

## Effect of copper slag addition on mechanical behaviour of asphalt mixes containing reclaimed asphalt pavement

A.C. Raposeiras <sup>a\*</sup>, A. Vargas-Cerón <sup>a</sup>, D. Movilla-Quesada <sup>a</sup>, D. Castro-Fresno <sup>b</sup>

<sup>a</sup> Civil Engineering Institute, Austral University of Chile, Valdivia, Chile

<sup>b</sup> Dept. of Transport and Technology of Projects and Processes, University of Cantabria, Santander, Spain

### ABSTRACT

Annually, copper production and refining processes generate large volumes of copper slag, and the disposal of this waste remains a major economic and environmental problem. This annual production causes an increase in the number and volume of landfills, as well as the quantity of slag that backs up landfills, it also produces leachates which contain metals such as Cu, Pb, Hg and SO<sub>2</sub>. In this research, friction and cohesive qualities of copper slag are exploited, in order to incorporate this slag as aggregate in asphalt mixes containing Reclaimed Asphalt Pavement (RAP). Results demonstrate that the use of copper slag in an addition percentage of 35% is favourable, because flow values increase and stability values decrease. The Marshall Quotient is reduced up to 27%, improving the performance of mixes with RAP and obtaining behaviour similar to a traditional mixture. This improvement is also reflected in an 8% increase in the indirect tensile strength, which stands the use of copper slag as a solution in RAP applications with more demanding tensile and fatigue requirements.

**Keywords:** Copper slag; Asphalt concrete; Reclaimed Asphalt Pavement; Marshall Quotient; Indirect Tensile Strength; Resilient modulus

### 1. Introduction

Recovery and reuse of wastes and byproducts from industrial activities and construction sector has become very important in recent decades in reducing the use of raw materials for road construction. The use of crumb rubber [1], recycled concrete [2] or granite countertops manufacture wastes [3, 4] have provided good results in hot mix asphalt, sometimes improving the results of a conventional mixture, and other limiting its use to low-traffic roads. However, the best results are generated with byproducts such as steel and thermal power plant slags [5], being able to replace the natural aggregates without reducing its mechanical properties and durability.

Based on these experiences, copper slag is expected to have similar results. Copper slag (CS) is a byproduct of copper smelting, and from the beginning it has been classified as industrial waste, being placed around the smelting plants. About 2.2 tons of CS are produced for every ton of copper, which has led to large accumulations in Chile exceeding 50 million tons [6]. CS is considered an environmental liability, heavy metals included in the CS can generate leaching problems due to the high toxicity of metals such as Cu, Pb, Hg and SO<sub>2</sub>, especially present in smaller size particles [7]. As an application to tackle the CS problem, this material has been used in the construction field, exploiting the properties of wear resistance, angularity, density and high hydrophilic property. CS in this regard is used as feedstock in the manufacture of tiles, bricks, concrete [8, 9, 10, 11, 12] and as a replacement for Portland cement [9, 11, 12, 13].

When CS is used as an aggregate replacement in an asphalt mixture, the leaching that copper slag could generate is controlled, as each of the slag particles is coated by asphalt binder and sealed of all voids. Some projects with copper slag as a fine aggregate replacement in the manufacture of asphalt pavements have been developed, with successful results for additions ranging from 5% to 30%. However there is some inconsistency in the results of stability, as some research shows that addition of the slag increases stability [8, 9], while in another study, stability was decreased [14]. This inconsistency can be attributed to temperature differences of the tests, as well as the size and percentage of the CS used.

On the other hand, the use of reclaimed asphalt pavement (RAP) in developing new materials is a technique increasingly relevant due to the economic advantages and environmental impact reduction [15, 16].

Some researches have been conducted with high additions of RAP in asphalt mixes, and the results show a change in the physical behavior of the mixes, affecting both durability and structural performance [17, 18, 19, 20, 21]. Additions between 40 and 60% of RAP increased stiffness and indirect tensile strength from 60 to 70%, and the Marshall flow of mixes was reduced between 20 and 50% as the content of the RAP increases. All these changes generate pavements that are more resistant to rutting, but susceptible to the generation of fatigue cracking when no additives or modified binder are used [18, 19, 20, 21].

To solve these inconveniences, softer penetration binder has been used and/or asphalt binder rejuvenating additives have been incorporated to the mixes, so that the initial rheological characteristics of binder can be partially recovered [15, 17, 18, 21]. Such solutions have achieved a stiffness increase of 25%, but tensile strength values increased 5% when incorporating up to 50% of RAP, also achieving Marshall flow values equivalent to those of a traditional mixture [21].

The behavior of mixes including high RAP content can be improved through friction and cohesive properties of the copper slag provided by the angularity of their particles and their calcium content. Also, it allows for a safe use of these slags since the toxicity of heavy metals is neutralized with the asphalt film which covers them, being safe for the environment [8, 22].

## 2. Methodology

This study was conducted by fabricating 140 samples of 101.6 mm diameter and 63 mm nominal height, in seven different types of asphalt mixes with different percentages of copper slag (CS) and RAP (Table 1). Each type of mixture was evaluated by eight equal samples with optimal binder percentage.

### 2.1. Materials

The materials used in this research were: aggregate extracted from quarry (AG), reclaimed asphalt pavement (RAP), copper slag dump (CS) and AC-30 asphalt cement (AC-30) according to the classification of ASTM D3381/D3381M [23]. An IV-A-12 semi-dense mixture gradation curve (Fig. 1) has been used for the development of the samples [24].

AG and RAP were included in all sieve sizes of the gradation curve, while CS was only included in sieves between No.4 to No.100 as appropriate, incorporating the highest percentage of this material as aggregates with a particle size larger than 2.5 mm. This distribution maintains the common size and percentage of RAP in the asphalt mixture, substituting certain percentage of AG for CS.

Volume dosing of the aggregates for the different asphalt mixes was carried out, while the incorporation of AC-30 was performed by weight [25]. This binder dosage considered the percentage of binder included in RAP, in order to not exceed optimal binder dosage for the mixes.

Three groups of samples according to the type of aggregates used can be distinguished in this research (Fig. 2). Physical characterization of the aggregates and binder was performed [26, 27, 28, 29, 30].

Physical characterization of materials such as AG and RAP showed no major differences and remained within the normal range, with density values close to 2600 kg/m<sup>3</sup>, water absorption of 1%, and wear quotient below 15%. However, the copper slag presented higher densities because of its iron oxide content, and a low water absorption due to its crystal structure. As result of this structure, CS also presented a low porosity and superior abrasion factors as other aggregates due to the greater number of edges that particles of this material have (Table 2). All these materials meet the requirements to be used as aggregates of an asphalt mixture [24]. The properties and characteristics of the asphalt cement are listed in Table 3.

To ensure that the different asphalt mixes can be compared with each other, proportions in the gradation were kept, according to their sieve size (Fig. 3). For RAP and CS immovable gradation curves were used, whereas AG dosage for each sieve size was modified for the different combinations. AG dosages are in charged to fulfill the gaps of CS and RAP in particle sizes in order to enter the semi-dense curve design.

Hot mix asphalt samples were manufactured according to the Marshall design [25]. To prevent aging of the binder included in the RAP, the rest of the added material was heated at a different temperature, keeping the RAP in temperatures from 80 to 100 °C for incorporation of low to high percentage of use respectively, based on previous experiences with this material [31].

### 2.2. Testing plan

Once optimum binder percentage obtained, eight samples for each combination were prepared for the mechanical tests. Four of them intended for Marshall stability and flow tests, and the remaining four for Indirect Tensile Strength tests (ITS) [32] and a non-destructive test, in order to determine the resilient modulus by diametric compression.

For sample conditioning, these were placed into plastic bags which were then sealed, in order not to come into contact with moisture when they are submerged in water at 25 °C for 6 hours of the conditioning. To obtain the resilient modulus, the samples were conditioned for a minimum of 24 hours at 15 °C as required by the ASTM D7369 [33].

## 3. Analysis of results

Once physical characterization of the materials is obtained, then the optimum binder content was determined for the seven combinations shown, including the content of binder provided by the RAP (Table 4).

The bulk density curves show that an increase in the incorporation of CS produces an increase in density from 3% to 16%, achieving denser mixes than ones with traditional aggregates (Fig. 4).

The stability test results show that the inclusion of CS generates lower stability values when compared to samples with only RAP addition (Fig. 5). Decrease rates vary from 14 to 23%, producing the greatest effect when the amount of CS exceeds RAP content. However in cases where the amount of RAP is below 30%, CS incorporation achieves values close to conventional mixes values. Furthermore, mixes with CS generated less dispersed stability values, property only attributable to the CS, which helped to generate more stable mixes.

Lower percentages of air void percentages in mixes are obtained for samples where CS was added, and voids decrease even more drastically with binder additions higher than 5% (Fig. 6). However, with the lowest content of RAP and greater involvement of CS, these differences were only 9%, under the values of a traditional mix. CS helps to reduce the quantity of asphalt binder to be used in an asphalt mixture when looking for optimal air void contents.

Once the optimal percentage of binder is obtained, test samples are produced. From Marshall stability and flow tests it was observed that the use of 15% of CS causes a general decrease in stability values of up to 46% (Fig. 7). It was further noted that, as the addition of CS increases, differences between stability values of samples with and without CS tended to be reduced, achieving values of on average 20% lower for samples with CS. This addition allowed the mixes with RAP to reach stability values very close to the average reference value of the traditional mixture analyzed. All the obtained values were higher than the minimal requirements on 9 kN [19].

These decreases are due to the low friction ability that the vitreous texture of the CS has, which prevails before the angularity of the particles when the binder changes its viscous behavior over 30°C, and in this case the adhesiveness between CS and other

aggregates does not increase. There is also an influence on the lime content, which prevails in the fine material from the CS, contributing to their adhesive properties and causing minor stability decreases, which are revealed by incorporating CS in percentages higher than 25%.

In turn, Marshall flow results (Fig. 8) show that the CS increases the value of this parameter for asphalt mixes with RAP when the addition rate is 35%. If 25% of CS is used, similar values are obtained, and a drastically decrease is observed when there are only 15% of EC.

All of the combinations with CS addition fulfilled the requirements for Marshall flow, but an excess of deformation was appreciated for higher contents of RAP when CS was not included, especially for 40% RAP. As the RAP content increased, the particles of the mixture tended to be less bonded, and this unbounding trend was reduced when CS was included. These data demonstrate the adhesive ability of the lime content from fine particles of CS.

Regarding Marshall stability/flow Quotient (Fig. 9), it was determined that the CS noted for its adhesive property and friction capacity with binder at maximum service temperatures, close to 60 °C, which is the sample conditioning test temperature. Also, the use of CS transforms the Marshall Quotient for mixes with RAP to values close to those obtained with the control mixture, with range differences smaller than 10%.

The behavioral model for both groups (with and without CS) for the Marshall stability and flow tests and Marshall Quotient can be observed in Table 5. For the Marshall stability and Marshall Quotient, a high level of influence from the CS was appreciated, while RAP percentage was more significant for the Marshall flow tests than the CS percentage (Table 6). According to the partial correlations with no influence of RAP, CS again had a direct relation with Marshall stability and Quotient, being neutral for the Marshall flow results (Table 7).

A second series of tests was carried out with the remaining four samples for each combination, where Indirect Tensile Strength (ITS) tests and resilience modulus (Fig. 10, Fig. 11) were evaluated. The results show that adding CS in mixes with RAP generates ITS increases between 8 to 9% in strength. Comparing these results with those obtained with traditional mixes, an increase up to 78% in the strength is obtained (Fig. 10). The observed increases stem from the elastic behavior of the binder to temperatures below 30 °C, together with the angularity of the CS particles and the calcium content, which provides adhesion and increases interlocking, the resulting mixture has a higher strength before fracture.

Resilient modulus results showed that by using CS, a greater elasticity is generated only when the addition is of 15%, differentiating of up to 25% more elastic behavior compared to the same mixture with RAP and with no CS addition (Fig. 11). With other additions (25 and 35% of CS) similarities were found between the behavior results. All results remained above 10000 MPa, widely surpassing the 6845 MPa mean obtained in traditional mixes. While the adhesive characteristics of the CS cause increased elasticity at 15% addition, a stiffening effect of the fine material with the asphalt cement is produced for higher incorporation of CS. This effect is similar to the bituminous mastic stiffening effect, which is generated when the relationship between the adhesive material and binder exceed optimum, becoming more rigid at low temperatures, and losing elasticity.

Both Marshall and ITS tests results indicate that asphalt mixes with CS behaves in a similar way as other slags, like steel or thermal power plant slags [5]. The slags produce a reduction in mixes rigidity, and slightly increases their deformation, which in case of RAP added asphalt mixes is a benefit, in an opposite way as other byproducts perform in these mixes, such as crumb rubber, which increases stiffness and reduces deformation and ITS [1].

#### 4. Conclusions

In this section, several conclusions about the effect caused by the addition of copper slag as aggregate replacement in hot asphalt mixes containing RAP are obtained:

Presence of the CS in all addition percentages increased the density of mixes up to 16%, due to the iron content that exists in this material.

The use of CS reduces Marshall stability values for additions exceeding 25%, reaching similar levels to the control mixture values. However, the indirect tensile strength (ITS) increased on average by 8%. This shows the thermal susceptibility of the behavior of this material in asphalt cement.

Asphalt mixes with CS take advantage of the friction and cohesive characteristics of this type of slag at low temperatures; while at higher temperatures, where the asphalt viscous behavior prevails, these fractioning properties disappear, stabilizing the decreases due to the cohesive properties of the lime existing in the fine material of CS.

The addition of 35% CS generates favorable effects on the Marshall flow of the mixes, while for the remaining additions it causes decreases in ductility.

For CS additions higher than 25%, the Marshall quotient is reduced to very close values to those generated by a traditional mixture, obtaining differences of below 10%.

There is a direct relationship between the fine material of the CS, the asphalt cement and the temperature at which it is subjected, since under certain conditions and low temperatures, a stiffening effect occurs, being the cause of the lesser elasticity of the mixes. This effect happens for additions higher than 25%, generating the same behavior as a mixture with RAP and without CS.

The use of copper slags in mixes with RAP improves the overall performance of the mixture for additions of 35% of CS in mixes with 20% of RAP. This addition improves the strength results in mixes with RAP, in percentages that are usually added, without the need for special additives or rejuvenating products, improving performance both flow and tensile strength.

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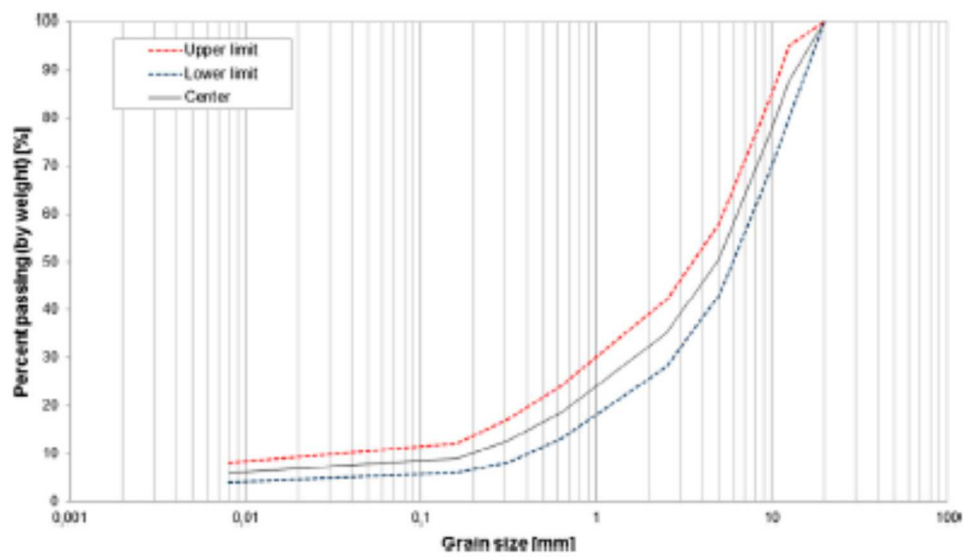


Fig. 1. IV-A-12 gradation curve.

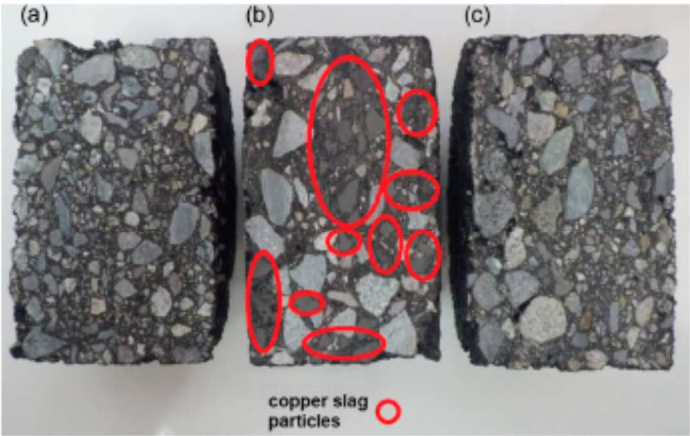


Fig. 2. Sample differences based on materials used: a) Combination VII, 100% AG; b) Combination III, 45% AG-20% RAP-35% CS; c) Combination VI, 80% AG-20% RAP.

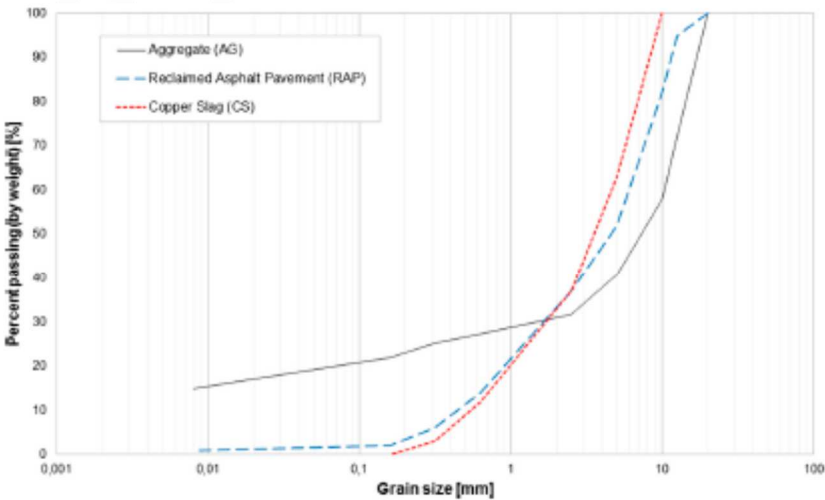


Fig. 3. Gradation curve for each material type.

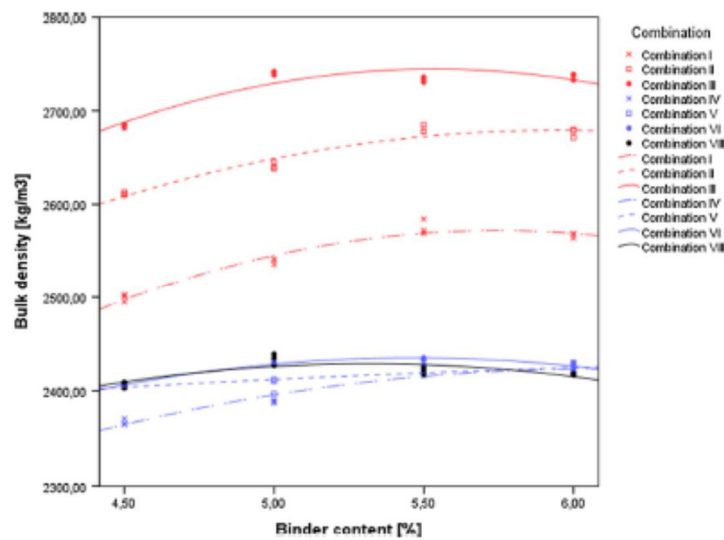


Fig. 4. Bulk density vs. Binder percentage.

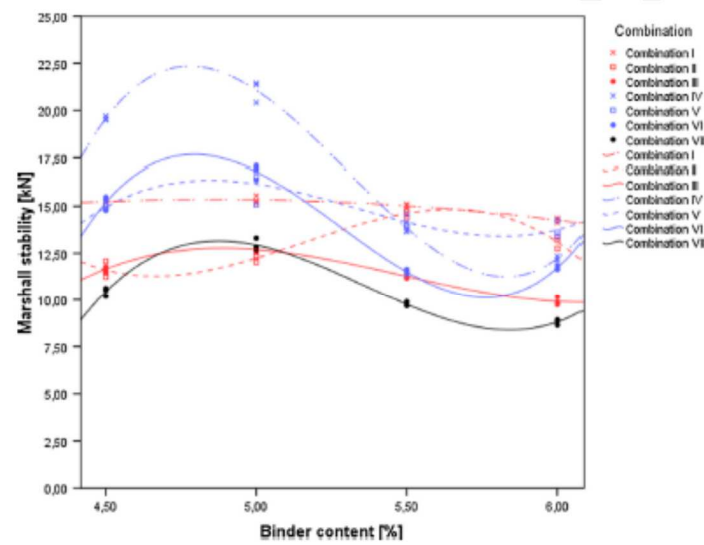


Fig. 5. Marshall stability vs. Binder percentage.

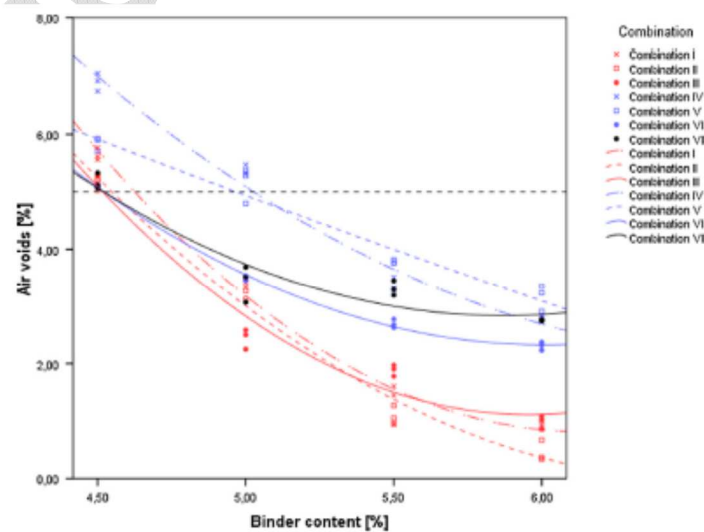


Fig. 6. Air void percentage in mixture vs. Binder percentage.



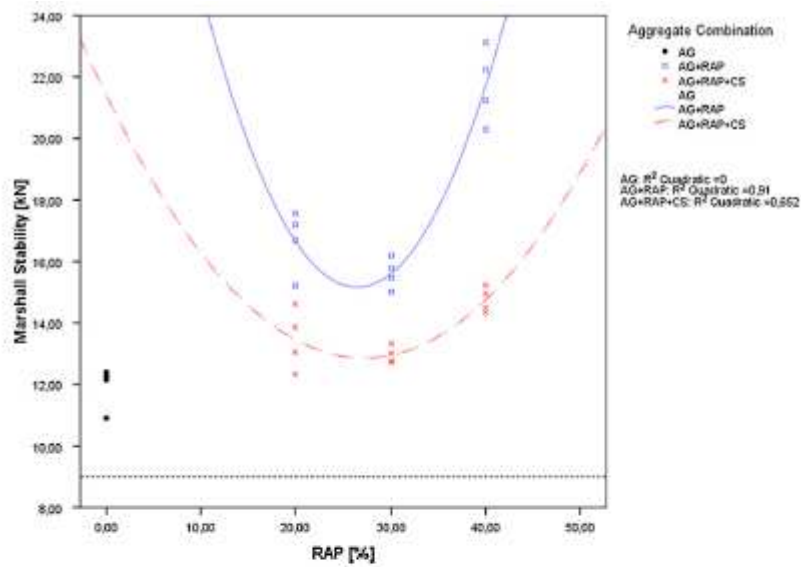


Fig. 7. Marshall stability vs. RAP percentage.

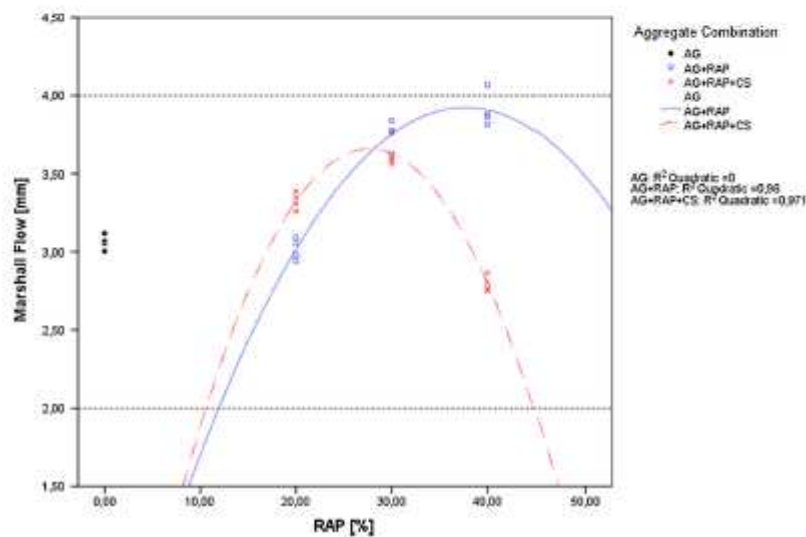


Fig. 8. Marshall flow vs. RAP percentage.

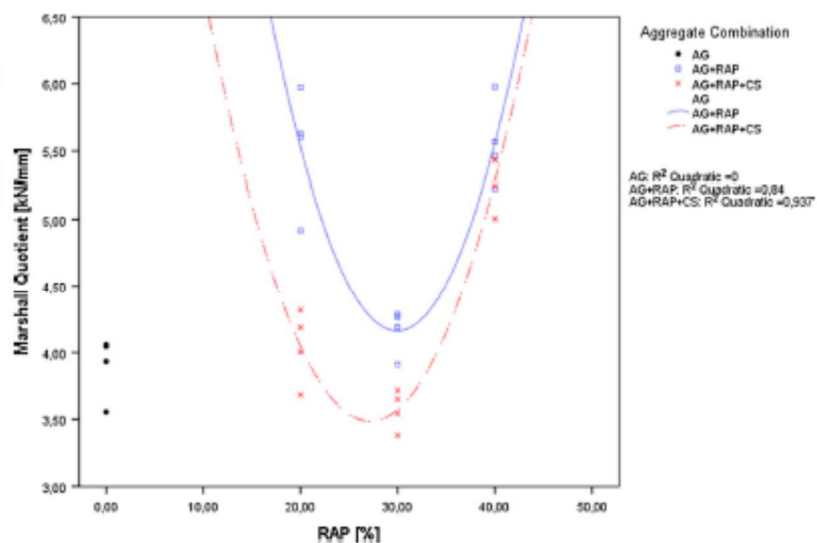


Fig. 9. Marshall Quotient vs. RAP percentage.



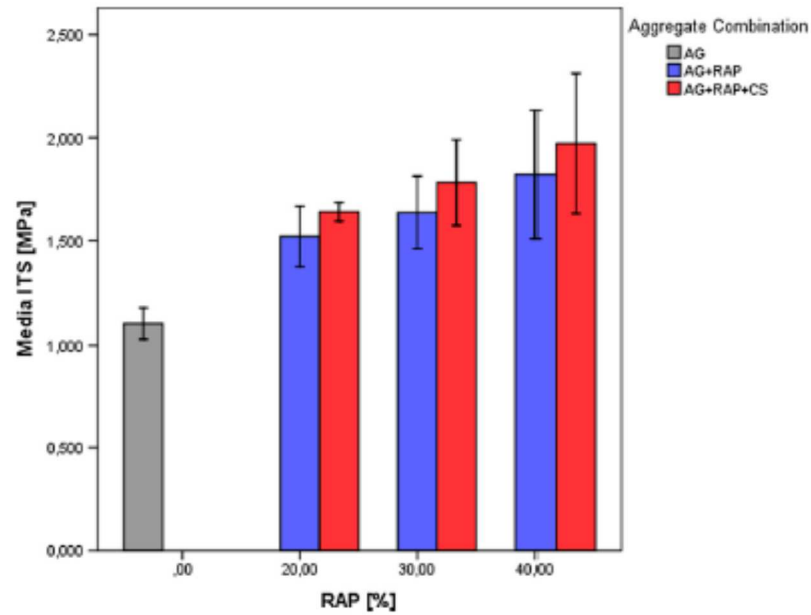


Fig. 10. ITS vs. RAP percentage.

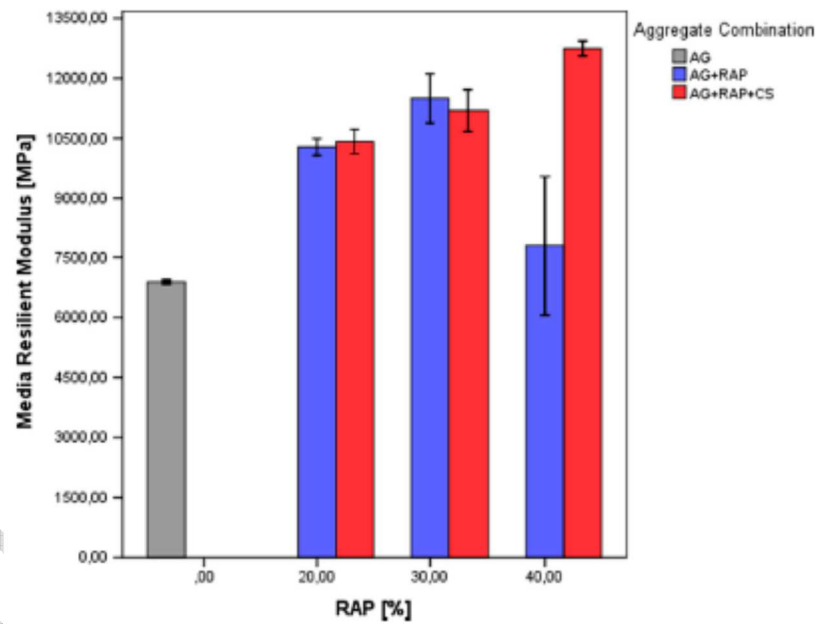


Fig. 11. Resilient modulus vs. RAP percentage.

**Table 1**  
Copper slag, RAP and aggregate combinations.

Combination	Volume dosage [%]		
	AG	RAP	CS
I	45	40	15
II	45	30	25
III	45	20	35
IV	60	40	-
V	70	30	-
VI	80	20	-
VII	100	-	-

**Table 2**  
Physical characteristics of the materials.

Material size	Property	AG	RAP	CS
Fine aggregate	True density [kg/m <sup>3</sup> ]	2675	2693	-
	Bulk density [kg/m <sup>3</sup> ]	2750	2767	-
	Water absorption [%]	1.01	0.99	-
Coarse aggregate	True density [kg/m <sup>3</sup> ]	2719	2692	3689
	Bulk density [kg/m <sup>3</sup> ]	2812	2782	3728
	Water absorption [%]	1.22	1.15	0.28
Resistance to degradation [%]		15.43	15.48	21.02
Fractured particles [%]		91.06	94.00	100.00

**Table 3**  
Binder properties.

Property	Unit	Value
Viscosity @ 60 °C	Poise	3305
Penetration @ 25 °C, 100 g, 5 s	mm	53
Ductility @ 25 °C	cm	>105
Density	kg/m <sup>3</sup>	1029
Softening point	°C	50.2
Mixing temperature	°C	155
Compaction temperature	°C	147

**Table 4**  
Optimal binder percentages, including RAP binder.

Combination	Binder percentage [%]
I	4.8
II	5.0
III	4.7
IV	5.1
V	5.1
VI	4.9
VII	4.9

**Table 5**  
Behavioural models for Marshall tests.

	Aggregate combination	Variable	Non standardized coefficients		Standardized coefficients Beta	t	Sig.
			B	Standard error			
Marshall Stability	AG+RAP	RAP [%]	−1.896	0.357	−5.540	−5.3060	0.000
		RAP [%]^2	0.036	0.006	6.308	6.0410	0.000
		(Constant)	40.262	5.053		7.9690	0.000
	AG+RAP+CS	RAP [%]	−0.628	0.236	−5.459	−2.6610	0.026
		RAP [%]^2	0.012	0.004	6.050	2.9480	0.016
		(Constant)	21.399	3.336		6.4150	0.000
Marshall Flow	AG+RAP	RAP [%]	0.219	0.034	4.500	6.4300	0.000
		RAP [%]^2	−0.003	0.001	−3.599	−5.1430	0.001
		(Constant)	−0.193	0.481		−0.4020	0.697
	AG+RAP+CS	RAP [%]	0.315	0.025	7.357	12.4590	0.000
		RAP [%]^2	−0.006	0.000	−8.012	−13.5690	0.000
		(Constant)	−0.697	0.357		−1.9510	0.083
Marshall Quotient	AG+RAP	RAP [%]	−0.828	0.121	−9.508	−6.837	0.000
		RAP [%]^2	0.014	0.002	9.569	6.881	0.000
		(Constant)	16.560	1.712		9.670	0.000
	AG+RAP+CS	RAP [%]	−0.594	0.079	−6.520	−7.485	0.000
		RAP [%]^2	0.011	0.001	7.229	8.300	0.000
		(Constant)	11.553	1.122		10.300	0.000

**Table 6**  
Parametrical and no-parametrical bivariate correlation.

			RAP [%]	Marshall Stability [kN]	Marshall Flow [mm]	Marshall Quotient [kN/mm]
Copper Slag [%]	Parametrical	Pearson Correl.	0.099	−0.449	−0.071	−0.430
		Bilateral Sig.	<b>0.614</b>	<b>0.017</b>	<b>0.719</b>	<b>0.022</b>
		N	28	28	28	28
	No-parametrical (Rho Spearman)	Correlation Coef.	0.051	−0.450	−0.143	−0.420
		Sig. (bilateral)	<b>0.798</b>	<b>0.016</b>	<b>0.467</b>	<b>0.026</b>
		N	28	28	28	28

**Table 7**  
Partial correlation controlled for RAP percentage.

Control variables		Copper Slag [%]	Marshall Flow [mm]	Marshall Quotient [kN/mm]
RAP [%]	Marshall Stability [kN]		Correlation	−0.630
0.333		0.710		
Bilateral Sig.		<b>0.000</b>	<b>0.090</b>	<b>0.000</b>
df	25	25	25	