More than 0.1	0.1–0.05	0.05–0.04	0.04–0.01	0.01–0.005	Less than 0.005
Sandy loam	15.7	12.90	5.82	42.95	5.70	16.93
Loam № 1	0.55	20.72	18.58	21.15	7.49	31.51
Loam № 2	0.2	9.33	9.56	29.86	9.35	41.70

Clay minerals are represented by Ca2 + montmorillonite, kaolinite, hydromica, mixed-layer formations, and by X-ray amorphous phase.

It was established that the studied rocks are composed of clay-dust aggregates and clay microaggregates, and the composition of the rock contains microaggregates of clay minerals less than 100 nm in size (Fig. 3), which makes it possible to consider these rocks as natural nanoscale raw materials.

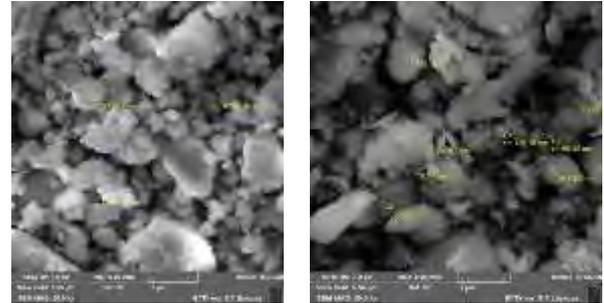


Fig. 3. The microstructure of sandy-clayey rocks, SEM, × 50,000: sandy loam (left), loam 2 (right)

As a binder used lump lime with an active CaO content of 78.3 wt. %. Lime was pre-crushed to a specific surface of 400 m² / kg. Lime and the test rock were thoroughly mixed in a predetermined ratio, moistened with the necessary amount of water and kept in a hermetically sealed cup. After slaking lime from the obtained raw material, samples were molded at a pressing pressure of 10 MPa. The formed samples were subjected to hydrothermal treatment in a steam chamber at a temperature of 90 ± 5 ° C. The results of the experiments are given in table. 3

TABLE III. PHYSICAL AND MECHANICAL CHARACTERISTICS OF SAMPLES

Physical and mechanical characteristics	The content of lime, % by weight of dry mix				
	5	10	15	20	25
Sandy loam based samples					
Compressive strength, MPa	11.50	14.06	11.82	9.59	8.80
Softening coefficient	0.65	0.76	0.77	0.75	0.70
Average density, kg / m ³	1685	1670	1665	1580	1555
Water absorption, %	13.15	13.85	16.56	17.97	18.52
Samples based on loam № 1					
Compressive strength, MPa	9.13	14.38	15.35	16.52	14.19
Softening coefficient	0.68	0.72	0.79	0.74	0.54
Average density, kg / m ³	1715	1655	1660	1600	1555
Water absorption, %	13.85	14.20	15.18	17.32	19.36
Samples based on loam № 2					
Compressive strength, MPa	7.39	13.79	13.02	11.08	11.27
Softening coefficient	0.65	0.72	0.79	0.75	0.88
Average density, kg / m ³	1540	1535	1505	1430	1415
Water absorption, %	14.04	14.26	15.35	17.44	19.89

Comparison of changes in the strength indicators shows that the maximum strength, in almost all cases, the samples reach with a lime content of 10–20 wt. %

The obtained physicochemical indicators of silicate samples indicate that the raw material under study under conditions of steaming at a temperature of 90–95 °C actively interacts with lime. At the same time, physicochemical processes take place that lead to the synthesis of a complex binder, forming a strong framework. Based on the data of differential thermal and X-ray phase analyzes (Fig. 4), the cementing compounds of the obtained samples are represented by slightly crystalline low-base calcium hydrosilicates and hydrogranates.

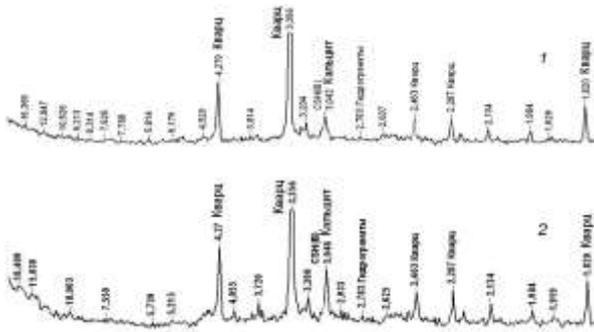


Fig. 4. Radiographs of samples with a lime content of 10 wt. %: 1 - sandy loam; 2 - loam number 2.

The neoplasms of crystalline, low-base calcium hydrosilicates, forming a network, fill the anisotropic pores between the relict structures of the clayey substance, which leads to compaction of the microstructure. In this case, a crystallization structure is formed (Fig. 5).

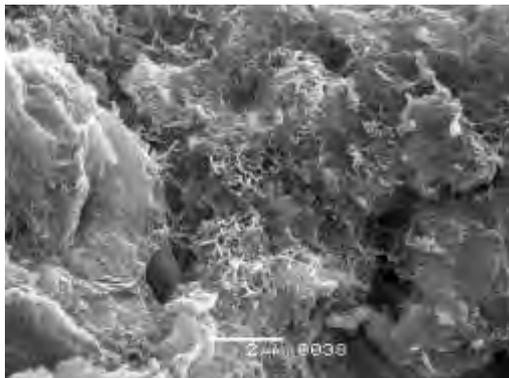


Fig. 5. Radiographs The microstructure of samples based on loam No. 2, samples, SEM, × 9500

The strength of the resulting material structure is significantly affected by the microfill grains, which are sand particles and their contact zone with the ground mass. The rock under study contains mainly non-rolled quartz particles, the surface of which is corroded to varying degrees. These particles of quartz are practically indistinguishable in a dense matrix of tumors of a substance based on lime and the finely dispersed part of the rock. By increasing the packing density, a stronger microstructure of the material is achieved, which gives the products a higher average density and compressive strength.

IV. CONCLUSION

Thus, the use of industrial waste or varieties of rocks for coloring silicate materials as coloring pigments will make it possible to impart the desired color to the product, and also to obtain effective painted high-hollow silicate bricks and stones, finishing materials, cellular concrete and acoustic materials, the use of which in construction will increase the comfort of living person.

The sandy-clay rocks studied are brown in color. The silicate materials obtained on the basis of these rocks acquire a light brown and brown color, which practically does not change with the time of hydrothermal treatment. In addition, under the conditions of hydrothermal treatment without pressure, in contrast to autoclave treatment, the negative effect of temperature on pigments found in sandy-argillaceous rock is reduced.

The operational characteristics of construction materials based on clay rocks of the unfinished lithogenesis stage and heteroporosity at the nano-, micro- and macro-level contribute to the creation of a comfortable habitat in living complexes, which can significantly increase the human life expectancy. Theoretical and practical approaches are to appear.

When unconventional clay rocks are used to synthesize "green" composites, a strong microstructure of the cementing substance is formed with the formation of crystalline low-base calcium hydrosilicates and hydrogranates, which leads to the formation of a strong condensation-crystallization and crystallization structure of the material, which provides high physical and mechanical properties of silicate products.

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References

- [1] A.A. Kuprina, V.S Lesovik, L.H. Zagorodnyk and M.Y. Elistratkin, "Anisotropy of materials properties of natural and man-triggered origin", *Research Journal of Applied Sciences*, Vol. 9 (11), 2014, pp: 816-819
- [2] A.A. Volodchenko, V.S. Lesovik, A.N. Volodchenko and L.H. Zagorodnjuk, "Improving the efficiency of wall materials for «green» building through the use of aluminosilicate raw materials", *International Journal of Applied Engineering Research*, Vol. 10(24), 2015, pp. 45142-45149.
- [3] M.Yu. Elistratkin and M.I.Kozhukhova, "Analysis of the factors of increasing the strength of the non-autoclave aerated concrete", *Construction Materials and Products*, Vol. 1, Issue 1, 2018, pp. 59 – 68.
- [4] T.V. Dmitrieva, V.V. Strokova and A.A.Bezrodnykh, "Influence of the genetic features of soils on the properties of soil-concretes on their basis", *Construction Materials and Products*, Vol. 1, Issue 1, 2018, pp. 69 – 77.
- [5] A.A. Volodchenko, V.S. Lesovik, A.N. Volodchenko, E.S. Glagolev, L.H. Zagorodnjuk and Y.V. Pukharensko, "Composite performance

- improvement based on non-conventional natural and technogenic raw materials”, *International Journal of Pharmacy and Technology*, 2016, Vol. 8(3), pp. 18856-18867.
- [6] L.A. Suleymanova, V.S. Lesovik, K.A. Kara, M.V. Malyukova and K.A. Suleymanov, “Energy-efficient concretes for green construction. Research journal of applied sciences”, Vol. 9 (12), 2014, pp. 1087-1090
- [7] L.A. Suleymanova, V.S. Lesovik, N.P. Lukutsova, K.R. Kondrash and K.A. Suleymanov, “Energy efficient technologies of production and use non-autoclaved aerated concrete”, *International Journal of Applied Engineering Research*, Vol. 10(5), 2015, pp. 12399-12406.
- [8] Danielle Klimesch and Abhi Ray, “Evaluation of phases in a hydrothermally treated CaO-SiO₂-H₂O system”, *Journal of Thermal Analysis and Calorimetry*, Vol. 70(3), 2002, pp. 995-1003.
- [9] S Bernstein, Thomas Karl Fehr, “The formation of 1.13 nm tobermorite under hydrothermal conditions: 1. The influence of quartz grain size within the system CaO-SiO₂-D₂O”, *Progress in Crystal Growth and Characterization of Materials*, Vol. 58 (2-3), 2012, pp. 84–91.
- [10] J. Brozovsky, “Rebound hammer tests of calcium silicate bricks—effects of internal compressive stress on measurement results,” *Applied Mechanics and Materials*, vol. 595, 2014, pp. 155–158.
- [11] Ali Olad , “Polymer / Clay Nanocomposites”, *Advances in Diverse Industrial Applications of Nanocomposites, InTech*, 2011. Pp. 113–138.
- [12] P. Kiliaris and C.D. Papaspyrides, “Polymer layered silicate (clay) nanocomposites: An overview of flame retardancy”, *Progress in Polymer Science*, Vol. 35, 2010, pp. 902–958.
- [13] A. Elamri, K. Abid, S. Dhoub and F. Sakli, “Morphological and Mechanical Properties of Nanoclay Coated Fabric”, *American Journal of Nano Research and Application. Special Issue: Nanocomposites Coating and Manufacturing*, Vol. 3(4-1), 2015, pp. 17-24.
- [14] S.J. Emery, S. Masterson and M.W. Caplehorn, “Sand-clay pindan material in pavements as a structural layer”, In the Proceedings of the 21st ARRB and 11th REAAA Conference. *Transport. Our Highway to a Sustainable Future*, 2003, p. 15.
- [15] A. Elamri, K. Abid, S. Dhoub and F. Sakli, “Morphological and Mechanical Properties of Nanoclay Coated Fabric”, *American Journal of Nano Research and Application. Special Issue: Nanocomposites Coating and Manufacturing*, Vol. 3(4-1), 2015, pp. 17-24.