LABORATORY INVESTIGATION ON ADHESION PROPERTIES AND WATER SUSCEPTIBILITY OF BITUMEN-AGGREGATE SYSTEMS

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Abstract: Challenges concerning pavement engineering are nowadays mainly addressed to a low-cost and time-saving preservation of infrastructures. The need for innovative materials and technologies, combined with the necessity of guaranteeing adequate in-service performance, must face the growing distresses of pavements. Moreover, several works are not properly executed, due to inappropriate materials with not well-known properties. To this regard, the present paper focuses the microscopic mechanisms developed at the bitumen-aggregate interface in order to fully understand the bonding and adhesion factors that affect the asphalt concrete serviceability. Experimental tests were carried out to investigate surface texture of different kind of aggregates and its correlation with aggregate-bitumen adhesion strength. The external surface of aggregate was characterized in terms of microscopic structure using a 2D contact profilometer; then the bonding with different bitumens, also in the presence of water, was investigated through boiling water and peel tests. A 50/70 pen bitumen was selected to assess the adhesion characteristics with limestone, porphyry, basalt and steel slag aggregate. Samples were prepared both with smooth and rough aggregate surfaces (in order to simulate different physical conditions) and were characterized in dry and wet conditions. Experimental findings showed a strong correlation between the bitumen-aggregate adhesion and the different kind of tested aggregates. Such property seemed to be also influenced by surface conditions and bitumen-aggregate blending processes. In particular, bitumen-aggregate systems prepared with limestone and basalt manifested good water resistance, whereas the use of steel slag and porphyry led to worse performance, also according to roughness characteristics.

Keywords: adhesion, bitumen, aggregate, water resistance, surface roughness, contact profilometer.

1. Introduction and State of the Art

The construction of flexible pavements for road or airport infrastructures and the design of asphalt mixtures generally occur after specific project steps and material characterization. Once laid and compacted, asphalt concrete must ensure adequate in-service performance and durability, safety and functionality.

Actually, few transport infrastructures are now exhibiting critical behavior due to a bad design and construction; however, asphalt pavements are often subject to early deterioration.

A cautious design of pavement, correctly based on multiple factors such as traffic, climate, etc., requires the adequate knowledge of the materials' characteristics and their working mechanism. To this purpose, one of the most important aspects interesting asphalt mixtures is linked to the interaction between the binder (bitumen) and the lithic matrix (aggregate). Cohesion and adhesion constitute the principal actors determining the bitumen-aggregate affinity. "Cohesion" refers to the force linking the single molecular particles of bitumen. The forces connecting external particles of bitumen and aggregates is named "adhesion". The evaluation and the understanding of these forces and factors is a valid instrument for a proper mix-design of asphalt concrete (as example, for the choice of compatible types of bitumen and aggregates) aimed at durable asphalt pavements. In order to better understand these mechanisms, several tests and protocols have been implemented. Bitumen-aggregate adhesion depends on certain factors difficult to be controlled, above all during laboratory testing. Indeed, aggregate properties deeply influence the bond behavior with bitumen; chemical composition, surface texture (roughness and porosity), shape and size, external impurities affect aggregate characteristics and adhesion with binders. Also bitumen properties (as examples viscosity and chemical structure) impact on the adhesion forces. Bitumen content in asphalt mixtures is another important aspect, since binder film thickness (greater or smaller depending on the quantity) which cover aggregates contributes to the bonding strength. Bitumen modification (rheological alteration or polymeric addition) could add other sources of variability in adhesion. In addition, considerable external factors which interest this interaction have to be accounted: e.g. water presence, temperature, traffic (typology and intensity), human behavior (Graf, 1986). Quite a few researchers (Hefer et al., 2005; Masad et al., 2005) faced the problem, proposing different theoretical approach to explain the behavior of bitumen-aggregate link (e.g. theory of boundary layers, mechanical theory, chemical bonding theory, etc.). Humidity represents a crucial element because the presence of water causes the adhesion weakening and the possible stripping of the aggregate grains. Goel & Sachdeva (2016) published a study concerning many arguments about stripping phenomena within asphalt mixtures, defining the dynamic of stripping, its main causes and the specific tests required to its investigation. Also Pasetto et al. (2017) dealt with stripping and bitumen-aggregate affinity evaluating water susceptibility at mastic-scale and at mixture-scale with two different procedures. Literature reports multiple researches oriented to the appraising of affinity in the case of many aggregate types: Cui et al. (2014), Zhang et al. (2015) Paliukaité et al. (2016) and Pasetto et al. (2017) worked with limestone, marble, granite and steel slags, different kinds of bitumens and some adhesion promoters. Each of them concluded that promoters guaranteed sensible improvements in the adhesion quality (limestone and marble, characterized by similar chemical composition, resulted more basic with respect to granite and owned a better bitumen affinity – oppositely, more acid granite and steel slags displayed worse bonding attitude). Various studies (Lottman, 1982; Voskuilen et al., 1996; Airey & Choi, 2002; Zhang et al., 2017;

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Pasetto et al., 2017) were based on standardized testing methods; however, specific procedures addressed to the measurement of adhesion forces were generally lacking. Qualitative tests (Static-Immersion test, Rolling bottle test, Boiling water test, etc.) for the assessment of the adhesion degree were developed, and calculated mainly the residual bitumen percentage after tests, compared with standard prescriptions of standardized quantities (PATTI test, pull-off test, peel test, etc.); in certain cases, stress needed for the adhesion breaking was calculated.

2. Objectives, Experimental Set-Up and Tests

Since the surface texture and the wettability of aggregate are believed two of the principal factors influencing adhesion force, the objective of the present paper concerns the development of specific tests useful to evaluate such characteristics on different kinds of aggregate.
The secondary target of the work regards the behavior description of marginal materials (steel slags and secondary materials derived from earlier processes) able to ensure benefits related to environmental and economical savings (avoiding quarrying activities and use of natural aggregate). Depending on the aggregate type and shape, pull-off and peel tests were planned. Developed procedures differed from common utilized methods (which involve smooth-surfaced and regular-shaped grains) allowing the utilization of "not-treated" aggregates, preventing possible alterations in the natural surface texture and bonding attitude. Tests were carried out in different conditions to evaluate the effect on adhesion functionally to: aggregate nature, hygrometric status and surface texture. Such characteristics (described in the following paragraphs) are summarized in Table 1. Four different types of aggregate were employed: limestone, basalt, porphyry and steel slag. The binder consisted of a traditional bitumen, with a 50/70 dmm penetration grade. Two different hygrometric conditions were considered: a dry condition, based on the natural ambient humidity; a wet condition, based on aggregate soaking (before the binder application) for two days, that represents the needed time for the complete saturation of surface permeable voids. Surface textures were varied: for each aggregate type, smooth surfaces (by blade sawing) or rough surfaces (without treating aggregates) were prepared. Only in the case of steel slag, it resulted impossible to obtain smooth surfaces because of some issues encountered during the mechanical cutting. Experimental activities were structured in successive stages: firstly, surface textures of smoothed and rough aggregates were characterized through the high-resolution surface profiler, then wettability tests were executed on aggregate-bitumen blends. Further, stripping tests and peel tests (for the evaluation of adhesion force) were performed (Table 1).

Table 1
Experimental set-up

Aggregate type	Surface type	Dry condition	Wet condition
Limestone	smooth	X	X
Limestone	rough	X	X
Porphyry	smooth	X	X
Porphyry	rough	X	X
Basalt	smooth	X	X
Basalt	rough	X	X
Steel slag	rough	X	X

The high-resolution surface profiler consists in an equipment (Figure 1) for the determination of surface texture of materials, generally named as roughness. It is mainly composed of a high-sensible extremity, able to translate in the three principal directions (x, y, z) in order to record the roughness of the tested surface (a transducer converts movements in electrical signals). The used equipment owned a high-resolution interval of 16,000,000:1. The stroke of the extremity used in the work was equal to 15 millimeters either for smooth or rough tested aggregates. Digital surface reconstruction and 3D representation (Figure 2) required a sampling area of about 9 mm^2 (3 mm long on the x and the y axes). Two different cut-offs (i.e. the wavelength of the filter for the signals recording) were then imposed in the equipment: the first one equal to 2.5 mm for the analysis of greater ripples (regarding only some wavelengths), the second one equal to 0.8 mm for a more detailed evaluation. R_a, R_{sk}, R_{ku} and R_{sm} were the determined parameters. R_a is the arithmetic average of absolute distances of the roughness profile with respect to the mean line (line calculated with the least squares method, balancing the areas below and above the line and minimizing their separation). R_{sk} is the measure of the density function asymmetry referred to the mean line (in the case of R_{sk} = 0, the roughness profile coincides with a Gaussian distribution; when $R_{sk}<0$ a quasi-linear profile is obtained; if $R_{sk}>0$ several peaks can be detected). R_{ku} represents the profile peak density (if R_{ku} = 3 the profile follows the Gaussian distribution, if R_{ku} > 3 a rounded profile is detected, etc.). R_{sm} is the average distance between profile peaks above the mean line (peaks are the highest points in the profile, which separate a rising from a descendant segment).

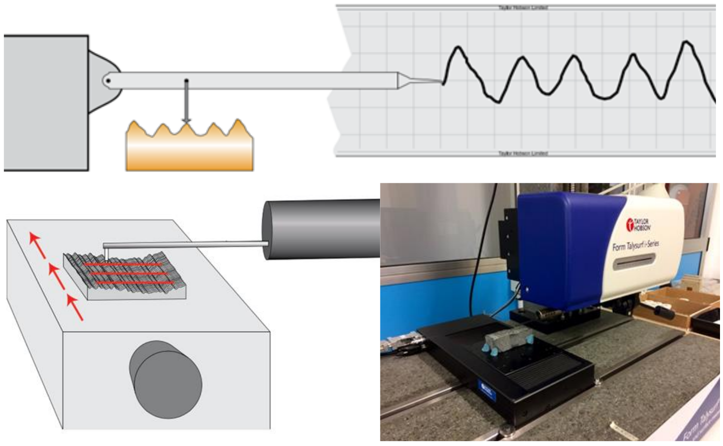

Fig. 1.
High-resolution surface profiler: equipment (right) and instrument functioning (left)

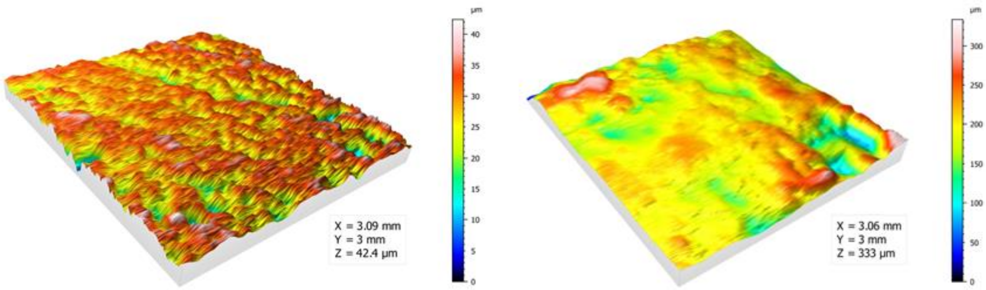

Fig. 2.
High-resolution surface profiler. Examples of 3D image: smoothed (left) and rough (right) limestone surfaces

The evaluation of the bitumen-aggregate adhesion in dry and wet conditions, as well as the analysis of wettability of aggregates by means of the Ancona Stripping Test (AST) proposed by Bocci and Colagrande (1993) were planned. The last is also adopted by EN 12697-11, except for the way of assessing final aggregate bitumen coverage. The procedure is based on the preparation of a blend of aggregate (60 g) and bitumen (3 g) mixed at temperature of 160 °C (Figure 3). When steel slags were used, proportions were changed using 75 g of aggregate in order to consider its higher specific weight and maintain an analogue blend coverage (volumetric properties). For AST, blends were placed in a 600 ml beaker with 200 ml of distilled water, laying the material in a suspended metallic net in order to avoid contacts with the beaker glass. The beaker was then placed in a greater one (of 2000 ml) containing 600 ml of boiling water. After 45 minutes, the material was extracted and cooled at ambient temperature, till the bituminous aggregate could be removed (Figure 3). Visual assessment of stripping percentage proposed by AST standard method was integrated by a Digital Imaging Processing (DIP) approach, implemented to separate residual areas of coated aggregates from stripped ones. In order to exclude further variables connected to ambient lighting and photo exposure, images of different samples were contemporary acquired in the same RAW format picture. DIP software allowed to adjust color palette to obtain representative stripping results, calculating pixel areas of coated and stripped portions (Figure 3).

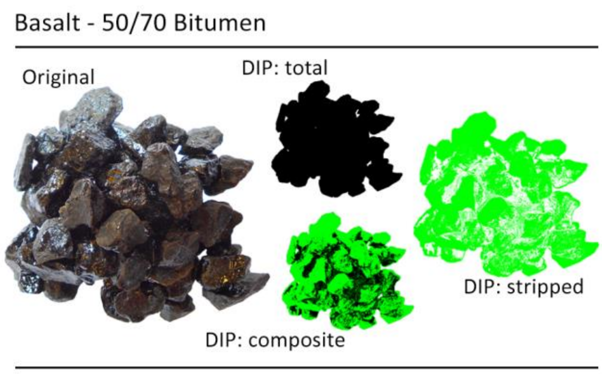

Fig. 3.
Example of blend before and after Ancona Stripping Test

Peel tests were already used in other researches (Zhang et al., 2016); similar operational procedures could be found in ASTM D6862-11 and EN 28510-1:2014 standards. A thick film of hot bitumen was placed on the surface of samples

(30 x 30 x 100 mm^3 size); suddenly, a 20 x 85 mm^2 rectangular aluminum stripe was then laid and bound to the sample. Each specimen was then cooled at room temperature for at least 30 minutes. An extremity of the aluminum stripe was bent in a perpendicular position (with respect to aggregate surface) to be clamped in the upper jaw of the machine used for the test execution (Figure 4). Figure 5 shows a sample at the end of this test.

Testing equipment was a universal machine composed by two jaws (Figure 4). In the lower one, a metallic shoe (able to shift in the horizontal direction) was interposed to clamp the sample and allow a traction force perpendicular (90°) to the aggregate surface (degrees of freedom of shoe adsorbed the non-perpendicular stresses). In the upper jaw, the aluminum stripe was clamped directly. Starting from the measured force F, the temporal evolution of vertical stress was determined for each sample. Punctual stress is calculated continuously as: $\sigma = F/A$, where A is intended as an infinitesimal constant area determined multiplying the stripe width for an infinitesimal length in the x direction, supposed equal to 0,01 mm (A value resulted constantly equal to 0,2 mm^2). Peel test speed was equal to 12 mm/min (as stated in ASTM D6862-11).

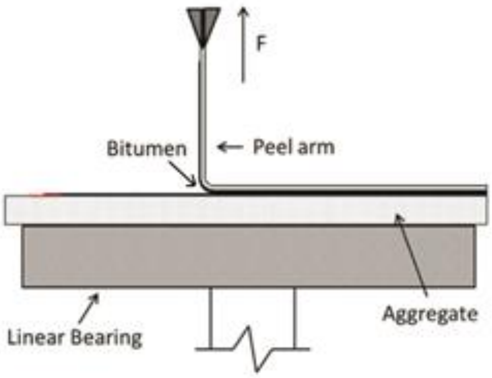

Fig. 4.
Peel test: machine (left), test setting (right)

Fig. 5.
Peel test results: dry smooth (left), wet smooth (right) limestone samples

3. Results and Discussion

Table 2 reports the results of physical-geometrical tests executed on aggregates to evaluate their main characteristics. Basic properties of used bitumen are described in Table 3. In general, all the aggregates showed rather low indexes. They seemed to match the requirements for bituminous mixtures stated by the main technical specifications for wearing courses. The best aggregate typology in terms of shape and flakiness indexes was the porphyry, but it exhibited a greater Los Angeles coefficient with respect to that of limestone or steel slag. Characteristics of bitumen seemed to be in line with typical properties of its class.

Table 2
Physical and geometric characteristics of aggregates

Aggregate type	Flakiness index [%] (EN 933-3)	Shape index [%] (EN 933-4)	Los Angeles index [%] (EN 1097-2)
Limestone	4	10	16
Porphyry	4	4	23
Basalt	7	6	14
Steel slag	5	6	12

Table 3
Basic properties of bitumen

Bitumen	Penetration @ 25°C [0,1/mm] (EN 1426)	Ring & Ball [°C] (EN 1427)	Ductility @ 25°C [cm] (ASTM D 113)	Elastic recovery [%] (EN 13398)	Fraass breaking point [°C] (EN 12593)
50/70	55	50	98	3	-6

Results from tests performed with the high-resolution surface profiler are presented in Table 4. Surface texture can be described through the parameters previously specified. Smoothed-surface samples presented almost similar R_a values: this proved that aggregate type minimally affected the surface roughness processed by the blade (cutting procedure furnished similar surfaces, regardless the aggregate type). Conversely, R_a values changed in the case of rough (untreated) surfaces. In this case, R_a values were always higher than those belonging to smoothed specimens. Roughness sensibly increased using basalt. Studying the sign of R_{sk} parameter, peaks trends of surfaces can be understood (R_{sk} >0 indicates greater peak presence, R_{sk} < 0 the greater presence of dips). Positive R_{sk} values were found for rough porphyry and basalt. As anticipated, also R_{ku} covers a key role in adhesion phenomena: recording R_{ku} values always higher than 3, all samples had jagged surfaces (in particular in the case of steel slag). R_{sm} parameters were quite high in all the situations, demonstrating a great distance from peak to peak and indicating that bitumens can show significant available spaces to develop adhesion (Table 4 reports that highest R_{sm} values were found with rough surfaces and cut-off of 2,5 mm).

Table 5 reports the results of the Ancona Stripping Tests. Residual bitumen quantity (%) and stripping (%) are indicated for each bitumen-aggregate combination. Examining material behavior, basalt and steel slag showed high percentages of stripping. Basalt suffered the stripping effects more than other aggregates, reaching a stripping percentage of 76,4%. Differently, limestone seemed to be a material particular resistant to this phenomenon. These behaviors could be justified by the chemical composition of materials. Indeed, limestone is a basic aggregate, thus hydrophobic; it does not contain silicon (SiO_2) but is full of calcium. For these reasons, it tends to assume a positive charge in presence of water and to generate salts with the functional groups of bitumen. These factors caused the better adhesive properties effectively recorded. On the contrary, porphyry is an acid aggregate, thus a hydrophilic material with a high content of silicon that, with water presence, generated negative charges responsible of mediocre adhesive characteristics. Analog behavior is attributable to steel slag due to a significant amount of calcium (despite silicon content was lower than that of porphyry). Different behavior of basalt (basic material) could be ascribed to a residual presence of impurities on the aggregate surface (before the blending with binder).

A graphical trend of tensile force related to time was built for each sample. Comparisons (Figure 6) were made fixing time to time texture conditions (smooth or rough) and hygrometry (dry and/or wet). It could be noticed that all aggregates in dry condition developed a good interaction with bitumen and good adhesion attitude; vice versa with wet cases (modest bitumen-aggregate adhesion). In general, comparing smooth (Figure 6a) and rough (Figure 6b) surfaces for the same material, the latter situation showed a greater (or similar) adhesive inclination. This finding could be expected considering that, generally, a rougher surface (with a huge number of asperities) should ensure more contact points, supporting the bitumen bond and causing the increase of the traction force needed for the separation. Overall, basalt (both smooth or rough samples) demonstrated force values greater than those of other aggregates. In wet condition, texture type (smooth/rough) seemed to affect the adhesion abilities to a limited extent. This can be ascribed to the fact that, in presence of water, the adhesion between aggregates and bitumen is mainly due to chemical affinity.

Figure 7 describes the tensile forces determined with Peel tests as a function of surface textures (in terms of R_a parameters) of all materials. Figure 7 shows mean R_a values for all the samples and stresses are calculated as mean of stress in the first 20% of test time. Figure 7a clearly depicts the different surface texture between smooth and rough samples; the increase of R_a values (increase of roughness) involved tension increments regardless the aggregate type. Despite a quite low coefficient of determination, tendencies shown in Figure 7a indicated that, as expected, stress increasing corresponded to roughness increments. Thus, a rough aggregate seemed to express a better adhesion with bitumen (with respect to a smoothed one). In Figure 7b the data of Figure 7a (dry samples results) and the wet samples results are presented and classified depending on the hygrometric condition (dry or wet). Dry samples (smoothed and rough) had a greater adhesion force with respect to wet ones and R_a seems to influence the degree of adhesion. On the other hand, R_a parameter did not affect the adhesion force in wet situations, probably because, with water presence, adhesion was mainly governed by chemical affinity, as demonstrated by stripping tests (AST).

Table 4
Results from high-resolution surface profiler

Aggregate type	Surface type	Cut-off [mm]	R_a (μm)	Rsk	Rku	R_{sm} (μm)
Limestone	smooth	0,8	3,80	-0,60	4,03	188,24
		2,5	4,57	-0,68	4,07	262,31

Aggregate type	Surface type	Cut-off [mm]	R_a (μm)	Rsk	Rku	R_{sm} (μm)
	rough	0,8	7,75	-0,49	4,64	317,81
		2,5	17,67	-0,48	3,64	1201,31
Porphyry	smooth	0,8	5,50	-0,34	4,40	230,19
		2,5	7,19	-0,88	4,83	389,80
	rough	0,8	5,63	0,15	4,21	508,12
		2,5	19,60	0,27	3,24	1982,22
Basalt	smooth	0,8	4,98	-0,49	3,65	189,76
		2,5	6,19	-0,68	4,08	269,28
	rough	0,8	9,98	0,04	4,07	446,11
		2,5	25,75	0,01	3,61	1439,15
Steel slag	rough	0,8	9,50	-0,74	8,39	387,30
		2,5	19,58	-0,62	5,68	1078,38

Table 5
Results from Ancona Stripping Test (AST)

Aggregate type	Residual bitumen quantity [%]	Stripping [%]
Basalt	23,6	76,4
Steel slag	32,7	67,4
Porphyry	60,8	39,2
Limestone	93,8	6,2

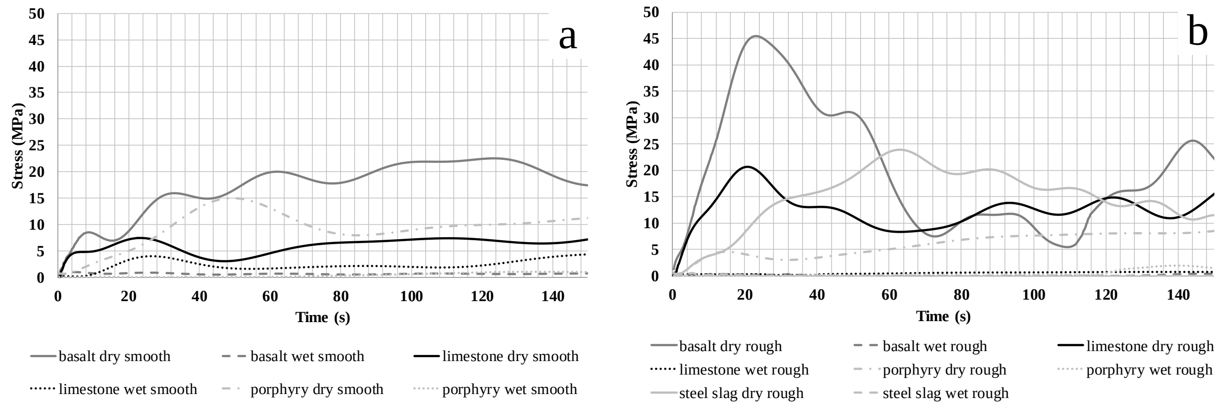

Fig. 6.
Peel test results: a) smooth and b) rough samples

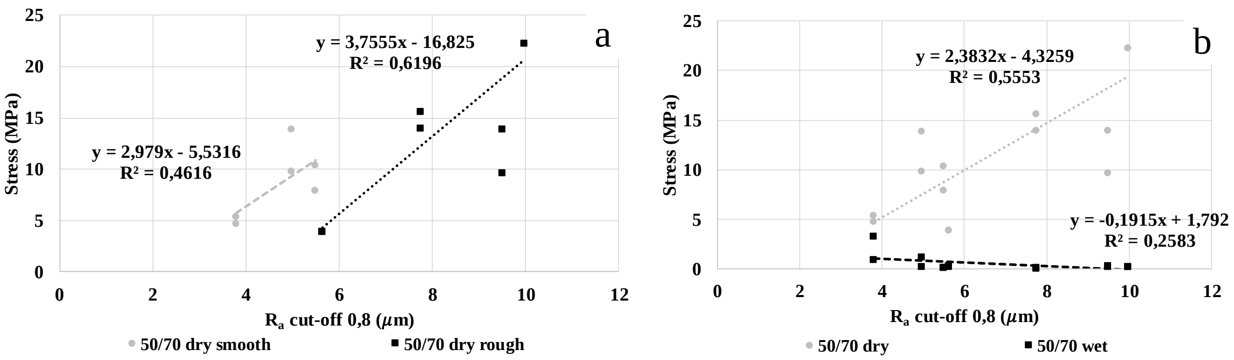

Fig. 7.
Correlations between stresses (MPa) and surface texture (in terms of R_a, in □m).

4. Conclusions

The present paper illustrates and discusses the study of physical-mechanical properties of aggregates and their influence to adhesion forces with binder. Specific emphasis was dedicated to the study of the surface texture through a high-

resolution surface profiler. Stripping (through Ancona Stripping Test) and bitumen-aggregate adhesion (through Peel test) were also evaluated. Several factors proved to influence the adhesion between bitumen and aggregate; the proposed experimental activity tried to focus the attention on the correlation between surface texture, bitumen-aggregate affinity and the influence of humidity on the aggregate surface. Texture of different typologies of aggregate seemed to play a crucial role in the bitumen-aggregate affinity. Authors used an innovative approach, using a complex equipment (high-resolution surface profiler) to evaluate the surface texture of aggregates. In this way, more evident correlations between aggregate nature and bitumen-aggregate affinity were found. A univocal test procedure easily to perform (in terms of equipment complexity and time-consumption) can be suggested based on this approach.
By performing stripping and adhesion (Peel) tests, it was verified that different kinds of aggregates exhibited different behaviors: limestone showed the best affinity with bitumen, while other aggregates demonstrated less affinity. For Peel tests, executed in dry (with natural ambient humidity) conditions, best adhesion was detected for basalt (and porphyry to a lesser extent). Moreover, smooth aggregates always displayed modest adhesion attitude, whereas adhesive forces resulted significant in the case of using rough-surfaced grains. Adhesion values of wet samples were rather small (low bitumen-aggregate affinity with water presence). This seemed to confirm that water was the main actor in the development of adhesion, even attenuating effects of different surface textures (smooth or rough) of analyzed aggregates. In this case, bitumen-aggregate affinity became more important. In conclusion, the proposed experimental method resulted suitable for preliminarily evaluating the bitumen-aggregate affinity. Further detailed analysis to be developed can concern the increasing of variables, studying surface textures and affinities in function of different kinds of binders and surface parameters. Intrinsic variability of all the examined parameters resulted significant, thus a statistic-based validation of obtained results should be performed also to gain indication about the possible extension of physical quantities already accounted.

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