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Estimated Adhesiveness of Asphaltic Bitumen to Natural Aggregtes Using Statistical Regression

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Abstract — **Usually the adhesiveness is defined as the capacity of a binder to cover an aggregate without dispersing itself when touching the water or the traffic aggressions. Therefore, the adhesiveness additives are products that improve the adhesiveness of the bitumen to a certain aggregate. The improvement of causeway's bitumen adhesiveness is becoming a current practice in our country, especially when is used acid (siliceous) aggregate. The used additives – amino derivatives - have a high stocking stability, a low toxicity degree toward the amine, diamine, polyamine-based additives and are liquid products perfectly compatible with all bitumens and easy to use, in comparison to the paste or solid additives, which must be made liquid to be used. The mineralogyc nature of aggregate is, also, very important because the bitumen „prefers" basic aggregates. However, in practice, the most used aggregates for hot (or cold) coating are acid. There is a continuous problem for specialists to find the optimal methods in increasing the percentage of bitumen capacity of coverage. So, because it is necessary to ensure a connection between the aggregate and asphalt; hence, we use more and more often the process of adding additives to asphalt (doping), namely a small amount of additive (0,1-0,5%). Although the percent of additive is small, the laboratory tests show that adhesiveness increases substantially after they were added. Mathematical regression analysis is using the least squares method to adjust a curve to a set of results. With it can be analyzed how a dependent variable (output) is influenced by the values of one or more independent variables (inputs). In this paper it is used mathematical regression method which defined a polynomial function that has to predict the amount of adhesive intervals where experiments aren`t able to performed. We were able to estimate the optimal values of the amount of adhesive that keeps adhesion greater than 80%.**

Keywords — bitumen, additives, adhesiveness, cover capacity, statistical regression

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I. Introduction

Adhesion is one of the complex phenomena which occur at the bitumen-aggregate contact, some of them being physical or physical – chemical phenomena (as are the texture of the aggregate surface and the porosity, bitumen viscosity and interfacial tension), others being chemical (mineralogical nature of aggregates or the chemical composition of bitumen).

Usually, adhesion is defined as being the capacity of a binder to cover an aggregate without dispersing in contact with water or under traffic aggression [1].

The study of the link between aggregates and bitumen is necessary to increase the lifetime of the roads, highlighting the importance of the mineralogy of the aggregates and also the importance of the force measured at the interface between bitumen and aggregates [2].

The adhesion agents must be capable of changing the superficial tension aggregate-bitumen and to allow the aggregate to be covered by bitumen. They also enhance the resistance of the binder to the destructive action of water.

Aggregates, which have on their surface an adsorbed water layer, have an electric charge due to the ionization of its molecules. The type of charge, positive or negative, allows their classification into acid aggregates or basic aggregates.

The acid aggregates (quartzite, granite, dacite, porphyry) are mostly silicates that have on their surface $SiO3^{2-}$ anions. The basic aggregates (limestone) are mostly carbonates that in the presence of water form Ca^{2+} cations. The natural silicates usually come from metamorphic or igneous rocks. The number of silicates in nature is very large and their structure very complex [3].

For adhesion determinations was used dacites as aggregates, which are neo-volcanic rocks, corresponding to granodiorites, with porphiric structure, massive texture and the following mineralogical composition: feldspars (20-35%), quartz (5-10%), hornblende (2-10%), biotite (2-5%), opaque minerals – magnetite (~1%), paste with hyalopilitic, pilotaxitic structure, made of small fragments of feldspars, quartz and glass (40-60%). The apparent density is 2600 kg/m3 and the compressive strength (in dry state): 160 N/mm2. Their color varies between grey and brown. They are slightly acidic or neutral [4].

Bitumen contains polar compounds like asphaltenes and resins. In these complex molecules, the presence of some acid groups leads to negative electric charge when water comes in contact with bitumen surface. These acid molecules locate themselves in the only possible direction, towards the surface, due to the acid group which is hydrophilic, covering the bitumen particles with a negative electric charge [5].

Using this simple concept, it will be easier to understand the fact that when bitumen comes in contact with a basic aggregate, an electrostatic attraction force is created between the ions with different electric charges on both surfaces, thus adhesion of a certain degree being created.

On the other hand, when bitumen comes in contact with a silica aggregate, the negative ions on both surfaces generate rejection hindering adhesion. Adhesion agents provide positive charge on the bitumen surface allowing the electrostatic binding with the negative silica aggregates, improving adhesion.

The additives used are structures of amidoamines, ethyleneamine-imidazolines and ethanol-amine-imidazolines. These display a high degree of storage stability and a low degree of toxicity comparing to the additives based on amines, diamines and polyamines and are liquid products perfectly compatible with all bitumens, easy to use comparing to solid or paste additives that must be brought in a liquid state in order to use them.

These additives are surfactant products with particularly polar composition and structure, being made of a lyophilic group which fixes itself on the bitumen and a hydrophilic group which fixes itself on the surface of the natural aggregate (Figure 1).

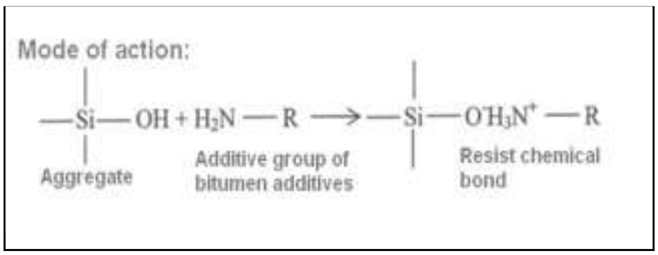

Figure 1. The way that the additive acts on the aggregate

There is no need for large doses of additives (usually the dosage values are between 0.1 and 0.5%). It has been noticed that exceeding the maximum used value does not lead to significant improvement of adhesion (on the contrary, sometimes when the dosage is raised unaccountably, a standstill or even decrease of adhesion value occurs).

In this paper was used mathematical regression to determine a function that will estimate the optimal amount of additive that makes adhesion to improve considerably.

II. Experimental part

In table 1, there are the additive percentages for each bitumen, the type of aggregate used (the same type of aggregate was used – dacite) as well as the adhesion values calculated through the classical method also called "The quantitative method for determining adhesion" [6].

The quantitative method consists in determining the adhesion of bitumen to the natural aggregate, calculated through the value of the adsorption of a pigment from a solution of given concentration by the bitumen coated aggregate (the pigment not being absorbed by the bitumen).

TABLE I. THE QUANTITATIVE ADHESION VERSUS ADDITIVE PERCENTAGES FOR EACH BITUMEN

Nr. of sample	The quantitative adhesion (%)	Bitumen (+ Additive)	Aggregate
1	38,4	Bitumen 70/100	Dacite
2	86,08	Bitumen 70/100 +0,3%Additive	Dacite
3	83,05	Bitumen70/100 +0,4%Additive	Dacite
4	79,84	Bitumen70/100 +0,2%Additive	Dacite
5	74,8	Bitumen70/100 +0,1%Additive	Dacite

The samples have processed effectively according to Romanian Normative [6]. It weighed 250 g of 5/8 sort natural aggregate in a porcelain capsule and 12,5 g bitumen in another capsule and introduced the capsules in the stove and heated them at 140^{o} C, thus it used a D70/100 bitumen for all samples.

After heating, it has introduced 250 g natural aggregate in the bitumen capsule and energetically mixed with a spittle until it became homogeneous. The capsule has took off the balneum and continued mixing until the bitumen film stopped dribbling from the natural aggregate surface.

The mixture was put in a Berzelius glass, covered with a watch glass and let it chill to the room temperature, for one hour. After chilling, it has poured 20-25^{o} C distilled water onto the composition. The water level should exceed the composition level with 7/8 cm. The glass is covered again and left to rest at the room temperature for 24 hours.

In the glass container of the re-circulating device was put the sifter 1/3 away from the container bottom and put the 250 g of 5/8 sort natural aggregate sample on the sifter. After that has added 1300 cm^3 Brilliant Red colourant solution of the set concentration (C) and started the recirculation (the recirculation lasts for 90 minutes for one sample). The operation has repeated as well for the aggregate samples covered with bitumen.

The sampled 20 cm^2 recirculated colourant solution that had the C_1 concentration for the non-wetted aggregate and the C_2 concentration for the bitumen filmed aggregate.

The sampled colourant solutions are separately analyzed specter-photometrically. The extinctions and the value of the C_1 and C_2 concentrations of the solutions are determined from the calibration plot.

Bitumen adhesion (Sn) is calculated using the formula:

$$S_n = 100 - \frac{C - C_2}{C - C_1} \cdot 100 \quad [\%] \qquad (1)$$

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where:

- C = initial concentration of the solution (percentage);
- C1 = solution concentration after its recirculation over the natural aggregate as it is (percentage);
- C2 = solution concentration after its recirculation over the natural aggregate coated with bitumen (percentage).
- Bitumen adhesion to the natural aggregate is considered to be adequate if its value (Sn) is at least 80%.

III. Statistical regression

The analysis through mathematical regression [8] uses the method of the smallest squares to adjust a curve to a set of results. Using this method we can analyze how a dependent variable called answer or output, is influenced by the values of one or more independent variables, called input data or parameters/ factors that can influence the result.

Regardless the type of regression used (linear or non-linear) to determine the approximation degree of the mathematical equation achieved through this method, usually two statistical indicators are used:

R^2 – called determination coefficient that shows how close to the reality of the experimental data the mathematical regression equation is.

Adjusted R^2 – called adjusted determination coefficient, its value being corrected in order to take into account factors as *N* (the total number of factors) and k (the numbers of factors included in the equation), allowing thus comparisons between more regression equations.

These indicators have values between 0 and 1. The better the degree of approximation of the equation, the closer to 1 the value of these coefficients gets. If they equal 1, then all the points of the dependent variable are located on the regression curve – the ideal case. This means that the variation of the dependent variable *Y* is very well explained by the variation of the independent variables X_i. If the regression equation is given by the equation of a straight line (2), we can say that the regression is linear.

$$Y = a \cdot X + b \qquad (2)$$

where a is the straight line slope and b is the intersection with the vertical axis, and they are called linear regression coefficients. The slope of this line also shows us the intensity of the relation between the dependent variable and the independent ones.

Mathematical regression plays an essential part in predicting the dependent variable *Y* in relation to the independent variables, on configurations of those which are not predefined but which are to be located in the defining space of the independent variables X_i.

Mathematical regression can be used to:

- Predict the value of the dependent variable in relation to the independent variable;
- Analyze the relation between the variables;
- Determine the function that defines the relation between the independent variables and the dependent one.
- Optimizing the processes using mathematical regression

The process of optimization [9] is a process of searching among the variables of a system for the combination of parameters that fulfills the operator’s requirements for a certain output value.

Using regression analysis, a polynomial function called objective function *f*(*x*) can be used to anticipate the behavior of a system, and that can be used further in the optimization process. With the help of this method, we can anticipate optimum values even outside the study field.

The general formation of a conventional optimization is expressed as follows:

$$\text{Maximize or Minimize:} \quad f(x) \qquad (3)$$

$$\text{Subjected to:} \quad hi(x) = 0,\ i = 1, p \qquad (4)$$

$$gj\ (x) \le 0,\ j = 1, m \qquad (5)$$

where: *f(x)* is the objective function, *hi(x)* is the equality constraint function, *p* is the number of constraints, *gj(x)* is the inequality constraint function and m is the number of constraints.

IV. Results and discussions

The highest values were achieved for the samples in which bitumen with additives was used [7]. One can notice that for the bitumen with no additives, in most cases, we cannot achieve the required value of 80% for adhesion, hence the conclusion that additive adding to bitumen becomes a necessity and not just another option.

As indicated in the table 1, the results – except for the first sample – fit the ± 5% deviation foreseen by Romanian Standard [6].

There are differences between the sample that contain pure bitumen and those for which additived bitumen was used (adhesiveness improvement is visible, regardless of the used method).

Using mathematical regression for the adhesion values achieved in relation to the additive percentage, we got a curve given by the regression equation:

$$F(X) = -1{,}74X^3 + 11{,}07X^2 - 15{,}96X + 81{,}43 \qquad (6)$$

where *X* is the additive percentage.

This curve is fitted in figure 2. The approximation of the curve is indicated by the value of R=1, which indicates a very good approximation.

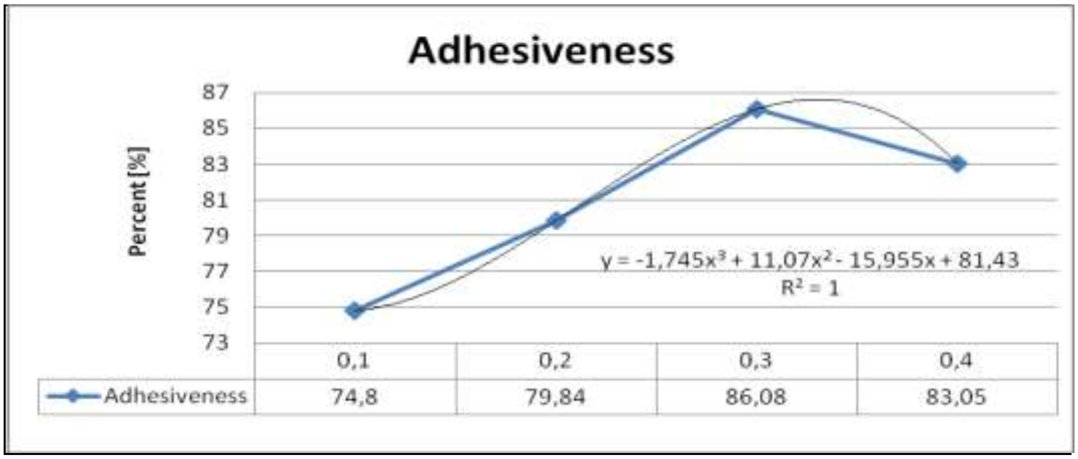

Figure 2. Adhesivness curve obtained by statistical regression

Using this function *F(X)*, we determined the optimum values of the additive percentage which maximize bitumen adhesiveness. We infer from this optimization that in order to get more than 80% adhesion, we must introduce a percentage of additives between 0.21% and 0.42% (Figure 3).

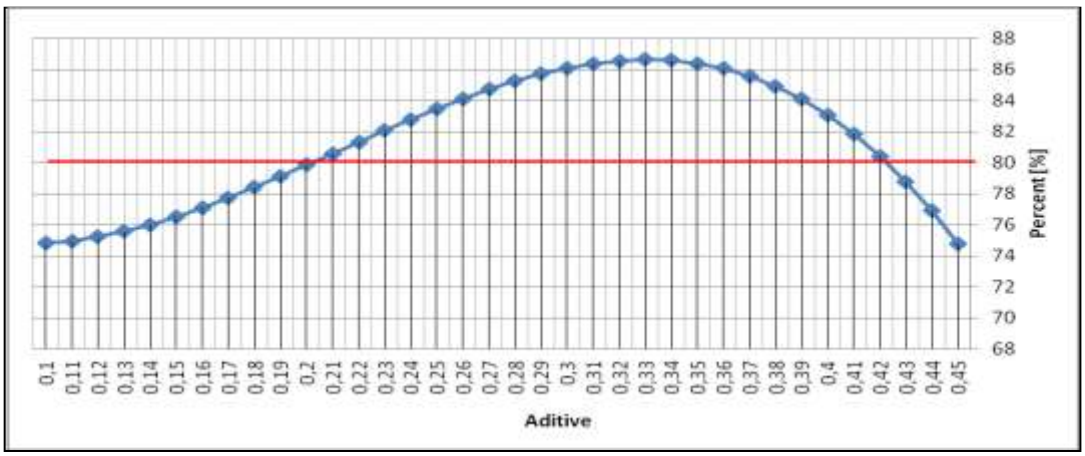

Figure 3. Optimal values of aditive

V. Conclusions

The adhesion of bitumen to aggregates can be improved through using adhesion additives that are able to change the interfacial conditions between the aggregate and bitumen in such a way that the bitumen adheres even to the aggregates that are difficult to cover. They, also, are able to enhance the adhesive bind between the aggregates and the bitumen, thus increasing the long term resistance to water's destructive action on aggregates.

Using mathematical regression we obtained a feature that helped us to estimate and then to optimize the amount of adhesive. Using this function we were able to determine the optimum amount of additive (ranging between 0.21% and 0.42% values) that provides adhesion located at around 80%.

The adhesiveness between bitumen and natural aggregates is a decisive factor for the resistance of the road. Very often, the problems appeared on the road surface may be attributed to the inadequate adhesiveness between bitumen and natural aggregates.

The achieved results are iterative and reproducible and allow the elimination of a certain number of parameters which distort the covering power of bitumens (in case of hot-applied coatings) or emulsions (in case of cold-applied coatings).

References

[1] D. Pavol, H. Marcela, V. Jozef, Methods for determination of affinity between aggregate and bitumen - June 13, 2011, [45th International Petroleum Conference, June 13, 2011, Bratislava, Slovak Republic]

[2] M. Horgnies, E.Darque-Ceretti, H.Fezai, E.Felder - Influence of the interfacial composition on the adhesion between aggregates and bitumen: Investigations by EDX, XPS and peel tests - International Journal of Adhesion & Adhesives, Vol.31 , 2011, pp: 238–247.

[3] J. Read and D. Whiteoak, *The Shell Bitumen Handbook* – Thomas Telford Publishing, 2003.

[4] R. Moraes, R. Velasquez, H. Bahia- Measuring effect of moisture on asphalt-aggregate bond with the bitumen bond strength test - Tansportation Research Board Annual Meeting, January, Washington, D.C., 2011, pp. 23-27 .

[5] M. Hugener, F. Beltzung, C. Angst - Affinity between bitumen and aggregates: improvement of the en static test method - 13-15th June 2012, Istanbul [5th Eurasphalt & Eurobitume Congress]

[6] Romanian Standard 10969/3-83: Adhesiveness of bitumen for roads to natural aggregates – quantitative determination method – replaced by SR 10969:2007

[7] M. Musteaţă – Research topics – Pollchimic Laboratory of Giurgiu, 2010.

[8] Montgomery Douglas C. 2001, Design and analysis of exper-iments. – 5th edition by Wiley & Sons, Inc., New York.

[9] Lepadatu D. et al. Optimization of springback in bend-ing process using FEM and Response Surface Methodolo-gy, International Journal of Advanced Manufacturing Technology, vol. 27, 2005, pp. 40-47.

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