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Tyre Recycling with Thermal Solvolysis Method Using Microwave Radiation

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Used tyres are one of the most widespread types of waste and one of the polymer materials which are most difficult to recycle. There are many different applications for used tyres today. Part of the tyres can be retreated and re-used, but most of the used tyres are recycled.

By dissolving the tyres, it is possible to filter out various chemical substances. These substances coming from the used tyres can be used for creating a new material or improving an existing one. One of the technologies involves adding chemically recycled tyres to the bitumen used in road pavement construction. The substances obtained from the recycled tyres can be used in the water proofing of bitumen. Methods of pyrolysis and solvolysis are used for recycling tyres into raw materials.

In this research tyre recycling with thermal solvolysis is studied by using a conventional oven and microwave oven in order to obtain waterproofing materials with improved exploitation properties. Promising results are obtained in the pre-studies on the tyre recycling in the microwave allowing to reduce energy consumption before thermal processing of tyres as well as during the thermal processing process compared with methods involving heating in traditional furnace.

Dissolving time of the recycled tyre pieces depending on the size of the pieces as well as energy consumption depending on furnace type, operating temperature and time have been assessed during the experimental part. Adhesive properties of the rubber-bitumen composite obtained during the experiments were tested. After testing the adhesive properties on the concrete surfaces it can be concluded that rubber-asphalt composite obtained in a microwave oven has the best adhesion because no waterproofing peeling has been found.

KEYWORDS: thermal solvolysis, rubber-bitumen composite.

Introduction

During the last decades all countries are paying increased attention to such problems as growing production and household waste stream including the used tyres and waste utilisation or recycling. Used tyres are one of the most widespread types of waste and one of the polymer materials which are most difficult to recycle.

Used tyre recycling problem is essential with regard to the ecology as the used tyres tend to accumulate in the locations, where they have been used (car services, aerodromes, production and public utility companies). When transported to the landfill sites (Fig. 1) or thrown out in the surrounding area, the used tyres create long-term pollution of the environment due to their resistance to such environmental factors as impact of sun, oxygen, ozone as well as microbiological impact.

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Fig. 1

Used tyres at landfill site (http://www.business-equipment.ru/images/utilizaciya-avtomobilnyh-shin.jpg, 23.11.2015)

Today there are several options how the used tyres could be treated. Small part of the used tyres is used for decoration purposes but the biggest part is recycled (Urtane S., 2012). Every year, around 2.5 million tons of tyres wear out only in Europe, and their recycling rate reaches 90%. Most of the tyres (about 40%) are burned for energy. Slightly lower proportion of the used tyres (about 30%) are shredded in crumbs for further use either in shredded form and 20% of the used tyres are restored and exported to be reused.

Currently a developing trend is used tyre recycling by applying the methods of chemical recycling. The chemical recycling involves irreversible changes of the tyres and collapsed structure of the polymers.

By melting the tyres it is possible to filter out various chemical substances added by the tyre producers. There are some additives that increase tyre freeze resistance or thermal resistance. Other additives increase their resistance to chemical substances (Zhitov R., 2013). By knowing how to preserve all these substances it is possible to change the composition of some material thus improving its properties (Baldino Noemi et al, 2013), (Ortega F.J. et al, 2015). For example, using bitumen together with recycled tyres it is possible to obtain lower costs and higher quality thus providing possibilities for the improvement the road surface quality.

There are several methods for used tyre recycling. Tyres burn easily, therefore the initial method for handling the used tyres was to burn them. Tyre burning is related to high emissions of fly ash that remain on the walls of chimney or in filters as well as on the leaves of plants and trees creating environmental pollution.

Applying the method of mechanical shredding the tyres are cut into small pieces. During the recycling process the rubber powder is obtained, which has an even increasing number of possible applications. The most important quality indicator of the powder is the size of particles ranging from 180 mkm to 12 mm (http://www.videsvestis.lv/content.asp?ID=41&what=12, 13.10.2015). Applying the granulation of tyres it is possible to use the rubber granules in the process of paving and asphalt production. This type of asphalt contributes to a longer service life of the new tyres.

Applying pyrolysis for the tyre recycling in a high temperature without air supply results into their decomposition, namely, gas condensate mixture, fly ash and metallic cord are obtained as the final products. Heating ability of the obtained condensate is equal to those of the fuel oil, therefore it can be used as the heating fuel (Papin A., 2015). Fly ash can be used as a filler in the rubber and plastic recycling but metallic cord - in the metal scrap recycling. Utilisation and further recycling of the used tyres proceeds in several stages: tyre tread and beads are removed with the side cutting equipment (for the dense stocking of the tyre parts), tyre parts are placed on the retort, the lid of the retort is closed and it is placed in the pyrolysis furnace set at 450^{0}C (http://ztbo.ru/o-tbo/stati/piroliz/piroliz-shin-i-pokrishek, 15.10.2015). Pyrolysis gas is formed during the decomposition; it moves through the cooler forming a pyrolysis oil. Part of this oil is redirected to the burner in order to sustain the pyrolysis process (Papin A., 2015).

One of the most promising used tyre recycling technologies is thermal solvolysis, which has been a research object of this scientific research. Thermal solvolysis of rubber crumbs proceeds in lower temperatures compared to the pyrolysis process. It allows to increase the amount of liquid product as less gas is formed as well as it is possible to modify quality of liquid products due to different solvent composition. Rubber decomposition with solvolysis requires shorter period of time compared to pyrolysis, therefore it is possible to process bigger amount of rubber (Andrejkov E. et al, 2005).

Materials and methods

Major and the most important component is rubber obtained from caoutchouc (Kalninsh M., 1988). Tyre components are given in Table 1.

Two granulometric fractions of the tyres have been used in the experimental part: 5-12mm (Fig. 2) and 0.2-1mm (Fig. 3). They were prepared using mechanical cutting equipment.

Table 1

Composition of tyres (raw materials)

Composition of tyres	
Rubber	36%
Fillers (carbon black, microsilica)	37%
Steel and textiles	18%
Plasticizers (substances added to ensure elasticity and freeze resistance of the product)	3%
Sulphur	1.3%
Zinc oxide	1.2%
Chrome, nickel, lead, cadmium	3.5%

Fig. 2

Rubber crumbs with the size 5-12mm

Fig. 3

Rubber crumbs with the size 0.2-1mm

Pure bitumen without foreign matters and caoutchouc additions is used for tyre dissolution. Properties of the bitumen used in the experimental part are given in Table 2.

Rubber crumbs from the recycled tyres constitute 20% ±0.5% from the total weight of the material used in the research aimed at assessing the interaction of bitumen and caoutchouc. Amount of the raw materials is given in Table 3.

Rubber crumbs without metal frame were used as they were intended for heating in microwave. All compositions have been mixed during the heating process in order to maintain even heat distribution among various components. Due to the high levels of fume the experiments were carried out in the premises with a special ventilation. Electric power consumption during the experiment has been measured with the electricity meter. Plan of the experiments is given in Table 4.

Table 2

Properties of the bitumen

Properties	Measurement units	Research method	Value
Penetration index at the temperature 25^{0}C	0.1 mm	EN 1426	20-30
Softening temperature	^{0}C	EN 1427	≥ 64
Fragility temperature	^{0}C	EN 12593	≤ -5
Paraffin content	%	EN12606-1	≤ 2.2
Dynamic viscosity at the temperature 60^{0}C	Pa*s	ASTM D 4402	≥ 5000
Inflammation temperature	^{0}C	EN ISO 2592	≥ 240
Density 25 ^{0}C	g/cm^3	EN ISO 15326+A1	1,0 – 1,1
Undissolved particle content	% m/m	ASTM D 4124	≥ 18
Weight changes after ageing	% m/m	EN 12607-1	≤ 0.4
Temperature increase after ageing	^{0}C	EN 1427	≤ 8.0
Penetration decrease after ageing	%	EN 1426	≤ 35

Table 3

Amount of the raw materials used

Number of the experiment series	Bitumen, g (%)	Rubber crumbs, g (%)	Total, g (%)
1.-4.	160g (80%)	40g (20%)	200g (100%)

Table 4

Marking of samples

Sample number	Number of the experiment series	Heating equipment	Amount of bitumen, %	Amount of rubber crumbs, %	Crumb size, mm
I	0	-	100	-	-
II	0	-	100	-	-
III	1.	Furnaces	80	20	5-12
IV	1.	Furnaces	80	20	5-12
V	2.	Furnaces	80	20	0.2-1
VI	2.	Furnaces	80	20	0.2-1
VII	3.	Microwaves	80	20	5-12
VIII	3.	Microwaves	80	20	5-12
IX	4.	Microwaves	80	20	0.2-1
X	4.	Microwaves	80	20	0.2-1

Solvolysis recycling proceeds in two stages: thermal solvolysis and further thermal oxidation with air. Heavy oil residues can be used as solvents, such as residues from the oil distillation (gudrons), oxidesed gudrons, etc. The most ration option from the economic point of view would be using the cheapest products, namely, gudrons. As the process takes place under the atmospheric pressure, the solvent should not contain any fractions that melt under the temperature of 360^{0}C (Andrejkov E. et al, 2005).

Fly ash particles and liquid products obtained from the used tyres with the thermal solvolysis serve as modifiers for the bitumen materials (Lebena P., 2004).

By changing the proportion of rubber crumbs and oil residues as well as conditions of the solvolysis and thermal oxidation it is possible to obtain various types of bitumen with different characteristic curves corresponding to the basic indicators of the existing norms and standards.

Solvolysis process happens at high temperatures (200-300^{o}C) or by using special additives (which are usually very expensive) promoting devulcanisation of rubber at lower temperatures. The process results in the rubber-bitumen composite used as a binder for asphalt (Amosova I., 2005).

To melt the rubber crumbs, the device, which is equal to the modern microwave operating at the microwave frequency 2450 Mhz and equipped with the mechanical mixing option and temperature indicator, have been used. According to the literature sources (Alekseenko V. et al, 2013) the crumbs melt at the temperature 230±5°C.

Result

The initial temperature of all samples was +20±2^{o}C and each composition of the bitumen and used tyre mix has been heated until 230±5^{o}C. It took about 200 minutes to reach the necessary temperature in the conventional electric furnace, while it took only 6-7 minutes in the microwave. Then the experiment continued with keeping the samples at the temperature 230±5^{o}C while the rubber crumbs completely melted and incorporated into the bitumen.

The rubber crumbs melt in 235-250 minutes in the furnace (Fig. 4), while in takes only 18-25 minutes in the microwave. Due to the consumption of the electric energy the production costs are significantly higher using the electric furnace compared to the experiments in the microwave. From the obtained information it can be concluded that for melting 0.1 kg of the bitumen and tyre mix 3.2 kWh of the electric energy is used in the electric furnace, while only 0.275 kWh –in the microwave (Table 5).

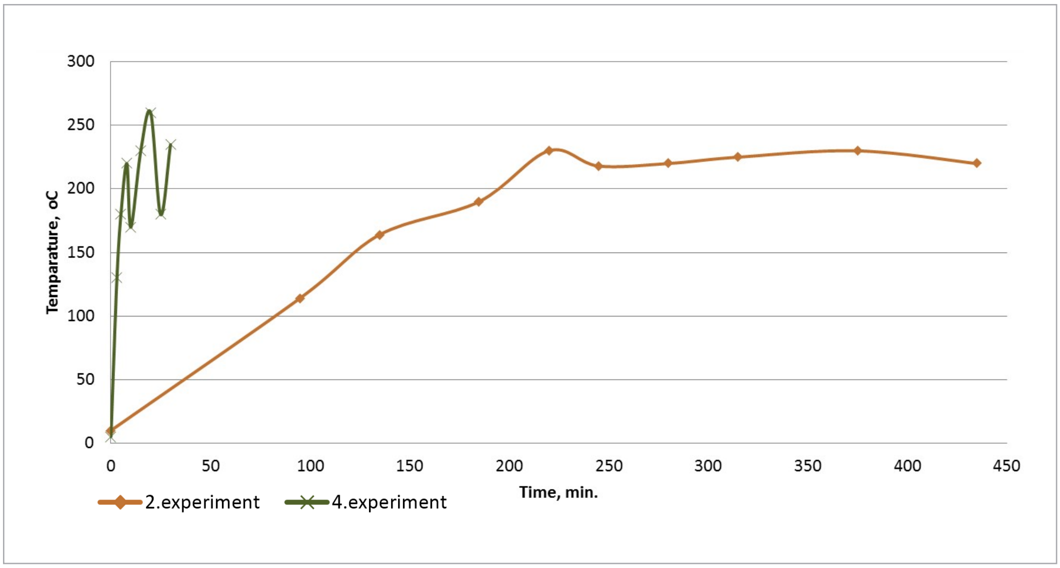

Fig. 4

Graphic relation of the consumed time (min) and temperature in the experiment series 2 and 4

Table 5

Comparison of the consumed time and electric energy for the weight 0.1 kg

Equipment	Fraction of rubber crumbs, mm	Time consumed, min	Electric power consumption, kW*h
Electric furnace	5-12	222	3.329
Electric furnace	0.2-1	206	3.092
Microwave	5-12	12	0.300
Microwave	0.2-1	11	0.252

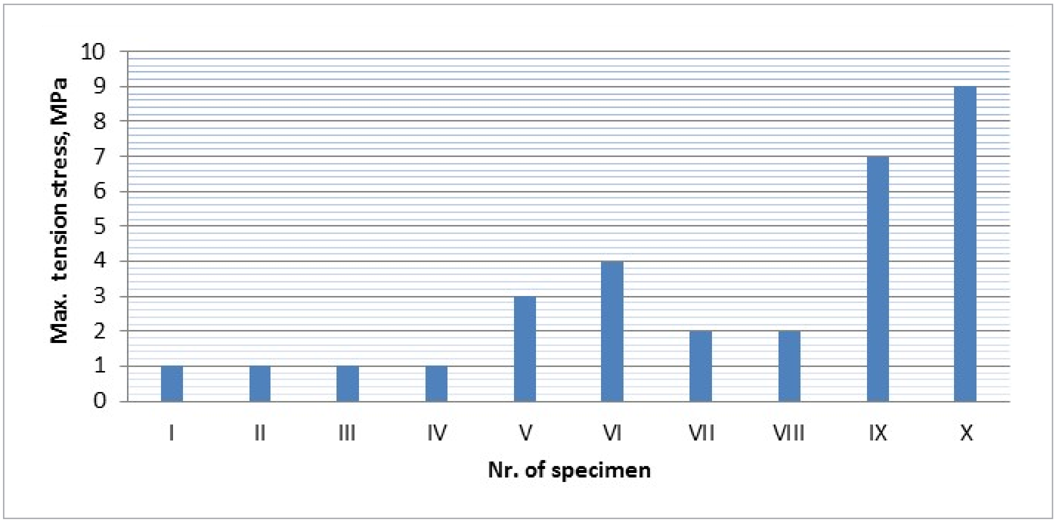

Fig. 5

Results of the adhesion tests, MPa

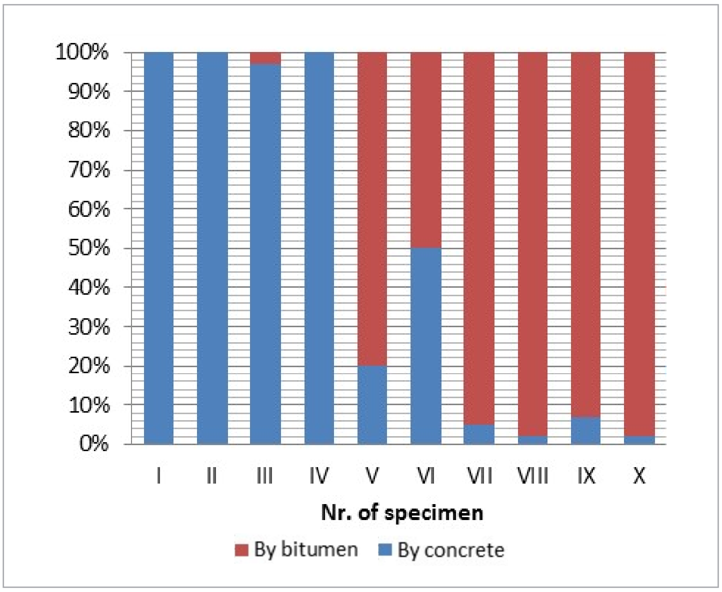

Fig. 6

Proportion of the pulling parts (on bitumen/on concrete surface)

By melting the tyres and incorporating them into bitumen the rubber-bitumen composite with the properties depending on the fillers and additives of the tyres is obtained. This material can be used in the road building industry as a binder for the road surface or in the construction industry for the water proofing of the structures.

One of the important properties of binder is adhesive property. Researchers (Somé Saannibe Ciryle et al, 2012) have investigated and made attempt to quantify the bonding quality between hot bitumen and other material.

The bonding properties of bitumen have been test basing on the standard EN 15322. To test the adhesive properties of the obtained material, 3 samples of the concrete glued with the bitumen composite material have been prepared for each composition of bitumen. The samples have been processed with the grinding machine and two pieces of concrete were glued with the different compositions of obtained bitumen. Each concrete piece has size 40x40x80 mm. Concrete specimens were glued by the surfaces with size 40x40 mm. The glued surface of the concrete samples was 1600 mm^2. All bitumen compositions and pure bitumen were heated before sealing for improving their fluidity properties.

Analysis

The experiments carried out within this research provide an idea and give a real-life experience about each of the two methods for the tyre recycling with solvolysis. It can be seen from the data included in the Table 5 that the processing of rubber crumbs in the microwave provide considerable cost advantage; however, it is related to the significantly higher release of air polluting substances (at least, as far as it can be concluded from the experiments). Unfortunately, it was not possible to determine the amount and composition of the substances released into the atmosphere in the form of fume. However, the following observations have been made:

1. During the experiments involving microwave there has been more fume than during the experiments with electric furnace;
2. Experiments involving microwave took significantly shorter time than experiments involving microwave. Therefore, it is possible to put forward a hypothesis that the total amount of fume can be even bigger using the electric furnace due to total time consumption.

The results of the adhesion tests vary; however, it is clear that the adhesive properties of the rubber-bitumen composite used together with the concrete are not lower than those of the bitumen alone (Fig. 5). In has been observed that the best adhesive properties are for the rubber-bitumen composite obtained in the microwave (Fig. 6). It could be explained with the better melting of rubber and its incorporation into the bitumen.

Conclusions

The results of the experiments show that 0.1 kg of the rubber-bitumen composite can be obtained in 11-12 minutes using the microwave, while it takes 230-250 minutes using the furnace. Therefore, it can be concluded that by using the microwave it is possible not only to shorten the melting time of the rubber but also to decrease the consumption of the electric energy around 10 times.

Results of the adhesion tests show that they differ depending on the fraction. During the experiments it was not possible to mix the rubber and bitumen evenly, which could possibly affect the adhesive strength. In general, the rubber-bitumen composite obtained in the microwave showed better results.

Used tyre recycling with solvolysis showed significant economy of the electric energy using the microwave compared to the conventional electric furnace. It allows reducing CO_2 in the used tyre recycling industry and obtaining raw materials of a good quality for the production of binder for the road surface or in the road building industry and for the water proofing materials used in various structures in the construction industry.

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