Reuse of paint sludge in road pavements: technological and environmental issues

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Abstract

This paper presents some of the results obtained in a research project focused on the use of paint sludge as a modifying agent in the production of bituminous binders for paving applications. The scope of the project was to provide an innovative, sustainable and profitable solution to waste management problems which automotive industries have to face worldwide as a result of the production of paint sludge. Following laboratory investigations carried out with the cooperation of an Italian automotive company, the authors applied for a patent which defines the industrial process for the production of paint sludge modified binders. This papers highlights some of the critical aspects which affect technological feasibility and environmental compatibility of such a process. Technological issues are addressed by referring to the flow behavior of binders during production and to their resistance to segregation during storage. Environmental issues are examined by considering gaseous emissions during binder preparation and the potential leaching behavior of corresponding bituminous mixtures employed in road paving operations.

Keywords: Paint sludge, bituminous binders, road pavements, gaseous emissions.

1. Introduction

Application of paints by spraying, extensively used in the automotive industry, represents a significant source of solid waste. Nowadays Italian plants produce from 2.5 to 5 kg of paint sludge (PS) per painted car (data from Fiat Chrysler Automobiles, 2008). Presently, the main fate of PS is disposal in landfills for hazardous wastes, in some case combined with pretreatments for the immobilization of contaminants (Arce *et al*., 2010), or incineration.

Since the early Nineties several possibilities concerning the management of PS have been considered. Researchers from Ford company investigated the technical feasibility of pyrolizing PS to an activated carbon-like adsorbent, useful to reduce emissions of Volatile Organic Compounds (VOCs) from spray booths (Kim *et al*. 2001). Other works considered the recycling of the inorganic pyrolysis residue, made of metal oxides, into the same source materials from which they originated (paint fillers), and the collection and subsequent burning of pyro-oil and pyro-gas (Muniz *et al*., 2003; Nakouzi *et al*., 1998). In the mid-Nineties ASTER proposed a solution to recycle PS into ingredients for automotive sealants (Gerace *et al*., 2002). In 2007 Indian researchers developed a treatment based on consecutive rinsing with several solvents, drying, milling and sieving for the conversion of PS into a reusable paint (Bhatia, 2007). Some attempts were also made in order to test the possibility of employing PS in building materials such as cement, concrete, mortar or asphalt. PS was tested as a reactive expansion additive in shrinkage compensating concrete, owing to its content of calcium compounds, (Soroushiam and Okwuegbu, 1996) and, in another experience, as a filler in asphaltic mixtures (Martinez-Gonzales, 1998). Results of this last investigation showed that incorporation of sludge in the mixtures improved their stability and satisfied ecological prescriptions of the local Environmental Agency.

With the support Fiat Chrysler Automobiles, the authors studied the use of PS in road pavements by focusing on the development of a process for the production of paint sludge modified binders (PSMBs). Such a process, which was the subject of a request for patent, entails the drying and milling of PS retrieved from painting plants followed by mixing with bitumen. Successful validation of the performance-related properties of PSMBs and of their corresponding bituminous mixtures was performed by means of an array of laboratory tests which considered PS of different types taken from several plants (Ruffino *et al*., 2011; Vercelli *et al*., 2016, Dalmazzo *et al*., 2017).

Key aspects which were taken into account in the design of all phases of the patented PSMB production process include its technological feasibility and environmental compatibility.

This paper presents some of the results obtained during the research project with respect to the abovementioned issues. Measurements and analyses which were considered relevant for the assessment of technological feasibility include evaluation of PS physical characteristics (which affect selection and performance of the drying system included in the PS processing cycle), of PSMB high-temperature viscosity (which influence the choice of the most appropriate type of full-scale mixer and may pose specific requirements for pumping in hot-mix plants), and of PSMB storage stability (which may lead to specific recommendations on storage, static or with agitation). Environmental issues were addressed by considering PS chemical composition (which defines the baseline for any further measurements), gaseous emissions during PSMB production (which are relevant in the context of the potential risk for workers), and leaching behavior of bituminous mixtures containing PSMBs (which needs to be considered in order to check potential impact on ground water reservoirs).

2. Materials and methods

PS samples considered in this paper were taken from two different plants and were both of the basecoat (PS A) and clearcoat (PS E) type. These materials were preliminary dried and milled and thereafter characterized in terms of their elemental (C, H, N, S, Cl) and metal (Al, As, Ba, Cd, Co, total Cr, Cu, Fe, Mn, Ni, Pb, Ti, Zn) content, heating value and residual ash content after combustion at 600°C. Carbon, hydrogen, nitrogen and sulphur contents were determined by using a Flash 2000 ThermoFisher Scientific CHNS analyzer. Metals were determined using a Perkin-Elmer Optima 2000 ICP-OES after a two-stage digestion in a Milestone 1200 Mega microwave oven in the presence of sulphuric acid (97%, Sigma Aldrich, 3 ml for 250 mg of sample, first stage) and nitric acid (65%, Sigma Aldrich, 3 ml for 250 mg of sample, second stage). The higher heating value (HHV) was evaluated by means of a calorimetric bomb; ashes at 600°C were obtained as the residue of combustion in the calorimetric bomb. Full characterization of PS samples was performed twice (in October 2013 and May 2015) in order to highlight possible variations due to changes in composition and/or of the plant pretreatment and recovery processes.

In order to reduce PS to powder form, suitable for binder modification, available samples were dried at 105°C and at 150°C and thereafter subjected to milling. PSMBs were prepared with a mixing procedure composed of preliminary binder heating in oven at 150°C, homogenization of the neat binder in the mixer with a spindle speed of 50 rpm, pouring of PS powder, homogenization of the blend at 450 rpm, and effective mixing at 150°C for 30 minutes with a spindle speed of 500 rpm. Blends considered in this paper were prepared with a PS dosage equal to 20% (b.w. of bitumen) and by employing two different 50/70 penetration grade base binders (identified as X and Y) provided by two Italian refineries.

VOCs in gaseous emissions sampled during PSMB production were monitored in time by means of a Ionscience Tiger Instrument up to a maximum mixing time of 50 minutes.

Binder viscosity was measured at 135 and 165°C by means of a Brookfield DV-III Ultra viscometer equipped with SC4-21 coaxial cylinders (ASTM D4402). Viscosity values were recorded at a constant shear rate equal to 18.6 s^{-1} (20 rpm).

Storage stability tests were performed on all PSMBs according to the procedure described in EN 13399. The test consists in the conditioning of a binder sample in oven at 180°C for 3 days followed by viscosity tests carried out on the top and bottom parts of the specimen at 135°C. The occurrence of phase separation is identified by comparing the two viscosity values which in fact should be approximately equal in the case of a homogeneous non-segregated binder.

Bituminous mixtures containing PSMBs were produced in the laboratory by combining natural aggregates with a fixed amount of binder (5.3% b.w. of dry aggregates). Target aggregate gradation and binder content of the mixtures considered in this paper were selected according to the acceptance limits provided by the Italian performance-based technical specifications for coarse dense-graded wearing course layers (CIRS, 2001). The job-mix formula obtained as a function of size distributions of available aggregate fractions was constituted by 21% of the 10/15 mm size fraction, 33% of 3/8, 30% of 0/5 and 16% of filler.

In order to evaluate the possible release of contaminants in the environment due to the presence of PS in bituminous mixtures prepared with PSMBs, the leaching test was performed (UNI EN 12457-2). Samples were taken from laboratory-prepared slabs (EN 12697-33) which were subjected to analysis before and after wheel tracking tests (EN 12697-22), with the purpose of highlighting possible additional effects due to traffic loading. Metals in the recovered leachate were determined by using a Perkin-Elmer Optima 2000 ICP-OES.

3. Results and discussions

Results of chemical and physical characterization tests carried out on PS A and E are shown in Tables 1 and 2.

As expected, significant variations of the total solid content (TS) were recorded when comparing different plants and/or samples taken from the same plant in different periods (Table 1). This is the result of the well-known inconsistency of PS production, which is affected by changes that occur in the treatment processes that are implemented and constantly adjusted for PS recovery. In particular, in the case of the plant where sample A was taken, the high increase of TS was probably due to a variation of the PS dehydration process. During the research project the authors pointed out that such a variability should be significantly reduced since it may affect the efficiency and the energy requirements of the PS drying phase included in the patented PSMB production process. The industrial partner involved in the project acknowledged the importance of homogenizing PS pretreatment processes and promoted actions to move towards such a target.

The other experimental results listed in Table 1 are not directly related to the technological and environmental issues discussed in this paper. Nevertheless, they are listed for all investigated PS samples since they provide evidence of the potential affinity with bitumen (high values of total volatile solids, TVS), of the constancy of elemental composition (C, H, N and S) and of the excellent heating performance (remarkable low and high heating values, LHV and HHV) (Vercelli *et al*., 2016).

Results of heavy metals analyses are reported in Table 2. Basecoat and clearcoat PS were found to be clearly different in terms of Chromium, Cobalt, Copper and Titanium. In particular, the basecoat products showed values up to two orders of magnitude higher than the clearcoat ones, probably as a result of the presence of such metals in the coloured pigments contained in basecoat paints. It was also observed that in time concentrations of some metals changed, as in the case of Aluminium for PS A (which decreased with the abovementioned change in dewatering treatment) and of Lead for both types of PS.

Figure 1 displays the results obtained by monitoring concentration of VOCs during laboratory PSMB mixing. It can be observed that after a mixing time of 50 minutes VOC concentration of binders containing PS was approximately equivalent to that of base binders X and Y. Thus, in the general philosophy of the PSMB production patent the mixing phase was considered as a supplementary PS treatment phase during which most of the VOC emissions should occur, with the elimination in controlled conditions of residual solvents, if any. From Figure 1 it should also be noticed that VOC concentration was definitely lower in the case of PSMBs produced by employing PS treated at a higher temperature (150°C instead of 105°C) before milling. Consequently, in the PSMB production patent it was stated that such an aspect of pretreatment should be adequately considered for the purpose of limiting environmental impacts.

Results of viscosity measurements performed at 135°C and 165°C are listed in Table 3, which highlights the different effect of PS on the viscosity of base bitumen. Clearcoat-derived PS E provided a stiffening contribution to bitumen X bitumen. On the other hand, basecoat-derived PS A had different effects depending upon binder type: it fluxed binder X while it slightly stiffened binder Y. It should also be observed that that the stiffening effect of PS A was found to be a function of PS drying temperature. In fact, by increasing pretreatment temperature (from 105°C to 150°C), it can be hypothesized that most of the solvents were removed from the blend, with its viscosity that tended to that of the base bitumen. Finally, it should be emphasized that regardless of PS type and pretreatment temperature, viscosity values were such that PSMBs may be used in hot-mix plants with no additional requirement on standard plumping and mixing operations.

Figure 2 shows the results of viscosity tests performed on PSMBs after storage stability tests. It can be noticed that binders containing PS dried at 105°C were subjected to phase separation, especially in the case of those prepared by the clearcoat product E, which displayed a strong difference between the viscosity of top and bottom samples. Although this may seem a problem for full-scale implementation, it can be easily solved by adopting, as in the case of other paving binders (e.g. those containing crumb rubber), an agitation system during the storage. It should also be noticed that binders containing PS dried at 150°C apparently showed a good storage stability. However, supplementary tests are required to confirm this observation.

As previously mentioned, leaching tests (UNI EN 12457-2) were performed before and after wheel tracking tests in order to evaluate the possible release of contaminants due to the presence of PS in bituminous mixtures. Results of these tests, displayed in Table 4, show that the concentration of metals and anions in the leachate of bituminous mixtures containing PSMBs (identified with codes UXA20 and UYA20) is substantially equivalent to that of standard bituminous mixtures (indicated as UXN). Moreover, repeated traffic loading did not show any relevant effect on recovered leachate.

Table 1. PS characterization: Total Solid content (TS), Total Volatile Solid content (TVS), elemental analysis (CHNS), high and low heating values (HHV, LHV).

Paint Sludge	TS	TVS	C	H	N	S	HHV	LHV
	%	%	%	%	%	%	kcal/kg	kcal/kg
A (10/2013)	26.8	75.0	50.3	6.92	3.19	0.08	5790	4230
A (05/2015)	41.3	74.5	50.0	6.68	2.96	< 0,01	-	-
E (10/2013)	39.2	94.5	58.2	8.25	6.8	< 0.01	6680	4820
E (05/2015)	33.6	89.6	55.2	7.86	6.19	< 0.01	-	-

Table 2. PS characterization: heavy metals analysis.

Paint Sludge	Cd	Cr	Fe	Mn	Zn	Ni	Al	Co	Ba	Pb	Cu	Ti
	mg/kg	mg/kg	%	mg/kg	mg/kg	mg/kg	%	mg/kg	mg/kg	mg/kg	mg/kg	%
A (10/2013)	1.3	64.1	0.65	338.0	371.0	6.9	1.45	140.0	7.2	8.7	94.4	6.820
A (05/2015)	ND	23.5	0.432	28.4	307	4.12	0.420	0.256	11.3	0.079	62.2	6.62
E (10/2013)	0.9	9.8	0.08	8.6	186.0	5.2	1.02	0.9	41.7	9.9	7.1	0.004
E (05/2015)	ND	7.72	0.109	34.2	251	10.2	0.737	0.439	17.4	0.242	10.6	0.115

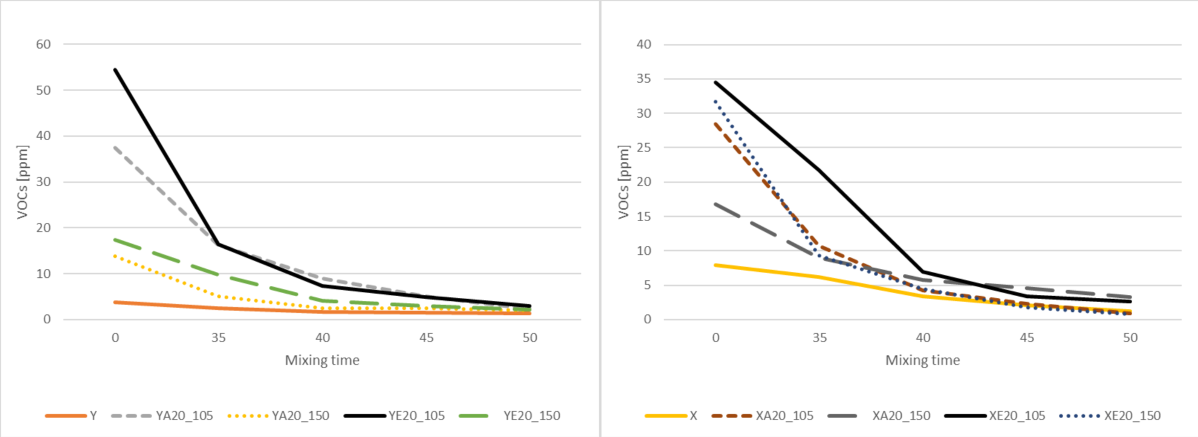

Figure 1. VOCs trend in gaseous emissions for PSBs with bitumen X and Y and PS A and E dried at 105 °C and 150 °C.

Table 3. Viscosity of PSMB modified with binder X and Y

	Viscosity [mPa·s]	
Material code	**@135°C**	**@165°C**
X	465	125
XA20_105°C	372	100
XA20_150°C	400	110
XE20_105°C	720	192
Y	432	115
YA20	442	115

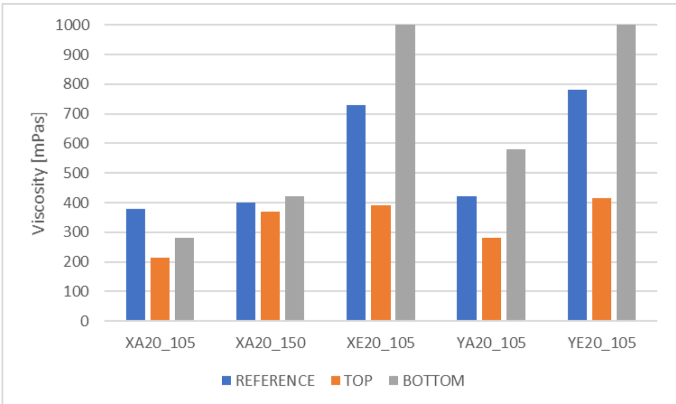

Figure 2. Storage stability for PSBs with bitumen X and Y and PS A and E dried at 105 °C and 150 °C.

Table 4. Leaching tests results for PSBs bituminous mixtures.

Parameter	Unit	UXN	UXN rutted	UXA20	UXA20 rutted	UYN	UYA20
Ba	microg/l	0.338±0.015	0.812±0.19	3.10±0.05	2.73±0.29	2.59±0.05	4.27±0.08
Cu	microg/l	< 0.54	< 0.54	< 0.54	< 0.54	< 0.54	< 0.54
Zn	microg/l	< 0.18	< 0.18	< 0.18	< 0.18	2.16±0.12	1.24±0.28
Co	microg/l	< 0.70	< 0.70	< 0.70	< 0.70	< 0.70	< 0.70
Ni	microg/l	< 1.5	< 1.5	< 1.5	< 1.5	< 1.5	< 1.5
As	microg/l	< 5.3	< 5.3	< 5.3	< 5.3	< 5.3	< 5.3
Cd	microg/l	< 0.25	< 0.25	< 0.25	< 0.25	< 0.25	< 0.25
Cr	microg/l	< 0.71	< 0.71	< 0.71	< 0.71	< 0.71	< 0.71
Pb	microg/l	< 4.2	< 4.2	< 4.2	< 4.2	< 4.2	< 4.2
Ti	microg/l	< 0.38	< 0.38	< 0.38	< 0.38	< 0.38	< 0.38
pH		9.55±0.04	9.50±0.01	9.55±0.01	9.48±0.01	9.19±0.04	9.13±0.01
EC	microS/cm	31±1	33±1	31±1	33±1	14±1	18±1

4. Conclusions

The experimental results presented in this paper show that the use of PS as a modifying agent of bituminous binders can be an attractive alternative to landfilling or incineration. The production of PSMB binders is feasible from a technological point of view and requires only minor adjustments of standard processes currently employed in hot-mix plants. Moreover, by making use of the PSMB production process which has been patented by the Authors, there are no true environmental issues related to gaseous emissions and release of contaminants in ground water.

As a result of these encouraging outcomes, the research project developed by the authors has entered in its full-scale implementation stage, in which pilot sections and larger field trials will be constructed and monitored in time. Further improvements will also be sought, with the fine-tuning of mixing processes and the possible performance enhancement by means of supplementary additives.

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