

## How Organic Waste Improves Bitumen's Characteristics

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### Article info

*Received:*

19 March 2021

*Received in revised form:*

27 April 2021

*Accepted:*

6 June 2021

### Abstract

The organic fraction derived from the differentiated collection of urban waste is mainly composed of fatty acids, medium molecular weight hydrocarbons and cellulose. This peculiar composition gave us insight into the possible use of organic waste to improve bitumen's characteristics (possible antioxidant, regenerating and/or viscosifying additive for road pavements). The issue of the disposal of organic waste is a global one and it's constantly of increasing concern. This study looks to alleviate this problem by finding ways for this waste fraction to be utilized for the greater good- in this case, as an additive for bitumen binder in road pavements. The present study is focused on the use of waste as it is and waste treated by the FENTON process (treatment with ferrous sulphate and hydrogen peroxide solution). Dynamic Shear Rheology (DSR) and aging tests (Rolling Thin Film Oven Test, RTFOT) showed that two of the additives tested in this study proved effective: one can be utilised as a viscosifying agent and the other can be used as a filler.

## 1. Introduction

Waste management is a problem of growing concern globally and industrialization amongst other avenues of waste production has contributed significantly to the production of organic waste in recent years. This has led to waste management problems as conventional waste disposal methods are not eco-friendly and are mostly inefficient [1]. About a million tons of waste are produced globally every day and incineration which is arguably the most common conventional waste disposal method is known to produce over 200 different types of toxic compounds such as nitrous oxide, sulphuric acids, fluorides, hydrogen chloride [2, 3]. Research in recent years is drifting towards looking for sustainable ways such as reducing landfill usage to treat and manage the environmental impact brought about by waste products that are produced from human and industrial activities in the 21<sup>st</sup> cen-

tury. This has facilitated the drift of research towards the development of green eco-friendly technologies to recycle waste thus creating a circular economy [4–10].

In the road pavement sector, bitumen is used as the binder for preparing asphalt conglomerate. More often than not, conventional bitumen does not have all the performance requirements for road pavement construction. These roads, depending on the geographical location, are increasingly subjected to conditions such as heavy traffic, harsh climatic conditions, oxidation and heavy loads. If the bituminous mixture obtained does not meet these performance requirements, the bitumen binder needs to be modified with additives to improve its performance thereby improving the performance of the asphalt conglomerate as a whole [11–13]. Most additives on the market are not eco-friendly and research in this field keeps looking for more eco-friendly and sustainable ways to improve asphalt performance. Bitumen is a viscoelastic complex mixture of organic compounds derived from the heavy petroleum fractionation process in the refinery. It is a

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substance used to bind inorganic conglomerates in road pavement preparation. This binder is mixed with the aggregates at high temperatures, and the presence of oxygen causes the bitumen to undergo strong oxidation which is a detrimental process reducing its mechanical characteristics.

In order to limit oxidation, specific additives are added to the bitumen before the mixing process. On the market, there are no eco-friendly additives [14, 15] and so recent attention is being paid to ecological additives with high performance in order to reduce the oxidation process [16–18].

Since further oxidation can take place also after paving in a slower process called ageing which also involves loss of more volatile components and structural aggregations [19], the same problem holds also for longer times. For this, additives, usually based on strong acids or bases, are present on the market [20–22], but eco-friendly additives are recently being studied [23–28].

The idea to use eco-friendly substances can be extended to eco-friendly processes such as the recycling of aged road pavements (the so-called Recycled Asphalt Pavement – RAP) to be partially reused in new road pavements while maintaining the performance and duration of the latter over time [29–31]. Also in this case, specific (regenerating) additives, generally based on amines, surfactants, oils, etc. are used [32–34].

To make the whole cycle entirely eco-friendly, research in recent years has focused on the use of recycled material from wastes to create new environmentally friendly additives for improving road pavement characteristics, with obvious advantages in terms of environment protection and safeguarding of human health [35–37].

Furthermore, the problem of the disposal of organic waste exists all over the world and so the purpose of this work is to identify possible reuse of this type of waste as an additive for road pavements in accordance with a circular economy facilitated by circular chemistry practices which are constantly gaining widespread acceptance in the industrial research sector [38–41]. One major limiting factor of organic waste is that there is never a homogeneous composition. This composition changes according to the nation, the geographical area, the number of inhabitants, ethnic culture, etc. To further improve the quality of the waste, it was chemically pre-treated (FENTON process) in the laboratory. On the final samples, rheological measurements were performed and to evaluate the performance of the additives as an antioxidant, the

ageing process in the plant was simulated in the laboratory using Rolling Thin Film Oven Test.

## 2. Experimental part

Waste was obtained in the ambit of the project “RESIFAC” (realization and experimentation of pilot plants for fast composting of civil and industrial organic waste). In the present study our attention is focused on the organic fraction of municipal solid waste, which includes food residues or food preparations and assimilable fractions, and which constitutes more than 30% of the total weight of municipal solid waste. Such residues were collected through a sampling procedure lasting 12 days. On each of these days, waste was taken from a group of four different families, each day a different one. The waste collected on each sampling day (about 5 kg) was mixed and milled to a millimeter size by means of a steel blender. The resulting minced material was stored at  $-20\text{ }^{\circ}\text{C}$  for 12 days. After this time, all the daily rates were defrosted and mixed; the material obtained was divided into bags of 200 g, re-frozen and then used after heat treatment at  $110\text{ }^{\circ}\text{C}$ . This was the starting matrix for the oxidative treatment. The oxidation reaction [42] was carried out using a laboratory-scale glass reactor apparatus. The compounds which are mainly oxidized by Fenton reagent are aromatic and aliphatic hydrocarbons. The details are reported in the work by Salvino et al. [43] which gives details on the whole oxidation process. The sample of organic material, 200 g, was placed into the glass reactor and the Fenton reagents were added: 10 mL of a  $\text{FeSO}_4$  solution followed by the addition of 10 mL of hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) solution. The mixture was kept under stirring conditions, the reactor was placed in a water bath, and the internal temperature was kept at  $60\text{ }^{\circ}\text{C}$  for the duration of the reaction. The pH of the mixture, initially at 4.5, changed to 3.0 during the reaction. The samples used were collected during the phases of oxidation procedure and were differentiated according to the different percentages of  $\text{H}_2\text{O}_2/\text{FeSO}_4$  used [43]. Six samples were prepared at different concentrations of  $\text{FeSO}_4$  and  $\text{H}_2\text{O}_2$  as shown in Table 1. Sample 1 is the reference sample and was not treated with either  $\text{FeSO}_4$  or  $\text{H}_2\text{O}_2$ . Samples 2, 3 and 4 had the same percentage of  $\text{FeSO}_4$ , 0.05%wt, but different concentrations of  $\text{H}_2\text{O}_2$ , i.e., 0.002%wt, 0.015%wt, and 0.02%wt respectively while for samples 5 and 6,  $\text{FeSO}_4$  was added in the percentage of 0.04% and 0.03% respectively and the  $\text{H}_2\text{O}_2$  percentage was constant at 0.002%.

**Table 1**  
Different percentages of H<sub>2</sub>O<sub>2</sub> and FeSO<sub>4</sub> used for treating the waste samples in FENTON process

Sample	% FeSO <sub>4</sub>	% H <sub>2</sub> O <sub>2</sub>
CD1	/	/
CD2	0.05	0.002
CD3	0.05	0.015
CD4	0.05	0.02
CD5	0.04	0.002
CD6	0.03	0.002

Dynamic rheological analysis, including viscosity measurements (steady state), Time cure and Frequency Sweep experiments, were recorded from room temperature up to 120 °C.

All rheological measurements were carried out using an SR5000 rheometer (Rheometrics, Piscataway, NJ, USA) controlled by shear stress and equipped with plate geometry (2 mm gap, 25 mm diameter).

### 3. Results and discussion

Bitumen with a penetration grade of 50/70 was used and this bitumen was aged by means of standardized oxidative processes Rolling thin film ovens (RTFOT) according to the EN 12607-1 standard [44]. This was done in order to create an “aged” reference to be treated with the additives

Both additives were added to the aged bitumen at a dosage of 2% by weight of the binder [45, 46].

This percentage was used because it appears to be the one commonly used for the regenerating additives available on the market. The resulting samples were analysed by dynamic and stationary rheological measurements to understand the effects on the mechanical properties and also on the structural and morphological variations induced by the single additives on the bitumen itself.

#### 3.1. Viscosity tests

The rheological analysis was done more thoroughly by recording the mechanical spectra.

To study the effect of the various additives on the rheological behaviour of bitumens, viscosity measurements were carried out at different temperatures for all the samples. Figure 2 shows the viscosity values for the various samples at the temperatures of 60, 80, 100, 120 °C. First of all, it must be noted that the general sharp decreasing of the viscosity with temperature is expected. This is the consequence of the disordering effect of temperature in soft matter since the thermal motion can easily loosen the bonds hold up by weaker interaction, which are quite common in soft matter and complex systems. As it can be seen, the only sample that brings a significant increase in viscosity and consequently shows good capabilities as a viscosifier is the CD2 sample. Its values appear to be higher while all the other samples show small variations with respect to the reference systems. It worthy to note that this speculation is valid at different temperatures (60, 80, 100, 120 °C).

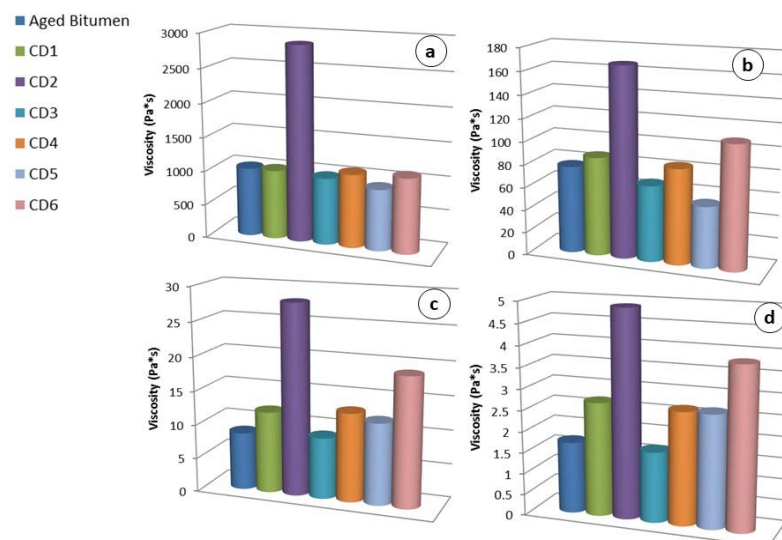


Fig. 1. Viscosity (steady state) of all the samples at different temperatures: a – 60 °C; b – 80 °C; c – 100 °C; d – 120 °C.

As a consequence of this finding, CD2 will be selected as the representative of all the FENTON-treated samples in the other tests.

We can try to give an interpretation of why the relative influence of the different additives does changes with temperature. We think that the lower amount of hydrogen peroxide in CD2 sample can guarantee disruption of bigger molecules (fat, proteins) into small components (fatty acids, peptides, alcohols) which are responsible for the observed behaviour (see for example the effect of polysaccharides [25]) preserving their integrity without further molecular fragmentations. Further oxidation would further deteriorate the organic waste to smaller chemical species with no action in the bitumen structure. Of course, due to the heterogeneities of the organic materials involved, the reaction pattern is complex with many chemical species involved, and this justifies why the relative influence of the different additives change with temperature.

### 3.2. Anti-aging tests

The effect of an anti-aging agent is evaluated by observing the plastic-fluid transition temperature as probed by rheometry [47–50]. Upon increasing temperature,  $G'$  monotonously decreases, but at a certain temperature it suddenly drops. This is the temperature at which the binder can be considered almost as a Newtonian fluid, so from the microscopic point of view, it can be intended as the temperature at which the thermal motion is sufficiently high to destroy the whole structure and therefore no storage of energy can be afforded by the sample.

Now, aging generally causes an increase in the transition temperature due to the oxidation of components; an additive must limit this phenomenon, maintaining the transition temperature, after aging, very similar to that of non-aged bitumen.

From Table 2, it can be seen that the organic waste did not bring any significant improvement compared to the starting bitumen leaving the transition temperature almost unchanged. The additive obtained from FENTON treated organic waste, CD2, at the same concentration 2% demonstrated a stricter interaction with bitumen, increasing the transition temperature of the unaged bitumen as well as the aged bitumen. However, the increase in the transition temperature when passing from unaged to aged bitumen (about 6 °C) is about of the same magnitude as in the case of non-additivated bitumen and the CD1 – additivated bitumen (4 °C).

**Table 2**

Transition temperatures (in °C) for unaged and aged bitumens with and without CD1 and CD2

Additive	Unaged	Aged bitumen
---	74.2	78.3
CD1 2%	74.3	78.6
CD2 2%	78.3	84.2

The tests were carried out using RT-FOT for 75 min due to the fact that road pavement administration agencies (such as ANAS in Italy, NCAT and NAPA in U.S.A., AIA in U.K., etc.) recommend that the difference between unaged bitumen and bitumen aged with RT-FOT be monitored closely and so there would have been no need to age the bitumen for 225 min with RT-FOT [51]. The test highlights that no anti-aging effect has been revealed but CD2 can be used to increase the resistance of bitumen to temperature.

### 3.3. Regeneration Tests

Regenerating agents are additives to be added to recycled asphalt (RAP), allowing its reuse in the production of new road pavements with a partial recovery of the pristine characteristics. To evaluate this effect, a 50/70 bitumen was aged by RTFOT for 225 min, and then 2% of CD1 or CD2 was added to the bitumen under stirring at 150 °C and left to mix for 15 min.

Time cure tests were carried out on these samples and the pseudoplastic-to-fluid transition temperatures were recorded. They are shown in Table 3.

Table 3 shows that the addition of CD1 or CD2 at 2% brings no regeneration to the oxidized bitumen since their addition does not bring the transition temperature value of the aged bitumen closer to the value of the pristine bitumen (74.2 °C).

**Table 3**

Transition temperatures for unaged and aged bitumens

Sample	Transition temperature (±0.5 °C) Bitumen 50/70
Bitumen 50/70	74.2
Bitumen 50/70 RTFOT 225	85.5
Bitumen 50/70 RTFOT 225 +2% CD1	85.0
Bitumen 50/70 RTFOT 225 +2% CD2	90.0



### 3.4. Frequency sweep tests

To understand whether the added compounds are engaged in effective interactions with the components of the bitumen thus stabilizing or destabilizing the supramolecular network, frequency sweep tests have been made and analyzed in the framework of the colloidal gel model.

According to the theory of Bohlin [52] and Winter [53], widely reported in literature as the “weak-gel model” [54], which can be applied in the context of the study of viscoelastic materials like bitumen [55], the material is characterized by a cooperative arrangement of flow units connected by weak physical interactions that cooperatively ensure the stability of the structure. This model matches also the complex inter-molecular structure typical of bitumen, where molecules are arranged together to form assemblies at various levels of aggregation and complexity [56], which are held up by weak interactions. In this model, the number of flow units interacting with each other to give the observed flow response  $z$ , can be derived by the use of the following equation:

$$|G^*(\omega)| = \sqrt{G'(\omega)^2 + G''(\omega)^2} = A\omega^{\frac{1}{z}}$$

where  $A$  is a proper constant related to the overall stiffness or resistance to deformation of the material within the linear viscoelastic region at an angular frequency of 1 rad/s, and  $G'$  and  $G''$  are the real and imaginary parts of complex modulus ( $G^*$ ) which are derived by rheological measurements.  $G'$  represents the elastic response of the material (storage of energy) under oscillation, and  $G''$  represents the inelastic contribution (dissipation of energy) during the same deformation.

In few words,  $A$  represents the force of interaction between the rheological units and  $z$  the coordination number, i.e. the number of rheological units interacting with a reference unit.

The derived values, calculated from the non-linear fitting of viscoelastic data to equation, are reported in Fig. 2. The figure shows a higher impact of CD2 on both  $A$  and  $z$  parameters, which confirms its more effective interactions with the structure of the bitumen.

### 4. Conclusions

Organic waste materials can be used in bitumen and asphalt concrete production with great benefits,

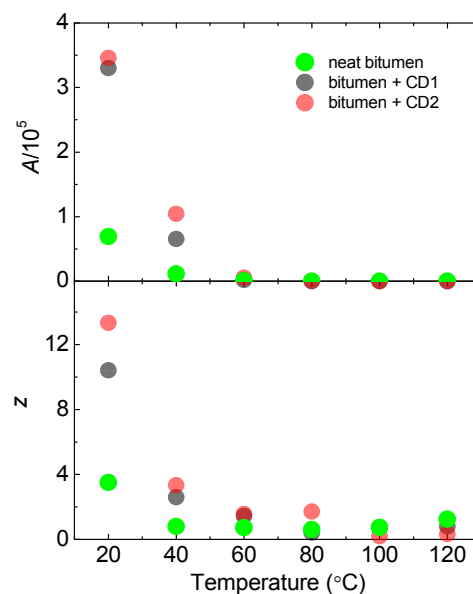


Fig. 2.  $A$  and  $z$  values as derived through equation for the neat bitumen, and that additivated by CD1 and CD2. The uncertainty associated to the values is of the same order of the symbol size.

providing better asphalt pavements and helping to develop proper and efficient waste management technology. The present study was prompted by the need of safeguarding the environment through a circular, eco-friendly and sustainable economy. We showed, by rheology, bare organic waste cannot be used as an anti-aging additive or as a regenerator of “aged” bitumen but could be used, instead, as a possible filler in bituminous conglomerates since it does not modify its characteristics/performance. We also showed that the FENTON-treated waste with low amounts of hydrogen peroxide can be used very suitable as a viscosifying agent, with a high capability of interacting with the complex structure of the bitumen.

FENTON-treated waste with high amounts of hydrogen peroxide can be used, instead, as filler in bituminous conglomerates, like the un-treated waste. These effects can be used for industrial applications for which further studies are needed to quantify the scaling-up convenience.

### Acknowledgments

The Authors acknowledge financial support from the Italian Ministero dello Sviluppo Economico (MISE), through the Project “RESIFAC”: Realization and experimentation of pilot plants for fast composting of civil and industrial organic waste (Project No. CUP: B28I17000070008, “Horizon 2020” – PON I&C 2014-2020).

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