

# 402-045 EFFECT OF THE NATURE AND FILLER CONTENT ON COHESION, ADHESIVENESS AND RHEOLOGICAL BEHAVIOR OF THE BITUMINOUS MASTICS

F. PÉREZ JIMÉNEZ<sup>1</sup>, M. BARRAL<sup>2</sup>, J.A. SOTO SÁNCHEZ<sup>3</sup>, J.A. NAVARRO JANSORO<sup>4</sup>

<sup>1</sup>Technical University of Catalonia. Jordi Girona, 1-3, Módulo B-1, 08034 Barcelona, SPAIN

<sup>2</sup>Campezo.Heraclio Fournier, 4 B, 1º. 01006 Vitoria-Gasteiz, SPAIN

<sup>3</sup>Proas-Cepsa. Ctra. DAganzo km 5.500. 28806 Alcalá de Henares, SPAIN

<sup>4</sup>Diputación Foral de Guipuzkoa. Departamento de Infraestructuras. Plaza de Guipúzcoa, s/n. San Sebastián, SPAIN

## ABSTRACT

*Bituminous mastic behavior depends on the filler/bitumen volumetric ratio and the nature of the filler employed. This work gathers the study made by the Diputación Foral de Gipuzkoa for the characterization of different types of fillers and, especially, fillers resulting from the manufacture of bituminous mixtures, bag-house fines.*

*The influence of these fillers in the rheological behavior of different types of bitumen (modified and non-modified) has been analyzed by means of the DSR equipment (Cepsa-Proas).*

*In addition, the effect of mastics on the cohesion and adhesiveness with acid and basic aggregate has also been studied. (Campezo, Road Research Laboratory of the Technical University of Catalonia, Spain)*

**Keywords:** mastic asphalt, mineral filler, cohesion, rheology.

## 1. INTRODUCTION

The combination of mineral dust (filler) with the bituminous binder to obtain a bituminous mastic has a positive effect on the behaviour of the bituminous mixture, as reported by several authors [1, 2, 3]. Bitumen viscosity and stiffness increase whereas, on the other hand, thermal susceptibility decreases. Tensile resistance is also enhanced, resulting in significantly improved mixture cohesion.

However, mixing mineral dust with bitumen causes a reduction in bitumen ductility, and excessive filler may render the mastic fragile and brittle [4].

Additionally, mineral dust may, in certain cases, exhibit a hydrophilic character by which it combines more readily with water than with the bituminous binder. This may lead to an uncoating process of the mixture in the presence of water, resulting in cohesion and resistance loss.

Different manufactured fillers (dust from the grinding of limestone aggregates, calcium carbonate, limestone, cement...) are currently being used, but their advantages and disadvantages remain unknown. Alternatively, the possibility of utilizing recovered filler as a manufactured filler is being investigated. This material is extracted during the aggregate drying and heating processes in the manufacture of bituminous mixes. In most cases, recovered filler is obtained by heating crushed limestone aggregates, the nature and composition of which are similar to those of manufactured fillers (calcium carbonates).

For the above reasons, the present work aims to analyze the effect of filler nature and content both on the rheological behaviour of mastics and the binding power with which they provide mixtures.

## 2. METHODOLOGY

Filler characterization and assessment were accomplished through the analysis of the effect of filler nature and content on the rheological behaviour of mastics and mixture cohesion and adherence. Five fillers of different origin and nature, added to different bitumens at different volumetric concentrations, were studied.

**Rheological behaviour.** The variation of the complex modulus and phase angle with temperature was determined using a dynamic shear rheometer to determine the optimal volumetric concentration of the assessed fillers. Additionally, the effect of concentration variation on one of them (calcium carbonate) was analyzed.

**Cohesion/tenacity.** The UCL method [5, 6, 7], based on the Cántabro test [8], is used to assess the effect of filler nature and content on the binding power (cohesion/tenacity) provided by bitumen to a set of aggregates. Binding power is understood as the bitumen's ability to hold aggregate particles together. This ability depends not only on bond resistance (cohesion) but also on the energy that must be supplied to completely deform and break the bond (tenacity).

The UCL method consists in mixing a selected number of aggregates of the same grading with a set amount of bitumen, specifically 4.5% by mass of aggregate. Aggregate grading is comprised by 80% of particles of size 5-2.5 mm and 20% of particles of size 2.5-0.63 mm. Marshall specimens are prepared with the above mixture and different quantities of mineral dust for subsequent testing in a Los Angeles machine, in accordance with the Cántabro test.

This test is used to determine the weight loss of tested specimens resulting from abrasion and impact against the drum wall. It has been demonstrated that the smaller the loss, the greater the capacity provided by the mastic to hold aggregate particles together. The test here, conducted at 25°C, allowed loss variation with different filler fractions to be known.

**Adhesiveness.** The ability of water to produce the uncoating process of the mastic was also evaluated by the UCL method. Thus, the increment in Cántabro losses experienced by specimens prepared according to the above procedure and after 4 days immersion in water at 45°C was calculated.

### 3. MATERIALS

Materials were characterized before testing to obtain full details, particularly about mineral dusts, which could facilitate the interpretation of mechanical results.

**Mineral dust.** Five fillers of different origin and nature, the characteristics of which are specified in Table 1, were employed. This table also illustrates the  $C_s$  values of the fillers that have been obtained according to the procedure explained below.

Results show that some fillers, such as calcium carbonate 2 and recovered filler, are characterized by a greater grain size. Both fillers have higher apparent density in toluene, and calcium carbonate 2 has thicker grading. Calcium carbonate 2 fails to comply with the Spanish standard because of its high apparent density in toluene.

On the other hand, lime and calcium carbonate 1 have greater fineness, which accounts for their low apparent density in toluene, indeed lower than the minimum allowed by the technical specifications of Spanish standard PG-3 [9].

**Bitumen.** Two different bitumens were used in the test: one conventional bitumen, B-60/70, and the other a polymer-modified bitumen, BM-3c, according to the Spanish standard [10]. Their characteristics are included in Table 2.



Filler Characteristics	Standard	Sieve (mm)	Cement	Calcined Lime	Calcium Carbonate 1	Calcium Carbonate 2	Recovered Filler
<b>Grading (% Passing)</b>	UNE EN 933-2	2	100	100	100	100	100
		0.125	100	98.7	100	88.7	98.2
		0.080	99.8	95.0	99.9	82.8	95.7
		0.063	99.1	92.3	99.6	77.9	93.4
<b>Methylene Blue (g/kg) <sup>(1)</sup></b>	UNE EN 1097-4	0.080	0.67	0.67	0.67	0.67	2.33
<b>Apparent Density in Toluene (g/cm<sup>3</sup>) <sup>(2)</sup></b>	NLT-176/92	0.080	0.71	0.31	0.48	0.87	0.80
<b>Real Density (g/cm<sup>3</sup>)</b>	UNE EN 933-9	0.080	2.76 <sup>(3)</sup>	2.15	2.58	2.59	2.57
<b>Solubility in Water (%)</b>	UNE EN 1744-1	(4)	-	0.5	1	0.3	0.8
<b>Calcium Carbonate (%)</b>	UNE EN 196-2	(4)	13.2	12.6	96.0	90.1	57.8
<b>Ca and Mg Oxides (%)</b>	UNE EN 459-2	(5)	-	98.6	-	-	-
<b>Critical Concentration (C<sub>c</sub>)</b>	IRAM 1542	(4)	0.307	0.154	0.214	0.410	0.440

(1) Spanish Specifications: < 10

(2) Spanish Specifications: 0,5 - 0,8 g/cm<sup>3</sup>

(3) In Toluene

(4) Without Sieving

(5) Calcined Sample without Sieving

**Table 1: Filler characteristics**

Bitumen Characteristics		Unit	Spanish Standard (NLT)	B-60/70	BM-3c
<b>Original Bitumen</b>					
Penetration (25 °C; 100 g; 5 s)		0.1 mm	124	63	61
Penetration Index			181	-0.2	2.8
Softening Point		°C	125	51.7	67.0
Fraass Brittle Point		°C	182	-14	-17
Ductility (5 cm/min) at 5 °C		cm	126	100	-
Solubility in Toluene		%	130	100	-
Storage Stability	Softening Point Difference	°C	328	-	0.5
	Penetration Difference (25 °C)	0.1 mm		-	2
Elastic Recovery at 25 °C		%	329	-	77
Relative Density ( 25 °C / 25 °C)		-	122	1.030	1.032
Asphaltene Content		%	131	18.0	21.3
Paraffin Content		%	345	1.7	-
<b>Rolling Thin Film Oven Residue</b>					
Mass Loss		%	185	0.23	0.42
Penetration (25 °C; 100 g; 5 s)		% p.o.	124	56	58
Softening Point Increase		°C	125	9.0	14.1
Ductility (5 cm/min) at 5 °C		cm	126	62	-

**Table 2: Characteristics of bitumens**

#### Mastic dosage. Tested filler/bitumen volumetric ratios.

Mineral dust was volumetrically added. In this way, the filler volume required in the sedimentation test to completely fill a given bituminous binder volume was determined for each filler and mineral dust type. The same was done for a material of lower viscosity (kerosene) since less viscous materials facilitate the sedimentation process. The volumetric ratio of maximum filler content, above which an excess of filler would occur, thus hindering complete covering of the filler by the bitumen film or liquid used (here kerosene oil), was established. This ratio is called critical concentration ( $C_s$ ) [11, 12, 13] and is given by the following expression:

$$C_s = \frac{V_{real\_filler}}{V_s} = \frac{P_f / P_{ef}}{V_s}$$

where

$P_f$  = poured weight of filler in g

$P_{ef}$  = specific weight of filler in g/cm<sup>3</sup>

$V_s$  = sedimentation volume in cm<sup>3</sup>

From this critical volumetric concentration various volumetric concentration levels were tested with respect to the critical concentration level,  $C_v/C_s$ . Thus,  $C_v$  was determined by the following expression:

$$C_v = \frac{V_{filler}}{V_{filler} + V_{bitumen}} = \frac{P_f / P_{ef}}{P_f / P_{ef} + P_b / P_{eb}}$$

where

$V_f$  and  $V_b$  = filler and bitumen volume in cm<sup>3</sup>

$P_f$  and  $P_b$  = filler and bitumen weight in g

$P_{ef}$  and  $P_{eb}$  = specific weight of filler and bitumen in g/cm<sup>3</sup>

Then,

For  $C_v/C_s = 1$ , we obtain the critical concentration.

For  $C_v/C_s \ll 1$ , a greater filler concentration is needed to fill the entire bitumen film.

For  $C_v/C_s \gg 1$ , there is an excess of filler.

The maximum mixture viscosity and tenacity were obtained for values of the  $C_v/C_s$  ratio close to 1.

## 4. ANALYSIS OF RESULTS

### 4.1. Rheological characterization

Comparison of mastics prepared with both bitumens (B-60/70 and BM-3c) and different fillers with a  $C_v/C_s$  ratio equal to 1 reveals that their behaviours are all very similar. For each of the bitumens, the mastics have comparable complex modulus and phase angle values, Figures 1 and 2.

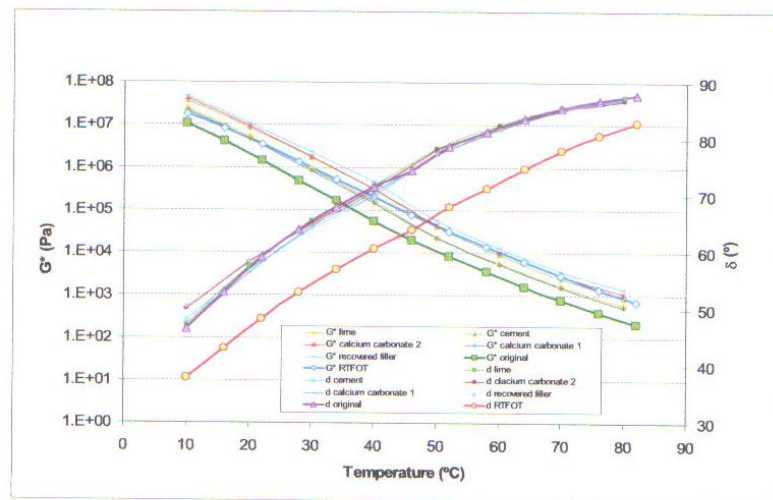


Figure 1: Effect of filler type on the behaviour of bitumen B-60/70 (frequency 1.59 Hz)

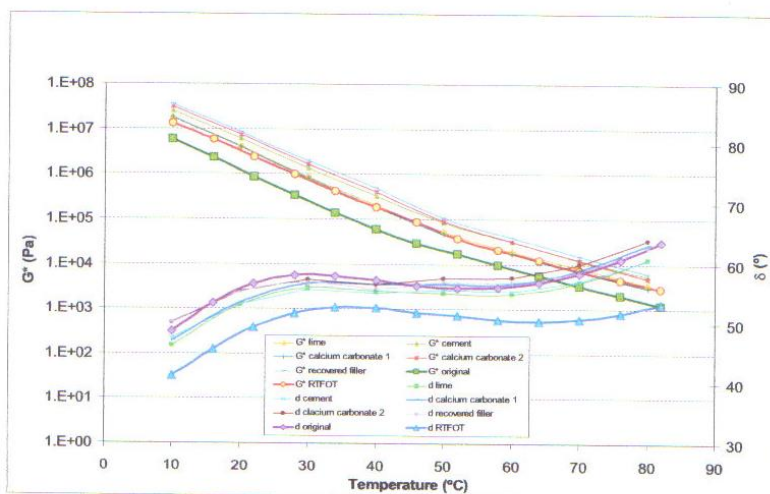


Figure 2: Effect of filler type on the behaviour of bitumen BM-3c (frequency 1.59 Hz)

The influence of the variation in the  $C_v/C_s$  ratio was analyzed for calcium carbonate 1 filler and bitumen B-60/70. The obtained results, Figure 3, indicate that the increase in the  $C_v/C_s$  ratio causes a slight rise in the mastic stiffness, making it impossible to determine the optimal content.

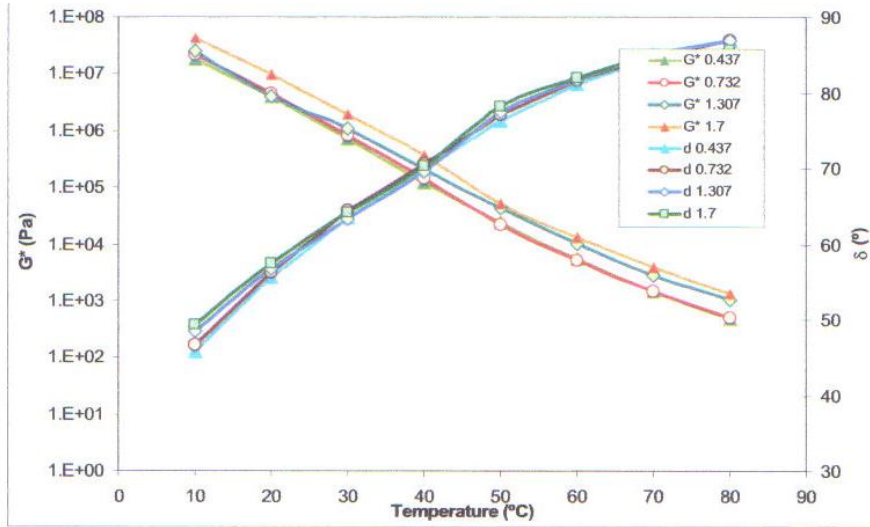


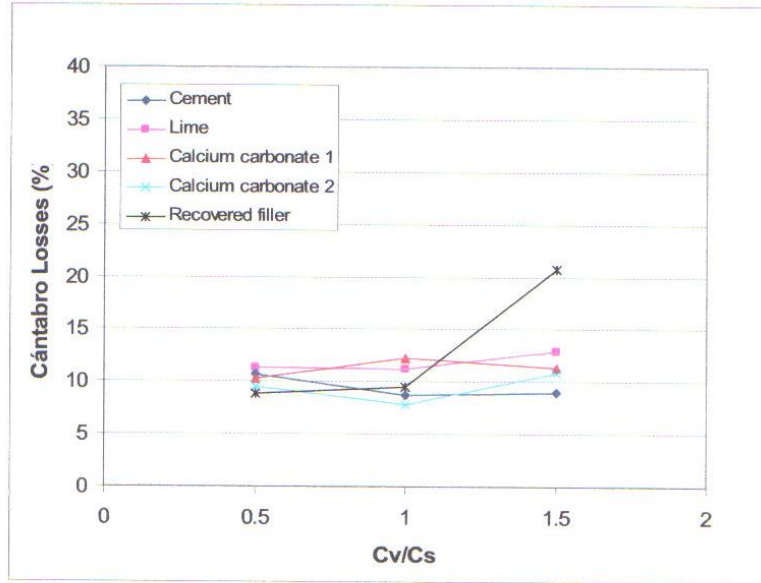
Figure 3: Isochrones (1.59 Hz) of mastic with calcium carbonate 1 and different filler/bitumen ratios

#### 4.2. Results of UCL method

Tests were conducted with two aggregate types, ophitic and limestone aggregates, and only one bitumen type, bitumen B-60/70.

Figure 4 shows the Cántabro losses obtained for bitumen B-60/70. As observed in the DSR tests, filler type does not seem to have a great influence on losses for  $C_v/C_s$  ratios smaller or equal to 1. As this ratio is exceeded, losses increase for fillers with higher  $C_s$  values (recovered filler or calcium carbonate 2), rendering the mastic fragile. However, for fillers with lower  $C_s$  values (lime and calcium carbonate 1), the process occurs for higher  $C_v/C_s$  ratios.





**Figure 4: Effect of volumetric concentration on Cantabro losses (dry conditioning). Limestone aggregate and bitumen B-60/70 (results obtained from Campezo)**

It must also be emphasized that while the mastic behaviour remains almost unchanged when the  $C_v/C_s$  ratio is taken as the parameter -for the same  $C_v/C_s$ , similar Cantabro losses are obtained- differences are observed when it is the filler/bitumen ratio by weight that is regarded as the parameter, Figure 5. In this case, fillers with higher  $C_s$  values, like recovered filler and calcium carbonate 2, allow higher filler/bitumen ratios. For these fillers, the optimal filler/bitumen ratios providing smaller losses correspond to values equal or greater than 1, i.e. between 1 and 2. On the other hand, for fillers with lower  $C_s$  values, such as lime and calcium carbonate 1, the optimal values are much lower than the above mentioned, specifically between 0.5 and 1. Thus, using fillers like lime to manufacture bituminous mixes, and selecting filler/bitumen ratios higher than 1 or 1.2, as recommended by the Spanish standard, may result in mixture fragilization due to excessive filler content. One of the most important conclusions drawn from this study is precisely the benefit of employing volumetric procedures, like the  $C_v/C_s$  ratio, rather than filler dosage by weight in bituminous mixes.

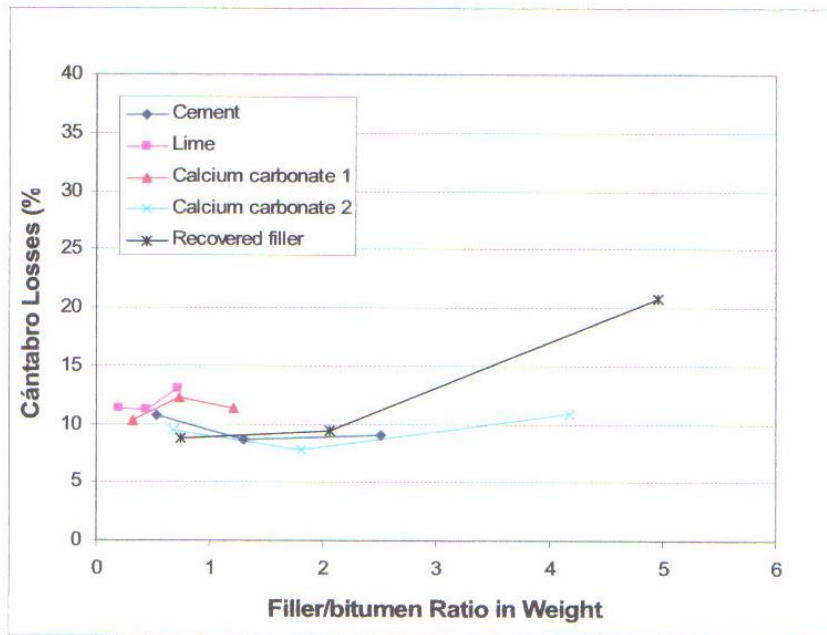


Figure 5: Variation in Cántabro losses (dry) with filler/bitumen ratio by weight (results obtained from Campezo)

The effect of each filler type on the mastic behaviour can be better observed if the influence of the variation in void percentage with the volumetric ratio, is analyzed, Figure 6. Fillers with higher  $C_s$  values (like calcium carbonate 2) are found to provide a thicker mastic film which fills more voids. Additionally, these fillers have a greater filler/bitumen ratio for the same  $C_v/C_s$  ratio. By contrast, the void percentage is hardly reduced by fillers with low  $C_s$  values.

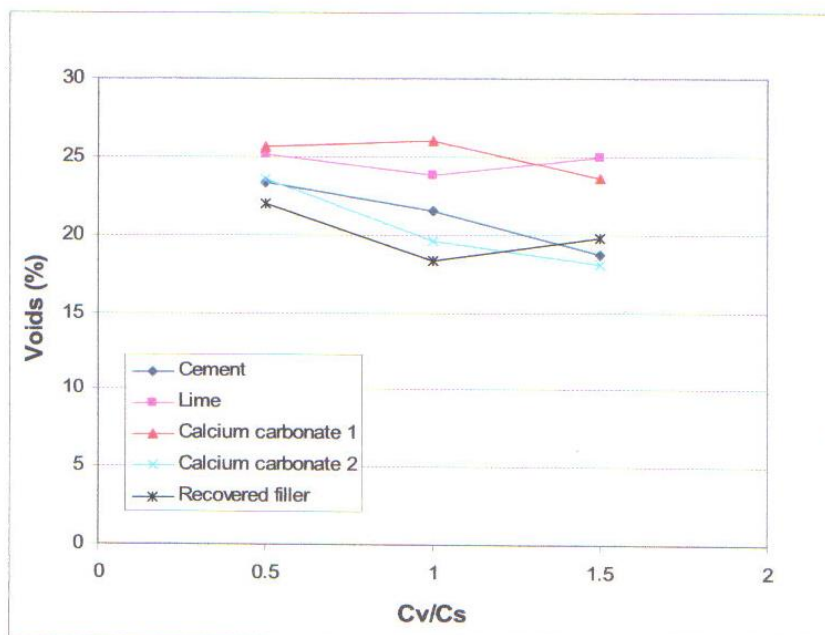


Figure 6: Effect of volumetric concentration and filler type on air voids (results obtained from Campezo)



The effect of water on filler behaviour was assessed by the UCL method by a wet conditioning. Thus, Cántabro losses were determined after 4 days immersion in water at 45°C, using two types of aggregate: limestone and ophitic aggregate, Figures 7 and 8.

Losses after immersion with limestone aggregate are higher than dry losses, especially for low  $C_v/C_s$  ratios, Figure 7.

For the studied range of values, the higher the  $C_v/C_s$  ratio, the lower the losses increase due to water effect, particularly in the case of fillers with the highest  $C_s$  (calcium carbonate 2 and recovered filler). This effect can also be appreciated in Figure 8, with ophitic aggregate, where more influence of the filler type is observed.

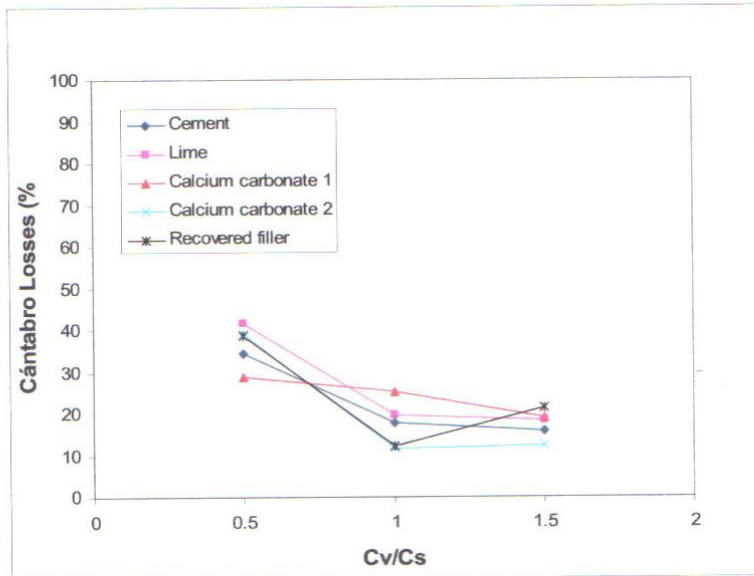


Figure 7: Effect of volumetric concentration and filler type on losses after immersion. Limestone aggregate and bitumen B-60/70 (results obtained from Campezo)

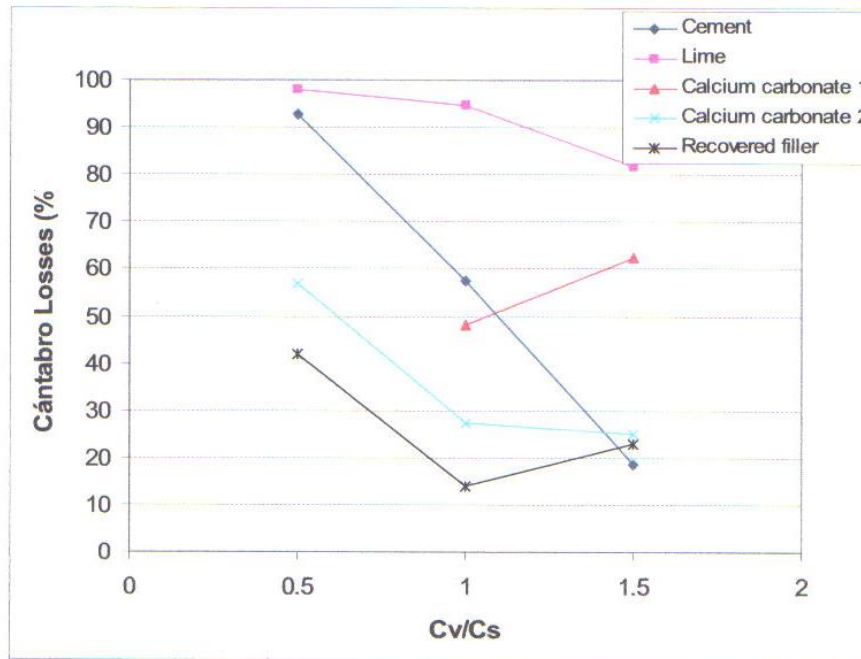


Figure 8: Effect of volumetric concentration and filler type on losses after immersion. Ophitic aggregate and bitumen B-60/70 (results obtained from Campezo)

## 5. CONCLUSIONS

The following conclusions can be drawn from the present study:

The rheological characterization of the analyzed mastics shows hardly any difference between them. An increase in the modulus,  $G^*$ , is observed with respect to the original bitumen (without filler), whereas the phase angle remains almost invariant. Moreover, it is impossible to determine the optimal filler content.

In contrast, characterization by the UCL method not only reveals differences in the response of mastics but also allows the point from which filler content results in excessive mastic fragilization to be clearly established. It is also possible to determine which mastic provides the mixture with greater resistance to the stripping action of water. Furthermore, the method demonstrates that filler dosage must not be accomplished by weight, but by volume.

It is further observed that there is no problem in using recovered filler of limestone aggregates provided its homogeneity and quality are controlled, especially its fineness and activity.

## REFERENCES

- [1] Craus, J.; Ishai, I.; Sides, A. "Some physico-chemical aspects of the effect and role of the filler in bituminous paving mixtures". *Journal of Association of Asphalt Paving Technologists*, Vol. 47, 1978, pp. 558-587.
- [2] Ishai, I.; Craus, J. "Effects of some aggregate and filler characteristics on behaviour and durability of asphalt paving mixtures". *Transportation Research Record: Journal of the Transportation Research Board*, No. 1530, TRB, National Research Council, Washington, D.C., 1996, pp. 75-85.
- [3] Gubler, R.; Liu, Y.; Anderson, D.; Partl, M. "Investigation of the system filler and asphalt binders by rheological means". *Journal of Association of Asphalt Paving Technologists*, Vol. 68, 1999, pp. 284-302.

- [4] M. Recasens, R. Miró, A. Martínez, F. Pérez Jiménez, U. Bianchetto. "Effect of filler on the ageing potential of asphalt mixtures". *Transportation Research Record, Journal of the Transportation Research Board*, No. 1901, TRB, National Research Council, Washington, D.C., 2005, pp. 10-17.
- [5] Pérez Jiménez, F.; Miró Recasens, R. "Characterization procedure of asphalt binders with the Cántabro Test. UCL Method". *Summaries and Papers, 5<sup>th</sup>. Eurobitume Congress*, Estocolmo, Jun. 16-18, Vol. 1A, 1993, pp. 209-213.
- [6] Pérez Jiménez, F.; Miró Recasens, R. "Binder Specifications based on Variation of the Cohesion with Temperature, Moisture and Ageing. UCL Method". *Euroasphalt & Eurobitume Congress*, Barcelona (Spain), 2000.
- [7] Bianchetto, H; Miró Recasens, R.; Pérez Jiménez, F.; Martínez, A. "Effect of calcareous fillers on bituminous mix aging". *Transportation Research Board 86<sup>th</sup> Annual Meeting*, Washington, D.C., 2007.
- [8] NLT-352/86 "Caracterización de las mezclas bituminosas abiertas por medio del ensayo Cántabro de pérdidas por desgaste". Normas NLT, Ensayos de carreteras. CEDEX, Madrid (Spain).
- [9] "Mezclas bituminosas en caliente". Artículo 542 Pliego de Prescripciones Técnicas Generales para Obras de Carreteras y Puentes, PG-3. Orden FOM/891/2004, *Ministerio de Fomento*, 2004.
- [10] "Conglomerantes hidráulicos y ligantes hidrocarbonatos". Pliego de Prescripciones Técnicas Generales para Obras de Carreteras y Puentes, PG-3. *Ministerio de Fomento*, 2000.
- [11] Ridgen, P. "The use of fillers in bituminous road surfacing – A study of filler-binder system in relation to filler characteristics". *Journal of Society of Chemical Industry*, No. 66, 1947, pp. 9-299.
- [12] Ruiz, C. "Concentración crítica de filler, su origen y significado en la dosificación de mezclas abiertas". Dirección de Vialidad de la Provincia de Buenos Aires, Argentina, Publicación N° 11, 1960.
- [13] Dorfman, B.; Rivara, Y. "Sobre la utilización de filler en las mezclas asfálticas en caliente". X Congreso Argentino de Vialidad y Tránsito, Buenos Aires, 1985, pp. 467-484.