WASTE POLYCARBONATE DERIVED PYROLYTIC OIL AS POTENTIAL BITUMEN REJUVENATOR

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Abstract: Increasing the use of reclaimed asphalt pavement (RAP) is a key objective in sustainable pavement engineering; however, high RAP contents often lead to performance degradation due to the aged and brittle binder. Rejuvenators are therefore required in the production of new asphalt mixes with high RAP content to restore binder properties. Meanwhile, incineration remains a common end-of-life option for plastic waste, which conflicts with zero-waste and carbon neutrality principles. Pyrolysis offers a sustainable alternative to incineration by a thermal conversion of plastic waste into oils that can act as bitumen rejuvenators, enabling the use of higher RAP content in new asphalt mix production.
This study investigates pyrolytic oil derived from waste polycarbonate as a rejuvenator for aged bitumen. The experimental programme includes oil characterization, compatibility evaluation, and assessment of its effects on the mechanical, rheological, and chemical properties of aged binder.
Results demonstrate that polycarbonate-derived pyrolytic oil is a promising rejuvenator for aged bitumen, enabling the useful utilization of waste polycarbonate and the use of higher RAP content in new asphalt mix production, thus supporting circular economy goals and plastic waste reduction.

Keywords: *circular economy, waste polycarbonate, pyrolytic oil, rejuvenator, reclaimed asphalt pavement.*

PIROLITIČKO ULJE IZ OTPADNOG POLIKARBONATA KAO POTENCIJALNI REJUVINATOR BITUMENA

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Rezime: Povećanje upotrebe struganog asfalta (RAP) je ključni cilj u održivom inženjerstvu puteva; međutim, visok sadržaj RAP često dovodi do slabljenja performansi zbog ostarelog i krhkog veziva. Stoga su u proizvodnji novih asfaltnih mešavina sa visokim sadržajem RAP potrebni aditivi tj. rejuvinatori kako bi se obnovila svojstva veziva. Istovremeno spaljivanje ostaje uobičajena opcija za kraj životnog veka plastičnog otpada, što je u suprotnosti sa principima nultog otpada i ugljeničnog neutraliteta. Piroliza nudi održivu alternativu spaljivanju termičkom konverzijom plastičnog otpada u ulja koja mogu delovati kao rejunivator bitumena, omogućavajući upotrebu većeg sadržaja RAP u proizvodnji novih asfaltnih mešavina.
Ova studija istražuje pirolitičko ulje dobijeno iz otpadnog polikarbonata kao alternativni rejunivator za ostareli bitumen. Eksperimentalni deo uključuje karakterizaciju ulja, procenu kompatibilnosti kao i procenu njegovih efekata na mehanička, reološka i hemijska svojstva ostarelog veziva.
Rezultati pokazuju da je pirolitičko ulje dobijeno iz polikarbonata obećavajući rejunivator za ostareli bitumen, što omogućava korisnu preradu i upotrebu otpadnog polikarbonata, kao i mogućnost povećanja procenta RAP u proizvodnji asfaltnih mešavina, podržavajući uz to ciljeve cirkularne ekonomije i smanjenje količina plastičnog otpada

Ključne reči: cirkularna ekonomija*, otpadni polikarbonat, pirolitičko ulje, rejuvenator, strugani asfalt.*

1. INTRODUCTION

Transportation infrastructure, particularly roads and pavements, is among the major contributors to embodied carbon emissions and natural resource consumption in the construction sector. Asphalt pavements, which dominate road construction worldwide, are especially significant in this context due to the high consumption of raw materials and energy [1]. For this reason, the use of reclaimed asphalt pavement (RAP), which involves reusing aggregate and bitumen from waste asphalt pavements, promotes the sustainability of pavement construction and reduces the environmental footprint of road construction [2,3]. According to the production figures published by EAPA over the latest years, the amount of available reclaimed asphalt in Europe is around 50 Mt [4]. However, available data indicate that, on average, the RAP content incorporated in new asphalt mixes amounts to only up to 15% [5], while maximum RAP content rates generally reach up to 35% [2,3]. Increasing the use of reclaimed asphalt (RAP) is a key objective in sustainable pavement engineering; however, high RAP contents often lead to lower performance due to the aged and brittle binder. Rejuvenators

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are therefore required in the production of new asphalt mixes with high RAP content to restore binder properties of aged bitumen [2,6-8]. Commercially available rejuvenators are predominantly derived from the petrochemical industry or from bio-oils obtained from recycling of waste machine or cooking oils [8]. To further reduce the environmental impact of asphalt mixtures containing high RAP contents, alternative rejuvenators with synergistic effects are required. Current research on alternatives to mineral-oil-based rejuvenators has mainly focused on pyrolytic oils from biomass or waste tires [7,9-12], while potential of waste plastics pyrolysis oils remains largely unexplored [13,14].

Polycarbonate (PC) is a thermoplastic polymer, which is widely used in industries including construction, automotive, medical devices, and electronics. In 2023, global PC demand reached 5,08 million tons, with projections estimating 6,82 million tons by 2032 [15,16]. Unfortunately, recycling of PC is hindered due to the lack of economically viable recycling technologies, resulting in a low demand for recycled PC, with landfilling and incineration remaining the predominant end-of-life options for the waste PC [17-20].

On the other hand, utilizing waste plastics in asphalt modification is offering a promising solution to both plastic waste management and the enhancement of road infrastructure [21]. Additional opportunities arise from the application of pyrolysis, which offers a sustainable alternative to incineration by an anoxic thermal conversion of plastic waste into gaseous phase, char, and oils. These oil products can act as bitumen rejuvenators, enabling higher RAP contents use in new asphalt mix production, as an additive for enhancement of asphalt properties, or improving the resilience of bitumen to aging [22-24].

This study focuses on the influence of the waste PC derived pyrolytic-oil (WPCPO) on the mechanical, rheological and chemical properties of laboratory aged bitumen, establishing the basis for useful utilization of waste PC in asphalt production with high RAP content.

2. MATERIALS AND METHODS

The virgin, aged and waste PC pyrolytic-oil modified aged bitumen (rejuvenated) were evaluated in terms of basic mechanical tests and rheological performance, as well through chemical characterization using Fourier Transform Infrared (FTIR) spectroscopy. FTIR spectroscopy was also used for characterization of the WPCPO.

2.1. Materials

The virgin – fresh bitumen used in the research was paving grade bitumen B 70/100 from ÖMV Schwechat.

The aged bitumen, which was the basis for the evaluation of rejuvenation effect of addition of waste PC derived pyrolytic oil, was obtained by means of laboratory aging of the virgin paving grade bitumen B 70/100.

The waste PC derived pyrolytic oil (WPCPO) was obtained from the pyrolysis of the waste PC at the laboratory scale pyrolysis reactor at CEROP d.o.o. (Center za ravnanje z odpadki Puconci), a Slovenian waste management facility.

To assess the effect of the rejuvenation, the WPCPO of ambient temperature was added to the aged bitumen at 150 °C and hand-stirred for 120 seconds, to obtain homogeneous modified aged bitumen. Additions of 5% and 7,5% of WPCPO to the aged bitumen were evaluated.

2.2. Methods

2.2.1. Laboratory aging

The ageing procedure for the virgin paving grade bitumen B 70/100 consisted of the Rolling Thin Film Oven Test (RTFOT) followed by Pressure Ageing Vessel (PAV) ageing. Bitumen subjected to this combined RTFOT and PAV protocol is hereafter referred to as aged bitumen (AB). RTFOT simulates the short-term ageing that occurs during the production, transport, and application of asphalt. Bitumen samples were aged for 75 minutes at a temperature of 163 °C, as specified in SIST EN 12607-1:2025. Long-term ageing, to which the bitumen is exposed during its service life, was characterized by the PAV procedure, in which the bitumen sample was aged for 20 hours under a pressure of 2.1 MPa at 100 °C (SIST EN 14769:2023).

2.2.2. Bitumen properties

Standard testing methods as defined in the European standards were used to determine the mechanical properties of bitumen, aged bitumen and its blends with WPCPO. To assess the consistency of the samples, the needle penetration test was conducted according to SIST EN 1426:2025 at 25 °C. The softening properties were determined using the Ring and Ball method in accordance with EN 1427:2015. The Fraass breaking point test, according to EN 12593:2015, was used to determine the brittleness of the samples at low temperatures.

In addition, the rheological properties of the studied binders were evaluated. Viscoelastic measurements were conducted using a dynamic shear rheometer (Physica MCR 301, Anton Paar) with a parallel plate geometry sensor system (PP25/1 mm). The DSR testing procedure according to SIST EN 17643:2022 was used to determine the complex shear modulus G* and phase angle δ of the bitumen samples. During the test, the temperature was increased from 20 °C to 90 °C at a rate of 1.2 °C/min. The sample was subjected to a constant shear stress of 0.5 kPa in oscillating mode at a frequency of 10 rad/s. Complex shear modulus G* and phase angle δ were measured and plotted against temperature. The output parameter is the temperature at which G* = 15 kPa and the corresponding phase angle.
The Multi Stress Creep and Recovery (MSCR) test was conducted in accordance with SIST EN 16659:2016. Creep and recovery testing is a non-destructive method for characterizing the viscoelastic properties of materials. In the creep phase, the specimen is subjected to a constant shear stress while the resulting shear strain is continuously recorded. The measured strain is time-dependent and reflects the viscoelastic response of the material under sustained loading. When the applied shear stress is removed, the recovery phase begins, during which the decrease in strain is monitored to evaluate the material's ability to recover from deformation. The binder specimen, maintained at a constant temperature of T = 60 °C, was exposed to a sequence of repeated creep–recovery cycles, each consisting of 1 s of constant shear stress (loading phase) immediately followed by 9 s of zero shear stress (recovery phase). The procedure was performed at two stress levels, 0,1 kPa and 3,2 kPa, with 10 consecutive cycles applied at each stress level. During the loading phase, a constant shear stress was imposed on the specimen, while during the recovery phase the stress was reduced to zero, allowing the material to exhibit its elastic recovery response. In this study, the test primarily serves as a comparative tool to evaluate relative changes in binder behavior, since paving-grade bitumen exhibits minimal elastic recovery.

2.2.3. FTIR spectroscopy

Fourier Transform Infrared (FTIR) spectroscopy enables the identification of characteristic absorption peaks due to the vibration and rotation of atoms in the sample by the measurement of infrared radiation absorption, unveiling specific functional groups present in the samples [7,25,26]. It has been widely used as a fast and accurate tool for detecting and analyzing organic materials [27] and is one of the most common techniques used to investigate bitumen aging from a chemical perspective [28]. The more significant changes in the FTIR spectrum associated with bitumen aging are the rise of carbonyl C=O (centered at around 1700 cm^{-1}) and sulfoxide S=O (centered at around 1030 cm^{-1}) bands [29]. In order to quantify the effect of bitumen aging the Carbonyl Index ($I_{C=O}$), Sulfoxide Index ($I_{S=O}$) and Chemical Aging Index (CAI) was applied:

$$I_{C=O} = \frac{A_{1700}}{A_{ref}}$$

$$I_{S=O} = \frac{A_{1030}}{A_{ref}}$$

$$CAI = I_{C=O} + I_{S=O}$$

where A_{1700} is the area of the C=O group peak centered at around 1700 cm^{-1}, A_{1030} is the area of the S=O group peak centered at around 1030 cm^{-1}, and A_{ref} is the area of the reference ethylene and methyl peaks, centered at 1460 and 1375 cm^{-1}, respectively [29].
The FTIR analysis was conducted on the virgin, aged and WPCPO modified aged bitumen to assess the effect of WPCPO, as well as on the WPCPO for the characterization purposes.
In the study FTIR spectrometer PerkinElmer FTIR Spectrum Two with diamond ATR was used. The FTIR ATR spectra for each sample were recorded over a wavenumber range of 4000 to 400 cm^{-1} at room temperature, with a resolution of 4 cm^{-1} and 4 scans per sample.
The areas of the peaks were determined by tangent baseline-correction method [30] using Spectrum IR 10.6.1 software.

3. RESULTS AND DISCUSSION

3.1. Results of mechanical and rheological properties of studied bitumen

The results for penetration, softening point, and Fraass breaking point of the virgin bitumen, aged bitumen (AB), and aged bitumen with WPCPO are presented in Table 1. Comparison of the virgin bitumen and AB shows that, as expected, the bitumen hardened and became more brittle during ageing (penetration decreased, while the softening point and Fraass breaking point increased). The addition of WPCPO to the aged bitumen made the binder softer, as the softening point and Fraass breaking point decreased and penetration increased. For the softening point and Fraass breaking point, 7,5% WPCPO was sufficient to achieve results similar to those of virgin bitumen, but the penetration grade was excessively high.

Table 1. *Results of basic mechanical tests on bitumen*

Sample	Penetration (1/10 mmm)	Softening point (°C)	Fraass braking point (°C)
Virgin bitumen	89	44,8	-13
Aged bitumen (AB)	30	59,2	-8
AB+5% WPCPO	69	48,6	-9
AB+7,5% WPCPO	106	44,4	-12

The softening effect of WPCPO is reflected in the reduction of the complex modulus (G*) of aged bitumen over the entire temperature range (Figure 2 (left)). The effect was more pronounced at higher WPCPO contents, with 7,5% WPCPO already reducing G* below that of the virgin bitumen. This is further evidenced by the decrease in the temperature corresponding to G* = 15 kPa (Table 2). A dosage of 5% effectively restored the binder properties close to the virgin state, while 7,5% caused over-rejuvenation.
In addition to softening, the phase angle of aged bitumen is partially restored at temperatures up to 70 °C and fully restored at higher temperatures (Figure 2 (right)). This suggests that WPCPO does not act solely as a simple softening agent, but may also help recover other viscoelastic properties and partially restore the internal microstructure of the aged binder.
Although the MSCR test response of rejuvenated aged bitumen moves partially toward that of the virgin binder (Figure 2), it does not fully recover, highlighting that WPCPO cannot completely reverse ageing effects in terms of MSCR test. MSCR test results show that rutting resistance decreases with the addition of WPCPO, as expected due to the softening of the binder. The non-recoverable creep compliance (Jnr) increased with WPCPO content (Table 2), indicating that rutting resistance decreases at higher dosages, while the increase in $J_{nr\text{-}diff}$ and R_{diff} shows greater stress sensitivity. This also demonstrates that the rejuvenated binder remains more sensitive to stress variations than the virgin material.

Table 2. *Results of rheological tests on bitumen*

Sample	T at G*=15 kPa (°C)	δ at TG*=15 kPa (°)	$R_{0,1\ kPa}$ (%)	$R_{3,2\ kPa}$ (%)	R_{diff} (%)	$J_{nr\ 0,1\ kPa}$ (kPa^{-1})	$J_{nr\ 3,2\ kPa}$ (kPa^{-1})	$J_{nr\text{-}diff}$ (%)
Virgin bitumen	46,1	81,8	-1,1	-2,3	-111,8	6,6	7,3	10,2
Aged bitumen (AB)	59,7	79,8	14,3	5,6	60,6	0,7	0,6	16,4
AB+5% WPCPO	49,5	77,1	5,0	-1,0	119,0	3,3	4,3	28,8
AB+7,5% WPCPO	45,2	77,2	2,5	-2,9	213,3	6,9	9,4	36,3

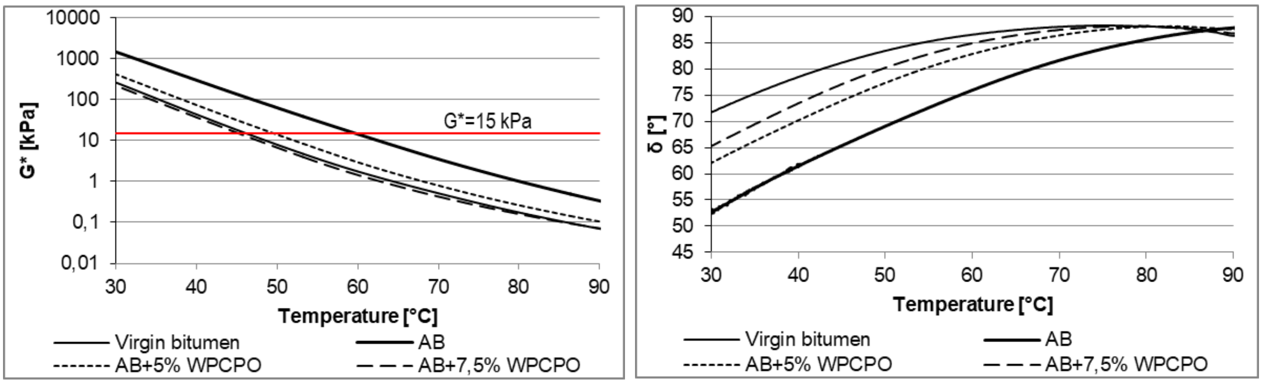

Figure 1. *Complex modulus G* (left) and phase angle δ (right) measured at DSR test*

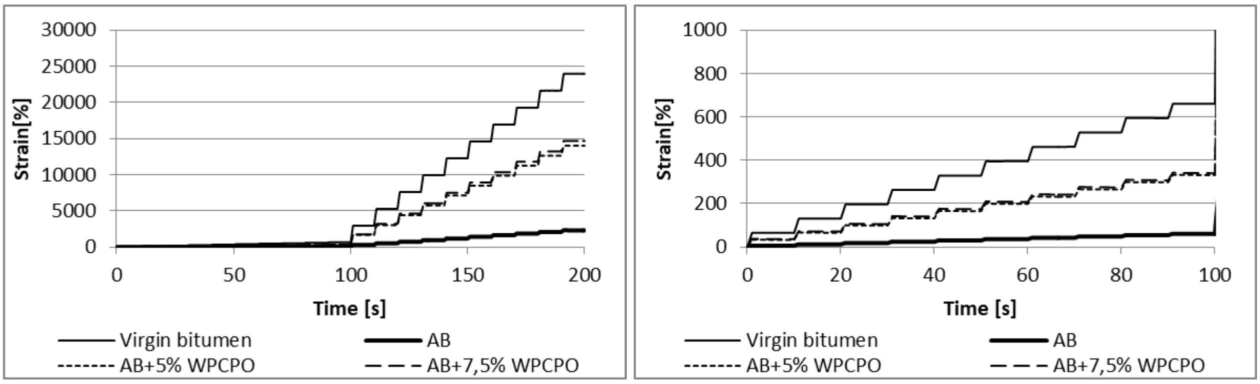

Figure 2. *Multi Stress Creep Recovery (MSCR) test results: entire test duration (left) and enlarged view of the first stress level at 0,1 kPa (right)*

3.2. Chemical composition of the investigated modifier and binders

3.2.1. FTIR Spectroscopy analysis of WPCPO

The properties of pyrolytic oils are largely depended on the degradation process, operational temperature and presence of impurities or catalysts [31-34]. In general, several phenolic products are generated during PC thermal degradation, as well as smaller molecules such as aldehydes, ketones and branched structures [31,35]. The chemical composition of waste PC-derived pyrolytic oil used in this study was characterized by FTIR spectroscopy and is presented in Figure 3.

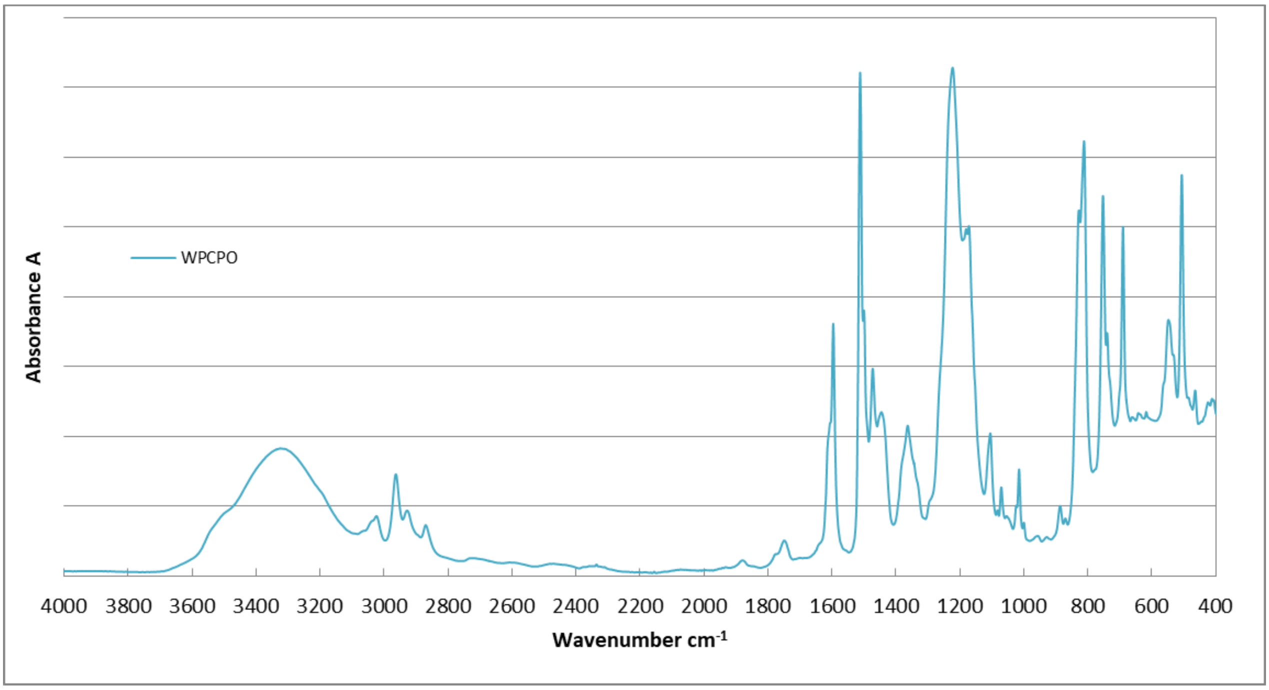

Figure 3. *FTIR spectra of WPCPO*

The FTIR spectrum of pyrolytic oil derived from waste polycarbonate (PC) indicates an aromatic-rich composition dominated by phenolic compounds. The presence of aromatic structures is confirmed by peaks at ~3050–3000 cm^{-1} (aromatic C–H stretching), ~1600 and ~1500 cm^{-1} (aromatic C=C stretching), and ~900–700 cm^{-1} (aromatic C–H out-of-plane bending). A broad band at ~3500–3200 cm^{-1} (O–H stretching) together with strong absorptions at ~1300–1000 cm^{-1} (C–O stretching) indicates phenolic derivatives, consistent with depolymerisation of the bisphenol-A backbone. Additional peaks at ~1770–1700 cm^{-1} suggest the presence of carbonyl groups from residual carbonate or degradation products. Minor aliphatic components are evidenced by bands at ~2950–2850 cm^{-1} (aliphatic C–H stretching) and ~1450–1375 cm^{-1} (CH_2/CH_3 bending). Overall, the spectrum indicates that the pyrolytic oil consists predominantly of aromatic and phenolic compounds with smaller contributions from oxygenated and aliphatic species [35-37].

3.2.2. FTIR Spectroscopy analyses of investigated binders

FTIR spectroscopy analyses were performed on virgin, aged (AB), and modified binders with 5% and 7,5% of WPCPO (AB+5% WPCPO and AB+7,5% WPCPO respectively). From the stacked FTIR spectra shown in Figure 4, it can be observed that the binders exhibit similar features, with the main peaks appearing at the same positions. The main differences between the virgin and aged bitumen are the appearance of the carbonyl group (C=O) at 1700 cm^{-1} and the increased intensity of the sulfoxide group (S=O) at 1030 cm^{-1} in the aged binder. For these peaks, a decrease in intensity after the addition of WPCPO to the aged bitumen can be observed, which is more pronounced for the binder with a higher addition of WPCPO, indicating the rejuvenation of the aged binder. In the spectrum of the binder with 7,5% WPCPO, some additional peaks can also be observed, mainly at 3500–3200 cm^{-1} (O–H stretching), 1511 cm^{-1} (aromatic C=C stretching), and approximately 1300–1100 cm^{-1} (C–O stretching), indicating the presence of phenolic derivatives presumably originating from the WPCPO or, even this is less probable, potential limited intermolecular interactions between WPCPO components and oxidized groups in aged bitumen.
Furthermore, the Carbonyl ($I_{C=O}$), Sulfoxide ($I_{S=O}$), and Chemical aging (CAI) indices were calculated for the investigated binders, as shown in Table 3. The analysis revealed that the carbonyl and sulfoxide indices of the rejuvenated binders were lower than those of the aged binder, suggesting the rejuvenation ability of the WPCPO.

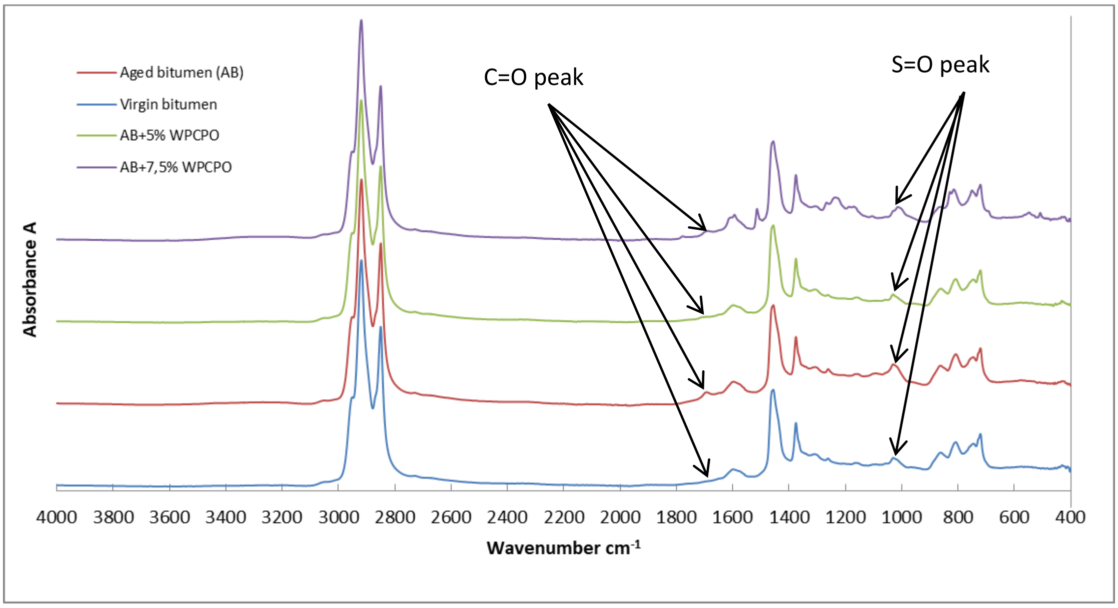

Figure 4. *FTIR spectra of investigated binders*

Table 3. *Carbonyl, Sulfoxide and Chemical ageing indices for investigated binders*

Sample	$I_{C=O}$	$I_{S=O}$	CAI
Virgin bitumen	0	0,0075	0,0075
Aged bitumen (AB)	0,030	0,015	0,0450
AB+5% WPCPO	0,0075	0,0100	0,0175
AB+7,5% WPCPO	0,0076	0,0025	0,0101

4. CONCLUSION

The mechanical and rheological bitumen test results show that WPCPO can effectively soften aged bitumen and improve its properties. This is largely supported by FTIR spectroscopy analyses. These analyses revealed that the Carbonyl, Sulfoxide and Chemical ageing indices of the rejuvenated binders were much lower than those of the laboratory-aged bitumen, confirming the chemical restoration or rejuvenation of the aged binder. The reduction of the Carbonyl index ($I_{C=O}$) in the case of 5% and 7,5% addition of WPCPO is very similar, amounting to 75,1% and 74,6% respectively to the aged bitumen indices. For the Sulfoxide index ($I_{S=O}$), a larger reduction of 83,1% is observed for the rejuvenated AB with 7,5% WPCPO addition, compared to 33,5% for AB+5%WPCPO. Since the Chemical ageing index provides information on the overall degree of rejuvenation, the higher reduction for AB+7,5%WPCPO is to be expected, amounting to 77,6% compared to 61,1% for AB+5%WPCPO.

When comparing the mechanical properties of rejuvenated and aged binders, a 5% dosage of WPCPO improves the characteristics of the aged bitumen almost to those of virgin bitumen, while a dosage of 7,5% tends to over-soften it, reducing stiffness and increasing sensitivity to stress. From a rheological perspective, the phase angle suggests that WPCPO does more than simply soften the binder; it also partially restores other viscoelastic properties, as supported by the FTIR findings.
The results of the study suggest that WPCPO is an effective rejuvenator, which can have synergistic effect in circular economy, combining use of waste plastic polycarbonate pyrolytic oil as rejuvenators to enhance amount of RAP used in production of new asphalt, promoting circularity in asphalt and plastic waste management. Nevertheless, careful optimisation on WPCPO dosage must be performed to avoid over-softening and to obtain up to the intended use tailored rejuvenated bitumen.
Since the encouraging results are based solely on laboratory-aged bitumen, a real case of rejuvenation on reclaimed asphalt bitumen and asphalt mixes using high proportions of RAP should also be studied. In addition, the influence of WPCPO on the ageing properties of the rejuvenated binder should be a focus of further research.

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