RECYCLING OF MODIFIED ASPHALT SHEETS FOR AUTOMOTIVE USE

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1. - INTRODUCTION

Asphalt is one of the by-products of petroleum refining, that has a high viscosity and black color [1,2]. The properties of asphalt depend on the refining process [3], but its structure can be modified to enhance its applicability [4,5]. The main asphalt modifiers are polymers [6] and the most commonly used are atactic polypropylene (APP) and poly (styrene-butadiene-styrene) (SBS). These polymers provide asphalt with elastic behavior, improve its fatigue resistance and increase resistance to deformation [2,4].

A special area of application for modified bitumen is the production of sheets or membranes. An asphalt sheet is a composite material made from modified bitumen, internal or external reinforcement, and external polyethylene layers. Modified asphalt guarantees impermeability and durability. It can also confer some specific characteristics, which depend on the modifiers added. The reinforcement improves the mechanical characteristics of the sheets because it facilitates the distribution of the applied loads and the proper lamination of the modified asphalt [7]. These asphalt sheets are generally used in the waterproofing of roofs, subway walls, and the damping and soundproofing of bodywork in the automotive industry. Their use in the automotive industry dates back to 1950, due to their low cost compared to other materials used as vibration reducers [8,9]. The soundproofing asphalt sheets are cut according to the specific drawings of each vehicle model. The resulting product is irregular in shape, and its cutting process generates a considerable amount of waste. For instance, from 2009 to 2013, the generation of waste associated with the cutting of asphalt soundproofing sheets for automotive use at IMPTEK® Ecuador averaged 112 tons per year [10].

Given the thermoplastic nature of asphalt sheets, most of their waste is currently recycled into hot asphalt mixtures for road paving. However, their recycling for the manufacture of new automotive-type soundproofing sheets (ATR) has not been studied in-depth and has not yet been applied at industrial scale [11]. Therefore, the present study was proposed to recycle ATR sheet cutting waste for its use in the manufacture of new soundproofing sheets in the Ecuadorian company IMPTEK®.

2. - MATERIAL & METHODS

2.1.- MATERIALS

The waste obtained from the cutting of automotive-type soundproofing asphalt sheets (called ATR sheet waste) was supplied by IMPTEK® Ecuador. A Somar by Schulz manual sheet shear was used to uniform the size of the waste. After cutting, pieces of approximately 1 x 1 cm^2 were obtained.

To prepare the asphalt mastic, AC-20 asphalt of Ecuadorian origin from the Esmeraldas State Refinery (REE), elastomeric polymer, and mineral fillers were used. To the original asphalt mastic, ATR sheet residues were added in percentages of 2.5, 5.0, 7.5, 10.0, and 15.0 wt%.

2.2.- METHODS

2.2.1.- Quantity of raw material required in each formulation

A formulation of 40 wt.% asphalt, 2 wt.% elastomeric polymer, and 58 wt.% mineral fillers was used in the preparation of the ATR sheets. This formulation was called "original asphalt mastic formulation". In the formulations with the ATR sheet residues, the amounts by weight of each component of the original formulation are always kept constant, taking into account that the ATR sheet residues have the same composition as the original mastic.

The formulation of each of the mastics analyzed and their components are presented in Table 1. The laboratory-scale formulations were labeled ATR-0 for the original formulation; and ATR-2.5, ATR-5, ATR-7.5, ATR-10, and ATR-15, for the blends with 2.5, 5.0, 7.5, 10.0, and 15.0 wt.% of ATR residues, respectively.

	Weight percentage (wt.%)					
Component	**ATR-0**	**ATR-2.5**	**ATR-5**	**ATR-7.5**	**ATR-10**	**ATR-15**
Asphalt AC-20	40.4	39.4	38.4	37.4	36.4	34.3
Elastomeric polymer	2.0	1.9	1.9	1.8	1.8	1.7
Mineral fillers	57.6	56.2	54.7	53.3	51.8	49.0
ATR waste	-	2.5	5.0	7.5	10.0	15.0
TOTAL	100.0	100.0	100.0	100.0	100.0	100.0

Table 1. Composition of the original ATR sheet mastic and composition of the formulations with cutting residues at contents of 2.5, 5.0, 7.5, 10.0, and 15.0 wt.%

2.2.2.- Laboratory scale testing and selection of the formulation

2.2.2.1.- Asphalt mastic processing

The asphalt mastic was prepared in two steps, i) dispersion of the polymer in asphalt and ii) addition of mineral fillers.

i) For the process of dispersion of the polymer in asphalt, a Charles Ross & Son Company mixer, model HSM-100LH-1, with a saw blade impeller, was used. Asphalt was added along with the ATR film residues at 2.5, 5.0, 7.5, 10.0, and 15.0 wt.%, and heated at 180 °C until homogenized. Once this temperature was reached, the elastomeric polymer was added and mixed for 120 min. Subsequently, the mineral fillers were added at a rate of 50 g/30 min.

ii) After the dispersion process, the resulting mixture was carried to a second mixer for the addition of fillers. In this process, a mixing equipment with a bow tie impeller was used. The mixture was heated to 200 °C and the mineral fillers were added at a rate of 600 g/30 min.

2.2.2.2.- Characterization of the obtained formulations

To determine whether the properties of the formulations of the original asphalt mastic (ATR-0) and the asphalt mastics containing cutting waste are within the established ranges, six control parameters were analyzed, which are shown in Table 2.

Control parameter	**Specification**		**Standard**
	Minimum	**Maximum**	
Viscosity 190° C, SC4-29, 5 rpm	40000 cps	-	NI-10-039
Softening point	110 °C	-	UNE 104281-1-3
Density	1.4 g/cm³	1.6 g/cm³	NI-10-034
Weight per area	6.8 kg/m²	7.4 kg/m²	UNE-EN 1849-1
Thickness	4.8 cm	5.2 cm	-
Thermal Flow	1 mm	10 mm	NI-10-028

Table 2. Established ranges for asphalt mastic properties

Viscosity was determined with a Brookfield® DV2T-RV rotational viscometer and a Thermosel® heating cell, using a SC4-29 spindle at 5 rpm. On the other hand, to measure the softening point, the NTE-INEN 920:2013 standard was used (ring-and-ball apparatus), using glycerin as an anti-adhesion agent, and a heat rate of 5 °C/min.

Density was calculated by the hydrostatic weighing method using 4 x 4 cm² specimens. Samples were taken from different places of the laminated mastic. Regarding the weight per area, 10 x 10 cm² test specimens were cut from three different places of the laminated mastic, and the UNE-EN 1849-1 standard was followed. The reported result is the average of three measurements, expressed in kg/m^2.

In the case of thermal flow, 25 x 5 cm^2 rectangular specimens were cut from different parts of the laminated mastic. The specimens were placed on galvalume steel sheets and their outline was marked. The specimens were placed on convex support and placed in an oven previously heated to 170 °C. After 30 min, the samples were removed from the oven and the displacement of the sheet with respect to the initial marks was measured. All property measurements were made in triplicate, and the average and standard deviation are reported.

2.2.3.- Industrial-scale process tests

For the test at industrial scale, the formulation that presented the best performance at laboratory scale was used. Ninety percent of the asphalt was added to the dispersion tank (vertical mixer with two shafts, the first with a propeller and paddle impeller, and the second sawtooth type) together with the entire elastomeric polymer, at 190 °C for 45 min. In another tank (horizontal mixer with one shaft of 6 paddles) the remaining 10% of asphalt was added together with ATR film residues (5.0 wt.% of the total final mix), at 200 °C. The mineral fillers were added after the complete incorporation of the asphalt residues. As in the laboratory scale, the process was carried out in triplicate and the parameters mentioned above were analyzed.

3. - RESULTS

3.1.- CHARACTERIZATION OF ASPHALT MASTIC FORMULATIONS

The results of the viscosity test are presented in Fig. 1. It was observed that the addition of ATR sheet waste increased the viscosity of the modified bitumen at all temperatures tested. In the case of 15.0 wt.% of waste, the viscosity at 170 °C is not reported because the value is outside the measurement range (maximum 200000 cps), according to the selected method.

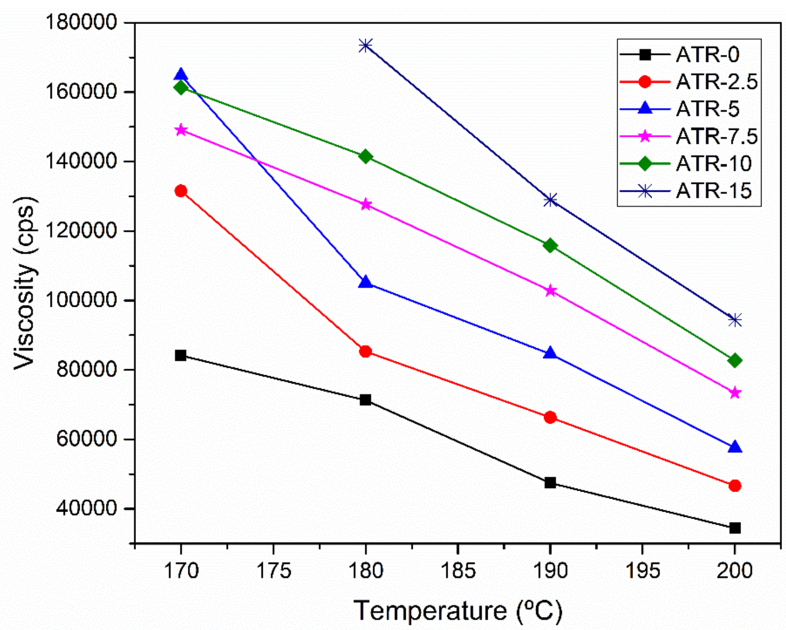

Fig. 1. Dynamic viscosity of the tested asphalt formulations as a function of the percentage of recycled sheet added

Fig. 1 shows that there is a linear relationship between the percentage of ATR film residues and the viscosity of the formulations. It is also observed that the formulations with the lowest percentage of ATR residues (2.5 and 5.0 wt.%) presented the closest behavior to the viscosity curve of the original formulation (ATR-0). This indicates that either of the two formulations will require the least effort from the motors in the mixing process, compared to the formulations containing the higher percentage of ATR residues.

The increase in viscosity due to the addition of ATR residues could be since the ATR sheets produce an absorbing of the maltene content effect. The absorption of maltenes leads to an increase in viscosity and a reduction in asphalt quality [12-14], which directly influences the modified asphalt characteristics.

The results of the softening point test are reported in Table 3. It is observed that the softening point of all formulations with residues increased an average of 43 °C with respect to the average value of the original formulation (ATR-0). The softening point reached a maximum of 164 °C in ATR-10 and a minimum of 160 °C in ATR-5. This increase in the softening point indicates that it is necessary to raise the temperature above this value to get the asphalt to start flowing, so the working temperatures should be around 180 °C to process any of the formulations studied. From an engineering perspective, it is important to consider that if working at temperatures close to 160 °C, the stiffness and thermal flow resistance of asphalt in this temperature range would lead to greater stress on the shafts of the mixing equipment and an increase in the frequency of maintenance. Likewise, the increase in the softening point and the increase in mastic viscosity are factors that must be taken into account when working at industrial scale, so as not to affect the processing equipment.

The increase in viscosity and softening point indicate that the asphalt present in the sheets has suffered thermal degradation due to the previous processing to which it has been subjected, and may have a lower amount of maltenes [6,15]. Additionally, it is important to mention that the ATR sheet waste has suffered aging due to UV radiation and climatic effects because they have been stored outdoors without any protection [16-18]. Therefore, the addition of these residues to the original mastic modifies its properties and quality and increases its viscosity [13,19].

Material	Softening point (°C)	Density (g/cm^3)	Weight per area (kg/m^2)	Thickness (mm)	Thermal Flow (mm)
ATR-0	119.5 ± 1.8	1.52 ± 0.03	7.02 ± 0.04	5.08 ± 0.02	1.1 ± 0.2
ATR-2.5	160.3 ± 2.1	1.54 ± 0.05	7.05 ± 0.04	5.19 ± 0.01	1.6 ± 0.1
ATR-5	160.0 ± 2.4	1.54 ± 0.07	7.03 ± 0.06	5.11 ± 0.03	1.6 ± 0.1
ATR-7.5	162.0 ± 1.3	1.54 ± 0.02	7.05 ± 0.03	5.15 ± 0.02	1.9 ± 0.2
ATR-10	164.0 ± 1.2	1.54 ± 0.06	7.01 ± 0.02	4.99 ± 0.03	1.1 ± 0.1
ATR-15	163.8 ± 1.9	1.54 ± 0.01	7.01 ± 0.03	5.02 ± 0.01	1.7 ± 0.1

Table 3. Softening point, density, weight per area, thickness, and thermal flow of asphalt mastics tested as a function of the percentage of recycled sheet added at laboratory scale

Table 3 also shows the results of the density, weight per area, and thermal flow tests. Regarding density, it can be seen that the addition of ATR film residues increased the density to an average value of 1.54 g/cm³, compared to the value of 1.52 g/cm³ corresponding to the original formulation (ATR-0). This variation is considered not much important since the density values of the formulations studied are within the control ranges (Table 2).

Additionally, Table 3 shows that there is no relationship between the percentage of ATR residues present in the mixes and the corresponding weight per area value of the film. This is because the weight per area is more influenced by the thickness of the film, which remained within the control range (between 4.8 - 5.2 cm). It is worth mentioning that all values are within the control range and that the variation between the maximum and minimum values is less than 5% of the control range.

The thermal flow results (Table 3) of all formulations revealed that all values are within the control range (Table 2) and the values are lower than the upper limit of the established control range (10 mm). It was determined that the thermal flow results are not related to the amount of ATR film residues incorporated in the formulation. The low thermal flow values detected are because the test was carried out at 170 °C. As observed in the viscosity test, at this temperature, all the formulations presented high viscosity, which is reflected in the obtaining of ATR films with higher resistance to flow.

The results derived from the characterization of the original ATR sheets and the studied formulations containing recycled ATR sheets show that all the control parameters are within the established ranges. The ATR-5 formulation was selected for industrial scale testing because it is the formulation that contains the highest content of recycled asphalt sheets while maintaining a viscosity close to the original mastic viscosity. This parameter conforms to industrial requirements (temperature between 170 to 200 °C and viscosity around 85000 cps) and avoids overstressing equipment and engines, which are already operating in this viscosity range.

3.2.- INDUSTRIAL-SCALE TESTING

The asphalt mastics obtained at industrial scale were labeled as L-ATR-0 (original formulation) and L-ATR-5 (5.0 wt.% of recycled sheet). The viscosity results determined for the industrial scale asphalt mastics are presented in Fig. 2. It is observed that the incorporation of 5 wt.% of ATR film residues increased the viscosity of the formulation, which agrees with the trend determined in laboratory tests. However, both formulations (L-ATR-0 and L-ATR-5) presented lower viscosity values than those obtained when laboratory scale tests were performed.

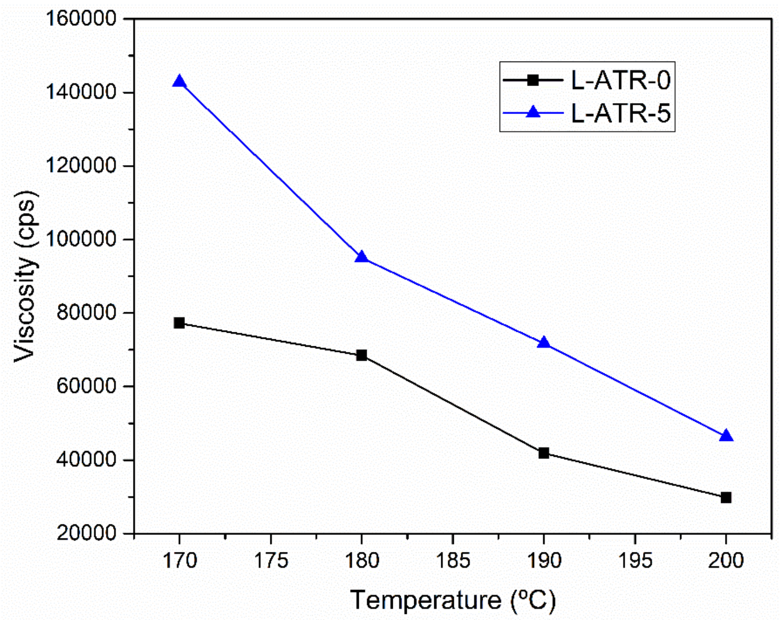

Fig. 2. Dynamic viscosity of asphalt mastics processed at industrial scale, as a function of the percentage of recycled sheet added

The control parameters of the L-ATR-0 and L-ATR-5 formulations processed at industrial scale are summarized in Table 4. All the results were within the control range (set out in Table 2). Like the behavior observed in the laboratory tests, the softening point increased with the addition of asphalt sheet residues, with respect to the original formulation (L-ATR-0). Additionally, Table 4 shows that, at industrial scale, there is no considerable variation in density, weight per area, thickness, or thermal flow with the addition of 5.0 wt.% of asphalt sheet waste.

Material	Softening point (°C)	Density (g/cm³)	Weight per area (kg/m²)	Thickness (mm)	Thermal Flow (mm)
L-ATR-0	121.6 ± 2.3	1.53 ± 0.02	7.10 ± 0.06	5.16 ± 0.04	3.2 ± 0.1
L-ATR-5	152.4 ± 2.7	1.52 ± 0.05	7.08 ± 0.05	5.08 ± 0.03	2.8 ± 0.1

Table 4. Softening point, density, weight per area, thickness and thermal flow of original formulation (L-ATR-0) and the selected formulation (L-ATR-5) processed at industrial-scale

4. - CONCLUSIONS

Asphalt sheets for automotive use (ATR) were produced using residues from the same process. The added residues caused an increase in the softening point and viscosity of the asphalt mastic. These changes were attributed to the fact that the ATR sheet wastes have undergone thermal degradation and UV aging, so the characteristics of the mastic asphalt and its quality are modified.

It was determined that the control parameters of all the formulations studied were within the range established for the commercialization of the products and that the formulations that contain 2.5, 5.0, and 7.5 wt.% of ATR sheet wastes presented very close values in their properties. The formulation with 5.0 wt.% of recycled ATR presented the best relationship between the amount of incorporated residue and the viscosity necessary to not modify the operating conditions of the equipment or cause overexertion in the engines. Therefore, it was selected for testing at industrial scale.

At industrial scale, it was determined that it is feasible to recycle ATR sheet wastes without affecting the properties of the new sheet and maintaining the same processing conditions in the production plant. The results obtained are favorable and show the possibility of reducing the environmental impact by reducing the waste generated.

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