Postprint of: Sienkiewicz M., Janik H., Borzędowska-Labuda K., Kucińska-Lipka J., Environmentally friendly polymer-rubber composites obtained from waste tyres: A review, Journal of Cleaner Production, Vol. 147 (2017), pp. 560-571, DOI: 10.1016/j.jclepro.2017.01.121

ENVIRONMENTALLY FRIENDLY POLYMER-RUBBER COMPOSITES OBTAINED FROM WASTE TYRES: A REVIEW

Maciej Sienkiewicz*, Helena Janik, Kaja Borzędowska-Labuda, Justyna Kucińska-Lipka
Polymer Technology Department, Chemical Faculty, Gdansk University of Technology
Gabriela Narutowicza Street 11/12, 80-233 Gdansk, Poland

Abstract

The paper is transdisciplinary. The complex overview on changes in policy and approach to waste tyres that leads to both solving environmental problem of post-consumer tyres and creating environmentally friendlier novel materials are presented. It is shown that in the world, the amount of polymer wastes increases every year. Most of them are in the form of post-consumer tyres. Recycling and recovery of such tyres is a serious environmental problem due to their very complex structure and composition. Moreover, storage of the whole post-consumer tyres takes a lot of space in landfills and pollute the environment. However, recently it has been reported a great progress in sustainable management of waste tyres. An important role in the progress of tyre recovery has played the establishment of the restrictive regulations, which prohibits stockpiling of tyres in landfills. This legislative steps have contributed to significant impact on the progress in the development of car tyres recovery. The most important is introducing of monitoring for improper stockpiling, producer responsibility for wastes tyres and the new tax system. With the help of mentioned changes and introducing environmentally friendlier technologies it appeared that the utilization of tyres can be cost effective. The applied technology can transform post-consumer tyres into a source of energy or valuable raw materials, from which can be obtained a wide range of

---

* Corresponding Author,
Phone: + 48 0583471093, Fax: + 48 0583472694
e-mail: macsienk@pg.gda.pl

sustainable polymeric composite materials characterized by quite good mechanical and functional properties. This new approach to post consumer tyres is undertaken in U.S., China, Japan and EU. From the overview it is clear that the grinded used tyres should not be treated any more as a pollutant but rather as a source of sustainable materials. Those materials belong to modern polymer-rubber composites obtained mainly from elastomers (natural and synthetic rubbers) or thermoplastics (PE, PP, PVC). The very important factor in making good composites is get the knowledge about the influence of amount, size and morphology of rubber granulate grains on interactions that occur in composites between the polymer matrix and tyre rubber grains and thus the properties. That last item was discussed in the final part of the overview.

Highlights

- Waste tyres pose a serious environmental problem.
- Key to the recovery of tyres with high performance is good law legislation
- Used tyres can be a source of sustainable new material.
- Spent tyres can be used to produce ‘environmentally friendlier’ composites.
- Sintering of rubber granulates is a way to obtain very cheap and useful materials.

Keywords: rubber-polymer composites, rubber granulate, used tyres management, recycling of tyres

1. Introduction

The dynamic increase of rubber wastes, especially in the form of used tyres has become a serious environmental problem. The main difficulties associated with the recovery and recycling of used tyres are due to the very complex structure and composition of tyre materials. The basic component, which makes up about 70-80% of the total mass of the tyres, is vulcanized (cured by sulfur) rubber, filled by carbon black and silica. 8 types of natural rubbers, 30 types of high-quality synthetic rubbers (especially styrene-butadiene rubber (SBR), butadiene rubber (BR) and butyl rubber (IIR)) and many kinds of chemical compounds, useful in the processing and vulcanization of rubber are involved in creation of materials for tyre. Moreover, tyres contain also steel cord and fibers made from nylon, polyester and cellulose, which have to be separated from the rubber during waste tyre recycling (Ramarad et al., 2015; Schnubel, 2014; WBCSD, 2008). Difficulties in the reuse and recycling of tyres also result from the complicated structure of the rubber, which is the blend of diene rubbers crosslinked by the sulfur. Due to this the tyres are insoluble and infusible material and they cannot be re-processed in such a simple process like the thermoplastics. Moreover designing the composition and the manufacturing of tyres is oriented towards obtaining a products which have high mechanical strength, flexibility and resistance to environmental factors, resistance to biodegradation and high (100ºC) and low (-30ºC) temperature. For these reasons the sustainable recovery and recycling of used tyres, from technical point of view, is very difficult and requires the use of novel advanced technology and specialized machinery and equipment (Ramarad et al., 2015; Karger-Koscic, 2013; Sienkiewicz et al., 2012).

However, the reports published by the largest associations of producers and recyclers of automobile tyres (CRIA, ETRMA, JATMA, RMA) show that in recent years a great progress has been made in sustainable management of used tyres. Thus used tyres should not be longer regarded in the world, as a hazardous waste, but as a source of ‘environmentally friendly’ materials, which can be transformed into new tyres (retreading), energy (incineration in cement kilns, paper plants, and industrial boilers) or novel group of composite materials with a broad range of application (recycling) (RMA, 2013; ETRMA, 2014 JATMA, 2015, CRIA,2010).

One of the major method of sustainable management of used tyres is grinding them and use of obtained in this process rubber granulate as a component or filler to create new polymer composites, which are thought to be ‘environmentally friendly’ as recycling them in this way leads to decreasing of the amount of post-consumer tyres. The rubber composites are mainly prepared by waste free technology and they have quite good mechanical and functional properties with reasonable economy of material manufacturing (Ramli et al., 2013). Ground tyre rubber is usually used as a filler in thermosets, thermoplastics and virgin rubber (Yehia et al., 2003; Kakroodi and Rodrigue, 2013a; Rocha et al., 2014; Kocevski et al., 2012). It is also used to modify asphalt (González et al., 2012; Nejad et al., 2012; Navarro et al., 2014; Liang et al., 2015; Miro et al., 2015) and concrete (Cheng et al., 2012; Bravo and Brito, 2012; Huang et al., 2013; Ossola and Wojcik, 2014; Koçak et al., 2012).

1.1. Models and methods of used tyres management in some countries

In different countries and regions in the world exist different systems of used tyres management, dependent on the local, legal, political and social standards. For example, in the EU and in the U.S. for the management of used tyres recovery and control are responsible individual countries and the states. Analyzing the development of methods and systems of management of the used tyres in the world it can be concluded that the key to the success of building high-efficiency models of waste management, which allows to achieve the level of recovery and recycling of used tyres near 100% is establishing appropriate legal regulations which limit or prohibit storage of whole and grinded tyres on the landfills. A huge impact on the efficiency of the recovery of used tyres have also the model of the management and its control. In Table 1 there are shown legal regulations and models of used tyres management introduced in recent years and adopted by the major manufacturers of these wastes (RMA, 2009; ETRMA, 2011; WBCSD, 2008).

Table 1. The legislation regulation and models in management of used tyres recovery in EU27, U.S., Japan and China (JATMA, 2015; Hu et al., 2014; ETRMA, 2011; ETRMA, 2013; CRIA,2010; RMA, 2009; Li et al., 2010; Wang, et al.,2009; WBCSD, 2008).

The latest statistical data reported that the total amount of used tyres generated in the world is constantly increasing, while the largest their quantities are produced in China (approx. 5.7 million tonnes, data cover only the tyres recovered by the recycling in 2010), U.S. (3.824 million tonnes in 2013) in the European Union (3.418 million tonnes in 2012) and Japan (1.052 million tonnes in 2014) (JATMA, 2015; Hu et al., 2014; ETRMA, 2013; CRIA, 2010; RMA, 2009; Li et al., 2010; Wang, et al.,2009). However it should be noted that in these countries, the recovery and recycling of used tyres became a separate branch of the industry, which produces energy or many kinds of useful materials and brings huge profits. In addition, in the world there are constantly being developed the high-level technologies of used tyres recycling and there are built up new centers of R&D, which create the new methods of used tyres recovering (ETRMA, 2011). For this reason these countries achieved a very high levels of recovery of the total amounts of produced used tyres, respectively in U.S. 95.9%, in EU countries 95% and in Japan 88%. (JATMA, 2015; ETRMA, 2013; RMA, 2013).

The tyres become a waste at moment of their dismantling from the vehicle, then the used tyres are collected and segregated to the Part-Worn-Tyres (PWT) and End-of-Life Tyres (ELT). Part-Worn-Tyres are a separated group of used tyres which can be used again in cars without their reprocessing (Reuse Tyres) or can be subjected to a process of retreading, which aims to rebuild the damaged tyre tread. PWT can be also exported, and this refers mainly to export of tyres from developed countries to developing countries. ELT are the group of waste tyres which cannot be used for its original purpose. For this reason, they have to be recovered in a different way (ETRMA, 2011). Data presented in Fig.1 demonstrate that the major methods of disposal of End-of-Life tyres in the world are the energy recovery and material recycling. Besides this ELTs can be utilized by many different methods e. g. by retreading or pyrolysis. The individual processes of material recovery and energy recovery of used tyres and the manner of their implementation have been comprehensively and clearly described in a previous publication (Sienkiewicz et al., 2012), and in several publications issued recently (Karger-Kocsis et al., 2013; Presti, 2013; Martinez et al., 2013; Shu and Huang, 2013; Williams, 2013; Polacco et al., 2015; Ramarad et al., 2015; Torretta et al., 2015; Thomas et al., 2016).

Fig. 1 Comparison of the ways of recovery and flow of used tyres (UT) in China, U.S., Japan and European Union countries (JATMA, 2015; Hu et al., 2014; ETRMA, 2010; ETRMA, 2013; RMA, 2013; Li et al., 2010; Wang, 2009).

In the EU, Japan, the U.S. and in China the main directions of the flow for used tyres in the process of their recovery are different (Fig. 1). The decisive influence on this have market conditions and current demand for recycled products, as well as the availability of appropriate recovery technologies of used tyres. In Japan and in the US, due to the high demand for the fuel and energy, ELTs are used primarily as an energy source (tyre derived fuel) and combusted with energy recovery in cement kilns, paper plants, and industrial boilers (JATMA, 2015; RMA, 2013).

In the European Union countries the recovery of used tyres is based mainly on the recycling process what is the result of very high demand for the new materials coming from tyre recycling. In the industrial scale it is realized by the mechanical grinding of used tyres, which yields rubber materials with different degrees of comminution. Rubber granulates obtained in this process could be reused as the source of raw materials for the production of different kinds of very valuable polymer composites, manufactured mainly by blending process with natural and synthetic rubbers or thermoplastics. However, in the UE the energy recovery of used tyres is also on the high level, compared with material recovery (ETRMA, 2013).

In China, in recent years, have started the adjustment of the legal regulations in recovery of used tyres to market and environment needs. Hu et al. (2014) reported that in this country still does not exist any system to regulate the collection and segregation of used tyres and that tasks are fulfilled by the private sector. But in China the instruments of used tyres management are no complicated and mainly they are based on administrative rulings. The statistics which describe the methods of waste tyres management in China are incomplete and on their basis it is difficult to characterize the market of recovery of used tyres in this country. However, based on some papers (Hu et al., 2014; Li et al., 2010; Wang, 2009) and data published by China Rubber Industry Association (CRIA, 2010) it can be concluded that the major direction of used tyres utilization is recycling realized by dynamic devulcanization (approx. 54% of total recycling), grinding tyres and production of rubber granulates (approx. 7% of total recycling), pyrolysis and tyre oil extraction (approx. 40% of total recycling). It should be emphasized that China is the largest producer and consumer of reclaimed rubber in the world. The scale of production of that material was estimated in 2010 on approx. 3 millions ton, what represents 81% of total world production of the reclaimed rubber (Hu et al., 2014; CRIA, 2010). In addition, in China over 2 millions tones of used tyres are recovered in the processes of illegal refining oil from tyres, which are mainly performed by small illegal refinery located in rural areas. In this process there is obtained the high energetic oil used later as a fuel. It is a huge problem in China, because the illegal refineries of oil from tyres emit a huge amount of pollution and make great damage in the environment. For this reason in China it was introduced a ban on the illegal oil refining and now this method of used tyres recycling is strictly prohibited.

2. Composites and polymer compositions obtained from rubber granulate

From the materials engineering point of view, the main merits of polymer composites are their small mass, great mechanical strength and resistance to fatigue elasticity and resistance to chemical and environmental agents (Strong, 2007). In recent years it has appeared a qualitatively novel group of sustainable composites obtained from polymer wastes (especially tyre rubber), which seems to be more attractive in relation to environmental protection and pollution control. In this case, post-consumer tyres and other rubber wastes (which containing high-quality natural and synthetic rubbers) have become the source of materials for obtaining ‘environmentally friendly’ composites with utilitarian properties. The advantages of using such materials include sustainable management of the vast quantities of used articles made from polymers that are currently burdening the environment, and the reduction in the costs of composite materials produced from cheap polymer wastes (Navarro et al., 2004).

Nowadays, the amount of used tyre recycling by grinding in the EU27, U.S., Japan and China is estimated about 3,6 million tonnes/year. According to the analysis of the market of tyre recycling can be stated that rubber granulates are no longer considered as a cheap filler but as a valuable component of sustainable rubber composites which are used in wide application by the producers of molded/ and extruded products (wheels, gasket, sole), producers of playgrounds, mulch and animal bedding, producers of artificial sports surfacing and automotive industries (JATMA, 2015; Hu et al., 2014; ETRMA, 2013; RMA, 2013; Li et al., 2010; Wang, 2009).

2.1. Composites obtained from rubber granulate and natural or synthetic rubbers

The first person to carry out research on the use of ground rubber waste in the form of a rubber compound filler was Charles Goodyear, the inventor of the vulcanization process, who in 1853 patented a process for molding polymer materials obtained from a rubber granulate and natural rubber (Goodyear, 1853). Nowadays, analysis of the rubber waste recycling market indicates that the rubber industry is the leader in the management of used tyres and other rubber products; it was the first to apply rubber granulates as a partial replacement for natural rubbers in rubber compounds.

The increasing level of consumption of rubber recycling products shows that rubber granulates are treated in the rubber industry as a raw material for producing composites with practical utilitarian properties and in an economic way. Unfortunately, in some applications, particularly in products with a demand for very high quality and strength, e.g. in new tyres, the use of rubber granulates is seriously limited. That is why the rubber composites obtained using rubber granulates are in practice used to manufacture cheap articles, where strength is not a priority, such a floor tiles and other flooring materials, windscreen wipers, washers, tapes, molds, cable housings and footwear soles.

It is reported in the literature that the properties of rubber granulates depend mainly on the method of obtaining them (cryogenic or ambient-temperature grinding), grain size, cross-linking, filler content and the type of natural rubber from which they were originally produced.

Gibala and Hamed (1994) presented the results of studies on compositions obtained from ground rubber wastes and styrene-butadiene rubbers (SBR). On the basis of those results, the Authors defined exactly the effect of rubber granulates on the rheological properties of rubber mixtures, their vulcanization process and the mechanical properties of the obtained composites. They showed that the addition of rubber granulate to the rubber compounds obtained from styrene-butadiene rubber (SBR) reduces the time of their subvulcanization and reduces the maximum vulcameter torque (ΔM). They also showed that the fall in value of the maximum torque measured during vulcametric analyses is due to the migration of sulfur from the elastomer matrix to the molecules of the rubber granulate. On the other hand, the reduction in subvulcanization time is due to the migration of vulcanization accelerators from the bulk of the rubber granulate to the fresh rubber phase. The process of migration of sulfur and accelerators in the rubber composite obtained from rubber granulate and SBR is presented in Fig. 2.

Fig. 2 Direction of migration of sulfur and accelerators in the rubber composites.

The analysis of the rheological properties of rubber compounds showed that compositions containing a rubber granulate grounded at ambient temperature have a higher

Mooney viscosity than those containing granulate obtained by cryogenic grinding of waste rubber (Gibala et al., 1996). It was explained by the occlusion of the elastomers from rubber compound by spongy particles of rubber granulates obtained at ambient temperature. In vulcanizate containing cryogenically grounded rubber granulates, in which grains have a smooth surfaces, occlusion was not found. This fact is reflected in the low viscosity of rubber compounds. Analysis of the mechanical properties of the compositions shown that the use of 30 phr of rubber granulate at ambient temperature reduces the tensile strength of vulcanizates from 28 MPa to 16.8 MPa and a reduction in ultimate elongation from 310% to 263% (Gibala et al., 1998). But it was also found that the presence of a rubber granulate in the fresh rubber distinctly improves its resistance to wear, (from 10.5 kN/m to 14 kN/m). This is partly due to the reduction in density of the cross-linked composition as a result of the migration of sulfur from the elastomer matrix to the granulate.

The mechanical properties of rubber composites (e.g., containing 30 parts by weight of granulates) can be improved by reducing their grain size or enlarging their specific surface area (Gibala et al., 1999). With granulate particle sizes obtained by grinding or manual cutting the improvement in the tensile strength of vulcanizates was 16.1-22.7 MPa (for grinded granulates) and 11.6-20.9 MPa (for cut granulates). Similar relationships were found for ultimate elongation: for grinded granulates values varied with decreasing particle size from 235 to 298%, and for cut granulates the corresponding values were 194-285%. The evident differences in the properties of composites containing two different types of rubber granulates are explained by the different nature of their interactions with the elastomer phase of the rubber compound. Microscopic research of rubber composites containing two types of rubber materials have shown that rubber granulates obtained by grinding have a larger specific surface area than the smooth-edged particles of grains obtained by cutting. As a result, they adhere better to the matrix, and during the stretching of composites they break, but without becoming separated from the natural rubber phase. On the other hand, the granulate obtained by cutting is very easily separated from the matrix.

Pyskło et al. (2000) also described the influence of the type and size of rubber granulate particles on the properties of composites obtained from SBR. They prepared composites with rubber dust obtained at the ambient temperature, rubber dust prepared during cryogenic grinding of rubber wastes and rubber pulp, obtained from the retreading of used tyres. Vulcanometric studies showed that the addition of 5% by weight of all types rubber granulate to rubber mixtures increases their viscosity and reduces their maximum vulcameter torque (ΔM), gradually diminishes their tensile strength, stress at constant elongation and hardness, and seriously lowers their resistance to multiple stretching. Similar relationships were also found with increasing grain size of the different rubber granulates. Ultimate elongation and tearing strength, slightly decrease as well. Similarly to the strength of composites obtained from rubber waste grinded at ambient temperature was much greater than those materials containing cryogenically obtained rubber dust. The Authors indicated that this is due to the different surface morphology of these two rubber materials. The irregular shape and the fuzzy surface of the particles of granulate obtained at ambient temperature is responsible for their better contact with the composite matrix. Cryogenic dust cause greater deterioration in the mechanical properties of vulcanizates than ambient temperature granulate even though the cryogenic dusts may have the same or smaller grain sizes, because cryogenic dusts have completely smooth surfaces. In the case of rubber pulp it was found that the properties of vulcanizates containing this kind of granulate are superior to those materials obtained from cryogenic dusts with a similar grain size range, and comparable to those materials made from coarse-grained cryogenic dusts with much larger grain sizes.

Kim et al (2003, 2007) showed that the mechanical properties and the course of vulcanization of rubber mixtures obtained from natural rubber and rubber granulates depends on the type of cross-linking agent used during their vulcanization. The reported that the use of 50 parts by weight of rubber granulate, obtained from cross-linked rubber using a conventional system consisting of sulfur and accelerators enables the subvulcanization time and the optimal time to vulcanize a rubber mixture to be shorten by ca 60% and 30%, respectively. In contrast, a rubber granulate obtained from rubber cross-linked with dicumyl peroxide practically does not affect the vulcanization parameters of the composition; only the maximum vulcameter torque is reduced. This provided evidence for the migration of sulfur from the rubber mixtures to interior of rubber granulate particles and to the vulcanization accelerators in the opposite direction. It also was reported that composites containing granulates cross-linked with sulfur have a far greater tensile strength, ultimate elongation and resistance to tear than materials obtained from granulates cross-linked with peroxide. In all composites analyzed, the increase in rubber granulate content decreased their mechanical properties, with the exception of the ultimate elongation which, for granulate contents of 10-50 phr, increased from ca 320-360% to 360-400%. The Authors explained that fact that the main reason for the deterioration in the mechanical properties of the vulcanizates is the reduction in matrix cross-linking density, which falls as the amount of granulate in the vulcanizates increases. The far inferior mechanical properties of composites containing peroxide cross-linked granulate were explained by their very weak adhesion to the matrix.

A similar improvement in mechanical properties due to granulate grains size in the mechanical properties of composites obtained from natural rubber and rubber granulate obtained from the cryogenic grinding of used tyres is reported in Naskar et al. (2000). The interesting aspect of these results is finding that the increased content of copper, iron, nickel, manganese and chromium, and the reduced levels of zinc and sulfur with decreasing granulate grain size. In the opinion of the Authors, the presence of large quantities of metals in fine-grained rubber dust is due to the difficulties of purifying it of the metallic residues left over after the grinding of the steel bead wires. The presence of these metals in the rubber material reduces the resistance to ageing of the vulcanizates after heating at 100°C for 36 h. In the case of composites containing granulate with the finest grains (0.1-0.15 mm) and at the same time the highest contents of the above-mentioned metals, because of ageing, their tensile strength was reduced by 69%, ultimate elongation by 53% and resistance to tear by 54%. In contrast, for a granulate of grain size 0.45-0.65 mm, the deterioration in the above properties was only 29%, 47% and 22%, respectively.

Han and Han (2002) demonstrated that the resistance to multiple bending (De Mattia test) for composites obtained from rubber granulate and natural rubber or rubber granulate and SBR increases up to 3-times. It is observed for the composites in case the rubber granulate content is in the range of 10 to 50 phr. It should be added that the improvement in resistance to multiple bending of composites is more distinct in materials obtained with the use of SBR and rubber granulates. In addition, the use of SBR as the composite matrix yields materials with a higher resistance to abrasion in comparison to the composite based on natural rubber matrix. Moreover, the changes in the content of rubber granulate and their grain size (in a range size 75-600 μm) do not significantly affect the resistance to abrasion of obtained composites.

In Table 2 there are summarized the results of literature analysis, which indicate that a significant influence on the improving of mechanical properties of the composite materials obtained by blending rubber granulate with rubber compounds have also methods of their preparing, type of used polymer matrix, particle size of rubber granulate, components ratio in the composite and the presence of additives. Based on the results shown in Table 2 following conclusions were formulated:

- Increase in the amount and particle size of rubber granulate reduce the mechanical properties of composites (Chandran et al., 2015).

- Good properties of polymer-rubber composites may be achieved by oxidation (Yehia et al., 2003) or plasma treatment (Zhang et al., 2009) of rubber granulate surface.
- Mechanical properties of composites may be improved by replacing part of rubber granulate by carbon black (Dekic et al., 2012), which enhances interaction between matrix and filler (Ismail et al.,2010).

Table 2. The influence of components type and ratio, particle size of rubber granulate, and processing on the properties of polymer-rubber composites obtained from rubber granulate and natural or synthetic rubber.

2.2. Elastic composites obtained in the process of sintering rubber granulate

A very interesting materials consisting of only ground rubber wastes were obtained by the process of sintering rubber granulates at elevated temperature and pressure (Fig. 3) (Farris and Morin, 2001; Morin et al., 2002). In this way very high amount of wastes are recycled and any other fresh material is not required to produce such materials. It is a very attractive group of rubber materials as they are environmentally friendlier and more economic than the composites obtained from ground rubber wastes and a new portion of fresh rubber component. The process by which such materials are obtained is based on the press molding of granulate at high pressure (0.5-26.0 MPa), which causes the consolidation and adhesion of grains and improves their adherence. At high temperature (80-240°C) the cross-links in the rubber granulate structure are broken up and the main chains are also partially disrupted. In the next stage, as a result of the recombination and regrouping of the radicals produced, new bonds are formed between the individual grains of the granulate, which yields a homogenous rubber material. The practical applications of such materials include the manufacture of washers, roofing materials, insulation boards, shoe soles and solid tyres.

Fig. 3 Process of sintering of rubber granulate.

The mechanical properties of sintered rubbers depend to a large extent on the temperature and time of the process, as well as on the type of rubber granulate and its grain size. Makarov and Drozdovski (1991) demonstrated that it is possible to obtain sintered rubbers with properties comparable to those composites produced from natural rubber and 70 wt% rubber granulates. Farris and Morin (2001) showed that the materials obtained in sintering process of rubber granulates, from natural rubber (NR) could achieve a tensile strength in the range of 3.5-6.5 MPa and an ultimate elongation of ca 330-530%. Improved properties of that material were obtained when the granulate was molded for 20-30 minutes at the temperature of 200°C and pressure equal to 8.6 MPa. These properties were much improved in sinters obtained from granulates containing SBR rubber, which have a tensile strength of ca 9.6-15.0 MPa and an ultimate elongation of 325-350%. It is interesting that the properties of sintered rubber obtained from polysulfide rubber granulate and vulcanizates obtained from the initial raw materials are comparable. The different properties of sintered granulates made from SBR and NR are due to the different mechanisms by which the bonds between the granulate grains are formed. In rubber materials containing NR granulates, the rubber particles are joined as a result of the recombination of radicals formed during bond breaking, whereas in SBR granulate cross-linking takes place through the mediation of unsaturated bonds present in SBR. The addition of sulfur to SBR granulate accelerates formation of composite and improves the tensile strength of obtained sinters. But if sulfur is present in amounts exceeding 10 parts by weight, obtained materials are hard and brittle, with an ultimate elongation of ca 5-6%. The mechanical properties of sinters obtained from NR granulate was improved by following the addition of low-molecular weight organic acids such as benzoic acid, salicylic acid, maleic acid and anhydride, phthalimide and phthalic anhydride (Tripathy et al., 2002).

Crespo et al. (2009) described the mechanical properties of natural crumb rubber (NCR), which was sintered at 160°C-220°C under the pressure between 4000-8000 kg. It was observed that elongation at break and the tensile strength of these sinters increase respectively from 100 % to 270 % and from 0.8 MPa to 1.3 MPa) when the molding temperature increase up to 200°C and pressure up to 6000 kg. The oxidation of the sulfur (from rubber) at temperature above 200 °C may be the explanation for decreases the tensile strength. The decrease elastic modulus (from 0.26 MPa to 0.15 MPa) in sintered NCR with increase the moulding temperature is caused by the thermal destruction of the polysulfide bonds (–Sx-) in rubber, and the formation of thermally activated free sulfur. That activated sulfur are diffused in the NCR materials and exhibits a high activity to conjugated dienes, present in the NCR waste material. With elastic modulus decrease, material becomes less rigid which leads to a decrease its hardness (from 41 °ShA to 21 °ShA). Crespo et al. (2009) found that the optimum parameters of sintering, which allows to obtain rubber sinters with the best mechanical properties are temperature of 200°C and compression pressure 6000kg.

In the same work the Authors have investigated mechanical properties of sintered products obtain from of NCR by using two kinds of adhesives, added to granulate in an amount to 50 wt%. The adhesives used in this study were water solution of natural rubber and polychloroprene latex with the maximum working temperature of up to 180°C. The sintering process were carried out at 160°C-180°C and pressure 6000kg. Sinters have the best tensile strength and elongation at break when they were obtained at 180°C with addition 5 wt% of latex (1.95 MPa, 310 % respectively), while the highest value of hardness (about 39 °ShA) have materials containing 5 wt% of adhesives which were obtained at 160 °C. In this case, addition of latex to rubber granulate in amount 5 wt% allows to achieve optimum properties of sintered materials. The latex addition above 10 wt% cause decreasing of mechanical properties distinctly. In composites obtained with addition of polychloroprene latex their tensile strength increases very fast, when the amount of adhesive are changed from 5 wt% to 10wt % (from 1.9 MPa to 3.1 MPa at 160 °C). The same situation can be observed in the case of value of elongation at break which increases from 240 % to 320 % in sinters obtained at temperature 160 °C. On the other hand the elastic modulus increases with amount of polychloroprene between 5 – 10 wt% independently from the applied sintering temperature. It was also observed that hardness in obtained materials increases nearly linearly from 41 °ShA to 51 °ShA when sinters was moulded at 160 °C and from 37 °ShA to 45 °ShA in materials sintered at 180°C.

The papers of García et al. (2007) and Stefani et al. (2005) showed that rice husk may be potentially filler which can be added to sintered rubber. It is low cost additive and quite good properties modifier. Adding of that additive to sintered rubber is leading to lower density of the material and enhanced biodegradability and recyclability what is also important in the case of ‘environmentally friendly’ production.

García et al. (2007) considered composites with three different particle sizes of rice husk filler (0.75 mm, 0.37 mm and less than 0.10 mm) to show that particle size is important parameter of mechanical properties in the sintering process. Samples were performed in a hot plate press with pressure and temperature control. The sintering was carried out at a constant pressure of 2 MPa at 180 °C. The heating cycle was selected by taking into account the relative thermal stability of components. Increase amount of filler in rubber sinters enhances the elastic modulus (from 3.42 MPa to about 12.08 MPa), decreases the tensile strength (from 2.16 MPa to 0.59 MPa) and the strain at break (from 105 % to 12 %) , whatever the particle sized was used. For composites obtained from the largest size of rice husk particles segregation during mixing was observed. For amount of filler 25 wt% the elastic modulus, the tensile strength and the strain at break were 6.48 MPa, 0.59 MPa and 12 % respectively. For composites containing granulates with the particle size about 0.1 mm the effectiveness of sintering process decrease. For amount of filler 25 % by weight the elastic modulus, the tensile strength and the strain at break were 10.59 MPa, 0.78 MPa and 15 % respectively. The Authors reported that the highest parameters of mechanical properties were obtained for rice husk size about in the same range of that the matrix (about 0,30 mm). They received the elastic modulus from 3.97 MPa to 12.08 MPa, the tensile strength from 1.85 MPa to 0.96 MPa and the strain at break from 105 % to 23 % for amount of filler 5-25 wt%.

2.3. Composites obtained from thermoplastic materials and rubber granulates

One of the ways of recycling ground rubber wastes is to use them as a component of composites in which thermoplastics are present; principally polyethylene (PE), polypropylene (PP), polystyrene (PS) and polyvinyl chloride (PVC) (Pittolo and Burford 1985; Duhaime and Baker, 1991; Tipanna and Kale, 1997; Phinyocheep et al., 2002; Naskar et al., 2002). These materials play an important role in the management of end-of-life tyres as they are materials low costs, easy for processing especially by injection and extrusion methods. They are nowadays very interesting composites in the polymer material market (Mangaraj, 2005).

The mechanical properties of this type of composites depend largely on the type of used thermoplastic matrix, the amount of rubber granulate and the degree of its dispersion in matrix as well as the nature of the interactions occurring between the composite components (Rajeev and De, 2004; Rajalingam et al., 1993; Pramanik and Baker, 1995).

Ismail and Suryadiansyah (2002) and Ismail et al. (2006) did show that rubber granulate in PP composites slightly improves their ultimate elongation in case a very fine grains' size in a range of 0,25-0,50 mm were used. In another papers (Tuchman and Rosen, 1978; Choudhury and Bhattacharya, 1996) it was also found that the physical modification of PP with a rubber granulate increases its impact strength, which may be greater by as much as 100% following the addition of 20% by mass of granulate.

It was reported in the literature that rubber granulates act as inert fillers. Very often it causes deterioration in the strength of the end products which strongly depends on the amount of rubber granulate and the type of matrix (Fuhrmann and Karger-Kocsis, 1999; Kim et al., 2000; Scaffaro et al., 2005; Egodage et al., 2009).

It was found that phase interactions between ground rubber wastes and thermoplastics are weak as they come from completely different group of macromolecular compounds with different physicochemical properties. The interactions can be improved, by adding various kinds of compatibilizers causing the reduction of interphase energy and increase of adhesion between the rubber granulate and the thermoplastic components. Moreover a better dispersion of the granulate grains in the matrix and less agglomerates after processing are observed in the final composites.

Lee and Roh (2007) presented the results of using compatibilizers like block styrene-ethylene/butylene-styrene (SEBS) copolymer grafted with maleic anhydride (SEBS-MA) and polypropylene grafted with maleic anhydride (PP-MA). Both types of compatibilizers substantially improved the mechanical properties of the proper composites in which granulates from wet grinding of tyres and isotactic polypropylene (i-PP) were used. What is more, multiple processing of those composites with compatibilizers has no effect on their mechanical properties.

Rajalingam et al. (1993) investigated the effect of an ethylene/acrylic acid copolymer on a mixture of rubber granulate and linear low-density polyethylene (LLDPE). They also examined the effect of the nature of the rubber granulate grain surface on the action of the compatibilizer. They found that using a rubber granulate grinded at ambient temperature with a more porous and rougher surface yielded composites with a somewhat better impact strength than those made with a smooth-grained cryogenic recyclate. Moreover, the processability of a recyclate grinded at ambient temperature with LLDPE is better.

Choudhury and Bhattacharya (1996) obtained an improvement in the properties of the composite made from rubber granulate and low-density polyethylene (LDPE) as a result of using a chlorinated polyethylene (CPE) as a compatibilizer. The Authors justified the use of CPE because of its affinity for both the rubber waste component and PE. This was explained by its elastic nature and structure of the main chain, which is almost identical to that in LDPE. Moreover, the flow index was clearly reduced with CPE as a result of the better phase interactions between LDPE and the rubber granulate. The similar findings were observed by Li et al. (2003), who developed composites with superior mechanical properties using an ethylene-propylene diene terpolymer (EPDM) for compatibilization of composite made from rubber waste granulate and high-density polyethylene (HDPE).

Better adhesion and mechanical properties of composites, especially the ultimate elongation, can also be achieved by adding to the system two different compatibilizers, namely, epoxidized natural rubber (ENR) and LLDPE grafted with maleic anhydride, methyl methacrylate and butyl acrylate (Guo et al., 2004).

A method of improving the compatibility between PP and a rubber waste granulate with the aid of natural rubber latex was presented by Awang et al. (2008). It is based on coating the granulate grains with a mixture of latex and sulfur. As a result, the tensile strength, ultimate elongation and elastic modulus of composites were substantially improved, as was their resistance to swelling in toluene and oil. Microscopic examination also showed that the latex coating gave a better grain dispersion in the matrix.

Oliphant and Baker (1993) discussed the possible improvement of the mechanical properties of LLDPE-based composites by the modification of the surface of the rubber waste granulate with an ethylene and acrylic acid copolymer (E/AA). The Authors showed that the improved tensile and impact strengths of the composites is due to the strong interactions and reactions of the functional groups at the surface of the granulate grains with the carboxyl groups of the compatibilizer.

Rajalingam and Baker (1992) showed quite big improvement in composite strength by modifying the rubber granulate surface with E/AA (40-50% w/w of modified granulate). The improvement of the properties like impact strength was achieved only in the case the matrix of the composite was LDPE but not HDPE.

The compatibility of thermoplastics with rubber waste granulates can also be improved by modifying their grain surfaces with sulfuric, nitric and perchloric acids (Colom et al., 2007), and with trichloroisocyanuric acid (Naskar et al., 2002). Research is also focused on the modification of granulates by physical methods such as UV radiation (Shanmugharaj et al., 2005) and γ radiation (Sonnier et al., 2007). However Sonnier et al., 2008 found that that reducing the grain size of rubber granulates, by expanding or modifying their specific surface area and using various types of compatibilizers, is not often satisfactory for improving mechanical strength .

An interesting approach of obtaining new composites with better mechanical properties can be achieved from rubber wastes granulates and thermoplastics processed in the way to give the features of thermoplastic vulcanizates (TPV), i.e., NR-thermoplastic composites, which are produced during the dynamic vulcanization of the NR phase with the molten thermoplastic (Coran and Patel, 1980a; 1980b; Coran and Patel, 1983; Karger-Kocsis, 1995; Karger-Kocsis, 1999; Radusch and Rzymski, 2001).

The papers of Menning et al. (1997a; 1997b) demonstrated that the additional cross-linking of PP-rubber granulate mixtures using sulfur and peroxides causes PP molecules to be grafted onto the surface of the rubber granulate grains, which are then both cross-linked (Hannes and Scholz 1996; Mennig et al., 1997a, 1997b; Michael et al., 1999). This gives better interphase adhesion and improves the mechanical properties of the thermoplastic vulcanizates obtained (TPV-granulate), which are much stronger than a physical mixture. The TPV-granulate compositions developed in this way (with grain sizes <0.4 mm) have a tensile strength of 14-17 MPa and a relative elongation of >150%, whereas a physical mixture of PP and rubber granulate of exactly the same mass ratio (50/50) has a tensile strength of 13 MPa and an elongation of 70% (Michael et al., 1999).

Naskar et al.(2001) observed that rubber waste granulate can serve as a partial replacement for EPDM terpolymer in thermoplastic vulcanizates (TPV) obtained from a mixture of EPDM and acrylated HDPE (AHDPE), subjected to dynamic vulcanization using dicumyl peroxide (DCP). The Authors found that their TPVs had optimal mechanical properties at a rubber to thermoplastic phase ratio of 60:40 and 1 part by weight of DCP. They also found that in the TPVs up to 50% of the EPDM could be replaced with waste rubber granulate without impairing their properties or processability.

The thermoplastic elastomers obtained from rubber granulates of ground end-of-life tyres types or other rubber wastes were used to produce materials like Symar T, from which automobile radiator linings and roofing materials were manufactured. The composition of such materials was patented by Jury and Chien (2001). They obtained thermoplastic materials from rubber dust (grain size 0.15-0.8 mm), PE, PP, EVA copolymer, SBS copolymer and auxiliary substances. The composites they developed, consisting of ca 50 wt% rubber granulate, depending on the proportions of constituents had tensile strength 5.0-7.2 MPa, relative elongation 200-260%, tearing resistance 32-52 kN/m, hardness 77-89 and compression set 60-75%. An advantage of these compositions is that they can be processed by injection or extrusion. In accordance with the inventors intention, they offer an alternative material to rubber used to manufacture products for the automobile industry.

The papers of Kowalska et al. showed that thermoplastic compositions containing ground rubber wastes (50% w/w) can also be used to manufacture many other articles of practical use like porous hoses for gardening purposes or as a sealant in anti-noise screens . The matrix of the composite materials can be PE, PP or PVC. and rubber granulate (50% by weight) can be used to manufacture (Kowalska et al., 1994; Kowalska et al., 1995; Kowalska and Wielgosz, 1999; Kowalska, 2002a; Kowalska et al., 2003). Parts of porous hoses obtained from LDPE, rubber granulate and phosphogypsum waste can be used as a substrate in sewage treatment plants (Kowalska et al., 1999; Kowalska et al., 2002b). PE and PVC wastes (from cable housings) in combination with rubber waste granulate are possible to be used to produce compositions for manufacturing street furniture (Kowalska et al., 1997; Kowalska and Wielgosz, 1999; Kowalska et al., 2002b; Kowalska, 2002c). PVC composites plasticized with dioctyl phthalate (DOP) and containing rubber granulate as filler, can be used as rubber linings (Kowalska, 2002a; Kowalska et al., 2002b).

Many others works are also related to the processing and methods of improving properties of polymer-rubber composite obtained from thermoplastics and rubber granulate. In Table 3 there are presented the most important achievements in this field. Based on this results it can be concluded that:

- The advantage of producing composites is the possibility of processing by injection molding or extrusion.
- Composites obtained from thermoplastics and rubber granulate are characterized by the low adhesion between thermoplastic matrix and filler (Crespo et al., 2010).
- Mechanical properties decrease when the amount of rubber granulate increases (Orrit et al., 2011).
- The properties of composites can be improved with thermoplastic matrix, by using of maleic anhydride (MA) and dicumyl peroxide (DCP) (Formela et al., 2013) or or bis-maleimide (BMI) (Magioli et al., 2010), which acts as compatybilizing system. In

turn, tensile strength can increase by the use of ethylene-1-ocetne copolymers (EOC) (Rocha et al., 2014).

- Increasing of rubber granulate content and addition of stabilizers improves stability against weathering (light, temperature, humidity) for composites obtained from maleate polyethylene and rubber waste granulate.

Table 3. The influence of type of components, particle size of rubber granulate, components ratio and processing on the properties of polymer-rubber composites obtained from rubber granulate and thermoplastics.

3. Conclusions

In this paper we have shown that the environmental problem of increasing number of used tyres in the world has been solved by the proper management and restrictive legal regulations, which prohibit stockpiling of postconsumer tyres on the landfill. It was the background for the development of new technologies for the production of new polymer-rubber composites obtained from rubber waste granulates. Such approach changed the role of rubber waste granulates. They should not be longer considered as a pollutant or as a cheap filler but should become 'environmentally friendly' materials with utilitarian properties. Regarding to the literature review showing the possibilities of using tyre granulates obtained from waste tyres to produce new polymer-rubber composites, it can be concluded that:

- Rubber waste granulates can be reused to produce various kinds of new rubber-polymer composites with the matrix of elastomers or thermoplastics.
- Rubber sinters are attractive group of materials due to their preparation only from rubber wastes what means sustainable and clean recycling.
- Production method of polymer composites obtained from recycled tyres is very simple. They can be manufactured using already known technology, machinery and equipment used in the rubber and thermoplastic materials industries. Therefore, implementation of these composites into industrial production does not require as much investment.
- Mechanical properties and performance of polymer composites obtained from waste rubber granulates depend on the type of used polymer matrix and therefore the nature of interactions between the matrix, the type and size of rubber grains and their amount in the composites.

References

Awang, M., Ismail, H., Hazizan, M.A., 2008. Processing and properties of polypropylene-latex modified waste tyre dust blends (PP/WTD_{ML}). Polym. Test. 27, 93–99.

Bravo, M., de Brito, J., 2012. Concrete made with used tyre aggregate: durability-related performance. Journal of Cleaner Production 25, 42-50.

Chandran, V., Raj, M.T., Lakshmanan, T., Kumar, S.M., 2015. Evaluation of Performance of Natural Rubber Composites with Different Sizes of Waste Tyre Rubber (WTR) and Precipitated Silica on C–M–M. Arab. J. Sci. Eng. 40, 1187-1196.

Cheng, X., Chen, H., Huang, S., Li, Z., Guo, X., 2012. Improvement of the Properties of Plasma Modified Ground Tire Rubber-Filled Cement Paste. J. Appl. Polym. Sci. 126, 1837–1843.

Choudhury, N.R., Bhattacharya, S.N., 1996. Effect of compatibilisation on mechanical and rheological properties of ground rubber tyre-polyethylene blends. Plast. Rub. Compos. Process. Appl. 25, 448 – 457.

Colom, X., Carrillo, F., Canavate, J., 2007. Composites reinforced with reused tyres: Surface oxidant treatment to improve the interfacial compatibility. Comp. Part A: Appl. Sci. Manuf. 38, 44–50.

Coran, A.Y., Patel, R., 1980a. Rubber-Thermoplastic Compositions. Part I. EPDM-Polypropylene Thermoplastic Vulcanizates. Rub. Chem. Technol. 53, 141-150.

Coran, A.Y., Patel, R., 1980b. Rubber-Thermoplastic Compositions. Part II. NBR-Nylon Thermoplastic Elastomeric Compositions. Rub Chem. Technol. 53, 781-794.

Coran, A.Y. Patel, R., 1983. Rubber-Thermoplastic Compositions. Part VIII. Nitrile Rubber Polyolefin Blends with Technological Compatibilization. Rub. Chem. Technol. 56, 1045-1060.

Crespo, J.E., Parres, F., Nadal, A., 2009. Mechanical behaviour analysis of sintered products of natural rubber crumb rubber (NRCR) using adhesives. Mater.-wiss. Werkst. 40, 211-217.

Crespo, J.E., Parres, F., Nadal, A., 2010. Research into the influence of ground tire rubber (GTR) in the mechanical and thermal properties of recycled thermoplastic materials. Mater.-wiss. Werkst. 41, 293-299.

CRIA - China Rubber Industry Association, 2010. Overview of Recycling and Reuse of Waste Tyres in China. http://www.etrma.org/uploads/Eventsmanager/CRIA%20%20PPT%20Report%205.1%20English%20version.pdf/ (accessed 31.01.2016).

Dekic, P.S., Temeljkovski, D.I., Rancic, B., Nusev, S., 2012. Application of recycled rubber powder (RRP) in NR/SBR compunds. J. Sci. Ind. Res. 71, 295-298.

Duhaime, J.R.M., Baker, W.E., 1991. Reactive blending of polyethylene and scrap rubber. Plast. Rub. Comp. Process. Appl. 15, 87–93.

Egodage, S.M., Harper, J.F., Walpalage, S.J., 2009. The development of rubber-thermoplastic blends from ground tyre rubber and waste polypropylene. Natl. Sci. Found. of Sri Lanka 37, 117-123.

ETRMA - European Tyre & Rubber Manufacturers Association, 2010. UT/Part Worn Tyres/ELT's Europe - Volumes Situation 2010. http://www.etrma.org/uploads/Modules/Documentsmanager/2011-07-11_ut-recovery-2010---table-for-press-release_final.pdf/ (accessed 30.05.2011).

ETRMA – European Tyre & Rubber Manufacturers' Association, 2011. End of life tyres - A valuable resource with growing potential – 2011 Report. http://www.etrma.org/uploads/Modules/Documentsmanager/brochure-elt-2011-final.pdf/ (accessed 31.01.2016).

ETRMA – European Tyre Rubber Manufactures Association, 2013. European tyre and rubber industry – Statistics. http://www.etrma.org/uploads/Modules/Documentsmanager/20131015---statistics-booklet-2013-final2.pdf/ (accessed 31.01.2016).

ETRMA – European Tyre Rubber Manufactures Association, 2014. European tyre and rubber industry – Statistics. https://www.mrw.co.uk/download?ac=1434852/ (accessed 31.01.2016).

Farris, R.J., Morin, J.E., 2001. Thermoset recycling methods. Patent US 2002/0101004/A1.

Formela, K., Haponiuk, J.T, Stankiewicz, P., Ryl, J., Konska, K., 2013. Effect of slightly crosslinked butyl rubber and compatibilizer additives on properties of recycled polyethylene-ground tire rubber blends. Przem. Chem. 92, 444-449.

Fuhrmann, I., Karger-Kocsis, J., 1999. Promising approach to functionalisation of ground tyre rubber -photochemically induced grafting: Short Communication. Plast. Rub. Comp. 28, 500–504.

Garcia, D., López, J., Balart, R., Ruseckaite, R.A., Stefani, P.M., 2007. Composites based on sintering rice husk-waste tire rubber mixtures. Mater. Des. 28, 2234-2238.

Gibala, D., Hamed, G.R., 1994. Cure and mechanical behavior of rubber compounds containing ground vulcanizates. I: Cure behavior. Rub. Chem. Technol. 67, 636-648.

Gibala, D., Laohapisitpanich, K., Thomas, D., Hamed, G.R., 1996. Cure and Mechanical Behavior of Rubber Compounds Containing Ground Vulcanizates. Part II—Mooney Viscosity. Rub. Chem. Technol. 69, 115-119.

Gibala, D., Hamed, G.R., Zhao, J., 1998. Tensile Behavior of an SBR Vulcanizate Containing a Single Rubber Particle. Rub. Chem. Technol. 71, 861-865.

Gibala, D., Thomas, D., Hamed, G.R., 1999. Cure and Mechanical Behavior of Rubber Compounds Containing Ground Vulcanizates: Part III. Tensile and Tear Strength. Rub. Chem. Technol. 72, 357-360.

González, V, Martínez-Boza, F. J., Gallegos, C., Pérez-Lepe, A., Páez, A., 2012. A study into the processing of bitumen modified with tyre crumb rubber and polymeric additives. Fuel Process. Technol. 95, 137–143.

Goodyear, C., 1853. British Patent 2933.

Guo, B., Cao, Y., Jia, D., Qiu, Q., 2004. Thermoplastic Elastomers Derived from Scrap Rubber Powder/LLDPE Blend with LLDPE-graft-(Epoxidized Natural Rubber) Dual Compatibilizer. Macromol. Mater. Eng. 289, 360 – 367.

Han, S.C., Han, M.H., 2002. Fracture behavior of NR and SBR vulcanizates filled with ground rubber having uniform particle size. J. Appl. Polym. Sci. 85, 2491–2500.

Hannes, M., Scholz, H., 1996. Recycling of scrap rubber to produce polymer processing compound with TPE characteristics. Patent DE 19607281.

Hu. N., Cheng. C.M., Wen. X.F., Chen. Y., Hu H.L., 2014. Comparison Study of Scrap Tires Management between China and the USA. Adv. Mater. Res. 878, 90-98.

Huang, B., Shu, X., Cao, J., 2013. A two-staged surface treatment to improve properties of rubber modified cement composites. Constr. Build, Mater. 40, 270–274.

Ismail, H., Suryadiansyah, M., 2002. Thermoplastic elastomers based on polypropylene/natural rubber and polypropylene/recycle rubber blends. Polym. Test. 21 (4), 389–395.

Ismail, H., Awang, M., Hazizan, M. A., 2006. Effect of Waste Tire Dust (WTD) Size on the Mechanical and Morphological Properties of Polypropylene/Waste Tire Dust (PP/WTD) Blends. Polym. Plast. Technol. Eng. 45, 463–468.

Ismail, H., Omar, N.F., Othman, N., 2010. Effect of Carbon Black Loading on Curing Characteristics and Mechanical Properties of Waste Tyre Dust/Carbon Black Hybrid Filler Filled Natural Rubber Compounds. J. Appl. Polym. Sci. 121, 1143–1150.

JATMA - Japan Automobile Tyre Manufacturers Association, Inc., 2015. Tyre industry of Japan. http://www.jatma.or.jp/english/media/ (accessed January 2015).

Jury, J.R., Chien, A.W.V., 2001. Thermoplastic elastomer composition. Patent U.S. 6313183.

Kakroodi, A. R., Rodrigue, D., 2013a. Degradation behavior of maleated polyethylene/ground tire rubber thermoplastic elastomers with and without stabilizers. Polym. Degrad. Stab. 98, 2184-2192.

Kakroodi, A. R., Rodrigue, D., 2013b. Reinforcement of Maleated Polyethylene/Ground Tire Rubber. J. Appl. Polym. Sci. 131, 1-8.

Karger-Kocsis, J., 1995. Polypropylene structure, blends and composites. Copolymers and Blends Vol.2. Chapman & Hall, London.

Karger-Kocsis, J., 1999. Polypropylene: An A-Z Reference. Kluwer Publishers, Dordrecht.

Karger-Kocsis, J., Meszaros, L., Barany, T., 2013. Ground tyre rubber (GTR) in thermoplastics, thermosets, and rubbers. J.Mater. Sci. 48, 1-38.

Kim, J.I., Ryu, S.H., Chang, Y.W., 2000. Mechanical and dynamic mechanical properties of waste rubber powder/HDPE composite. J. Appl. Polym Sci. 77, 2595–2602.

Kim, S.W., Hong, K.H., Seo, K.H., 2003. Effects of ground rubber having different curing systems on the crosslink structures and physical properties of NR vulcanizates. Mater. Res. Innov. 7, 149–154.

Kim, S.W., Park, H.Y., Lim, J.C., Jeon, I.R., Seo, K.H., 2007. Cure characteristics and physical properties of ground-rubber-filled natural rubber vulcanizates: Effects of the curing systems of the ground rubber and rubber matrix. J. Appl. Polym. Sci. 105, 2396–2406.

Koçak, Y., Subaşı, S., Yaman, Y., Uğur, L.O., Ateş, A., 2012. 1st International Conference on Architecture & Urban Design 1235-1240.

Kocevski, S., Yagneswaran, S., Xiao, F., Punith, V.S., Smith Jr., D.W., Amirkhanian, S., 2012. Surface modified ground rubber tire by grafting acrylic acid for paving applications. Constr. Build. Mater. 34, 83–90.

Kowalska, E., Ostrowski, E., Borensztejn, M., Wielgosz, Z., 1994. Thermoplastic polymeric composition containing rubber wastes. Patent PL 177682.

Kowalska, E., Ostrowski, E., Borensztejn, M., Wielgosz, Z., 1995. Apparatus for making porous tubes of polymeric compositions containing rubber discards. Patent PL 175637.

Kowalska, E., Ostrowski, E., Borensztejn, M., Wielgosz, Z., Cyrkiewicz, M., Kleszczewski, J., 1997. Thermoplastic polymeric composition containing waste rubber. Patent PL 321612.

Kowalska, E., Wielgosz, Z., 1999. The effect of vulcanized rubber scrap and thermally modified phosphogypsum used as additives on thermoplastic compositions properties. Elast. 3, 20-27.

Kowalsk,a E., Borensztejn, M., Ostrowski, E., Wielgosz, Z., Rog, S., Kosinski, W., Grazewicz, J., 1999. Porous material for use in sewage and water treatment processes. Patent PL 332918.

Kowalska, E., 2002a. Secondary raw materials used as fillers in theroplastic polymer compositions. Przem. Chem. 81, 374-379.

Kowalska, E., Wielgosz, Z., Pelka, J., 2002b. Use of post-life waste and production waste in thermoplastic polymer compositions. Polym. Polym. Compos. 10, 83-92.

Kowalska, E., 2002c. New developments in recycling of thermoplastics wastes. Przem. Chem. 81, 171-176.

Kowalska, E., Żubrowska, M., Borensztejn, M., 2003. Heterophase thermoplastic polymer compositions modified with rubber wastes. Polim. 48, 633-640.

Lee, H.J., Roh, H.S., 2007. The use of recycled tire chips to minimize dynamic earth pressure during compaction of backfill. Constr. Build. Mater. 21, 1016–1026.

Li, Y., Zhang, Y., Zhang, Y., 2003. Mechanical properties of high-density polyethylene/scrap rubber powder composites modified with ethylene–propylene–diene terpolymer, dicumyl peroxide, and silicone oil. Journal of Appl. Polym. Sci. 88, 2020–2027.

Li, X., Xu, H., Gao, Y., Tao, Y., 2010. Comparison of end-of-life tire treatment technologies: A Chinese case study. Waste Manag. 30, 2235–2246.

Liang, M., Xin, X., Fan, W., Sun, H., Yao, Y., Xing, B., 2015. Viscous properties, storage stability and their relationships with microstructure of tire scrap rubber modified asphalt. Constr. Build. Mater. 74, 124–131.

Magioli, M., Sirqueira, A.S., Soares, B.G., 2010. The effect of dynamic vulcanization on the mechanical, dynamic mechanical and fatigue properties of TPV based on polypropylene and ground tire rubber. Polym. Test. 29. 840–848.

Makarov, V.M., Drozdovski V.F., 1991. Reprocessing of tyres and rubber wastes, Ellis Horwood, New York.

Mangaraj, D., 2005. Role of Compatibilization in Recycling Rubber Waste by Blending with Plastics. Rub. Chem. Technol. 78 (3), 536-547.

Martinez, J.D., Puy, N., Murillo, R., Garcia, T., Navarro, M.V., Mastral, A.M., 2013. Waste tyre pyrolysis – A review. Renew. Sustain.Energ. Rev. 23, 179–213.

Mennig, G., Michael, H., Scholz, H., Rzymski, W. 1997a. Proc. 7th International Conference on Machinery Recycling Bydgoszcz/Polen. 203-211.

Mennig, G., Michael, H., Rzymski, W., Scholz, H., 1997b. Thermoplastic elastomers from polypropylene-powdered rubber scrap blends. Polim. 42, 491-493.

Michael, H., Scholz, H., Mennig, G, 1999. Blends from Recycled Rubber and Thermoplastics. Kautsch. Gummi Kunstst. 52, 510-513.

Miro, R, Martinez, A.H., Navarro, F.M., Gamez, M.C.R., 2015. Effect of ageing an temperature on the fatigue behaviour of bitumens. Mater. Des. 86, 129–137.

Morin, J.E., Williams, D.E; Farris, R.J., 2002. A Novel Method to Recycle Scrap Tires: High-Pressure High-Temperature Sintering. Rub. Chem. Technol. 75, 955-968.

Naskar, A.K., De, S.K., Bhowmick, A.K., Pramanik, P.K., Mukhopadhyay, R., 2000. Characterization of Ground Rubber Tire and its Effect on Natural Rubber Compound. Rub. Chem. Technol. 73, 902-911.

Naskar, A.K., Bhowmick, A.K., De, S.K., 2001. Thermoplastic elastomeric composition based on ground rubber tire. Polym. Eng. Sci. 41, 1087–1098.

Naskar, A,K.,. Bhowmick, A, K, De, S. K., 2002. Melt-processable rubber: Chlorinated waste tire rubber-filled polyvinyl chloride. J. Appl. Polym. Sci. 84, 622–631.

Navarro, F.J., Partal, P., Martínez-Boza, F., Gallegos, C., 2004. Thermo-rheological behaviour and storage stability of ground tire rubber-modified bitumens. Fuel 83, 2041–2049.

Navarro, F.M., Sanchez, M.S., Gámez, M.C.R., Martínez, M.S., 2014. The use of additives for the improvement of the mechanical behavior of high modulus asphalt mixes. Constr. Build. Mater. 70, 65–70.

Nejad, M. F., Aghajani, P., Modarres, A., Firoozifar, H., 2012. Investigating the properties of crumb rubber modified bitumen using classic and SHRP testing methods. Constr. Build. Mater. 26, 481–489.

Oliphant, K., Baker, W.E., 1993. The use of cryogenically ground rubber tires as a filler in polyolefin blends. Polym. Eng. Sci. 33, 166–174.

Ossola, G., Wojcik, A., 2014. UV modification of tire rubber for use in cementitious composites. Cem. Concr. Compos. 52, 34–41.

Orrit, P.J., Mujal-Rosas, M.R., Rahhali, A., Genesca, M.M., Fajula, C.X., Punseti, B.J., 2011. Dielectric and mechanical characterization of PVC composites with ground tire rubber. J. Compos. Mater. 45, 1233-1243.

Phinyocheep, P., Axtell, F.H., Laosee, T., 2002. Influence of compatibilizers on mechanical properties, crystallization, and morphology of polypropylene/scrap rubber dust blends. J Appl. Polym. Sci. 86, 148–159.

Pittolo, M., Burford, R.P., 1985. Recycled Rubber Crumb as a Toughener of Polystyrene. Rub. Chem. Technol. 58, 97-105.

Polacco, G., Filippi, S., Merusi, F., Stastna, G., 2015. A review of the fundamentals of polymer-modified asphalts: Asphalt/polymer interactions and principles of compatibility. Adv. Coll. Interface Sci. 224, 72–112.

Pramanik, P.K., Baker, W.E., 1995. Toughening of ground rubber tire filled thermoplastic compounds using different compatibilizer systems. Plast. Rub. Compos. Process.Appl. 24 (4), 229–237.

Presti, D., 2013. Recycled Tyre Rubber Modified Bitumens for road asphalt mixtures: A literature review. Constr. Build. Mater. 49, 863–881.

Pyskło, L., Parasiewicz, W., Stępkowski, R., 2000. Characteristics of rubber powders manufactured by means of different methods and evaluation their influence on rubber compounds properties. Elastom. 5, 15-25.

Radusch, H.J., Rzymski, W.M, 2001. Thermoplastic vulcanizates. I. Essence as well as material and structural-morphological requirements of their properties. Elastom. 5, 19-28.

Rajalingam, P., Baker, W.E., 1992. The Role of Functional Polymers in Ground Rubber Tire-Polyethylene Composite. Rub. Chem. Technol. 65, 908-916.

Rajalingam, P., Sharpe, J., Baker, W. E., 1993. Ground Rubber Tire / Thermoplastic Composites: Effect of Different Ground Rubber Tires. Rub. Chem. Technol. 66, 664-677.

Rajeev, R.S., De, S.K., 2004. Thermoplastic Elastomers Based on Waste Rubber and Plastics. Rub. Chem. Technol. 77, 569-578.

Ramarad, S., Khalid, M., Ratnam, C.T., Luqman Chuah, A., Rashmi, W., 2015. Waste tire rubber in polymer blends: A review on the evolution, properties and future. Prog. Mater. Sci. 72, 100–140.

Ramli, S., Ratnam, C.T., Ahamd, S.H., 2013. Mechanical, thermal, and morphological properties of ethylene vinyl acetate/waste tire dust (EVA/WDT) blends. Malays. Polym. J. 8, 27-32.

Rocha, G.M.C., Leyva, M.E., Oliveira, M.G., 2014. Thermoplastic Elastomers Blends Based on Linear Low Density Polyethylene, Ethylene-1-Octene Copolymers and Ground Rubber Tire. Polim. 24, 23-29.

RMA - Rubber anufacturers Association (USA), 2009. Scrap tire markets in the United States 9th Biennial Report. http://www.rma.org/scrap_tires/ (accessed January 2015).

RMA - Rubber Manufacturers Association (US), 2013. 2013 U.S. Scrap Tire Management Summary. http://www.rma.org/publications/scrap-tire-publications/ (accessed January 2015).

Scaffaro, R., Dintcheva, N.T, Nocilla, M.A., La Mantia, F.P., 2005. Formulation, characterization and optimization of the processing condition of blends of recycled polyethylene and ground tyre rubber: Mechanical and rheological analysis. Polym. Degrad. Stab. 90, 281-287.

Shanmugharaj, A.M., Kim, J.K., Ryu S.H., 2005. UV surface modification of waste tire powder: Characterization and its influence on the properties of polypropylene/waste powder composites. Polym. Test. 24, 739–745.

Shu, X., Huang, B., 2013. Recycling of waste tire rubber in asphalt and portland cement concrete: An overview. Constr. Build. Mater. 67, 217-224.

Schnubel, M., 2014. Today's Technician: Automotive Suspension & Steering System, Tires and Wheels. Cengage Learning, Stamford, pp 49-94.

Sienkiewicz, M., Kucińska-Lipka, J., Janik, H., Balas, A., 2012. Progress in used tyres management in the European Union: A review. Waste Manag. 32, 1742-1751.

Sonnier, R., Leroy, E., Clerc, L., Bergeret, A., Lopez-Cuesta, J.M., 2007. Polyethylene/ground tyre rubber blends: Influence of particle morphology and oxidation on mechanical properties. Polym. Test. 26, 274–281.

Sonnier, R., Leroy, E., Clerc, L., Bergeret, A., Lopez-Cuesta, J.-M., Bretelle, A.-S., Lenny, P., 2008. Compatibilizing thermoplastic/ground tyre rubber powder blends: Efficiency and limits. Polym. Test. 27, 901–907.

Stefani, P. M., Garcia, D., Lopez, J., Jimenez, A., 2005. Thermogravimetric analysis of composites obtained from sintering of rice husk-scrap tire mixtures. J. Therm. Anal. Calorim. 81, 315-320.

Strong, B.A., 2007. Fundamentals of Composites Manufacturing: Materials, Methods and Applications. Society of Manufacturing Engineers, USA.

Thomas, B.S., Gupta, R.C., 2016. A comprehensive review on the applications of waste tire rubber in cement concrete. Renew. Sustain. Energ. Rev. 54, 1323–1333.

Tipanna, M., Kale, D.D., 1997. Composites of Waste, Ground Rubber Particles and Poly(Vinyl Chloride). Rub. Chem. Technol. 70, 815-819.

Tripathy, A.R., Morin, J.E., Williams, D.E., Eyles, S.J., Farris, R.J., 2002. A Novel Approach to Improving the Mechanical Properties in Recycled Vulcanized Natural Rubber and Its Mechanism. Macromol. 35, 4616-4627.

Torretta, V., Rada, E.C., Ragazzi, M., Trulli, E., Istrate, I.A, Cioca, L.I., 2015. Treatment and disposal of tyres: Two EU approaches. A review. Waste Manag. 45, 152-160.

Tuchman, D., Rosen, S.L., 1978. The mechanical properties of plastics containing cryogenically ground tyre. J. Elastom. Plast. 10, 115-128.

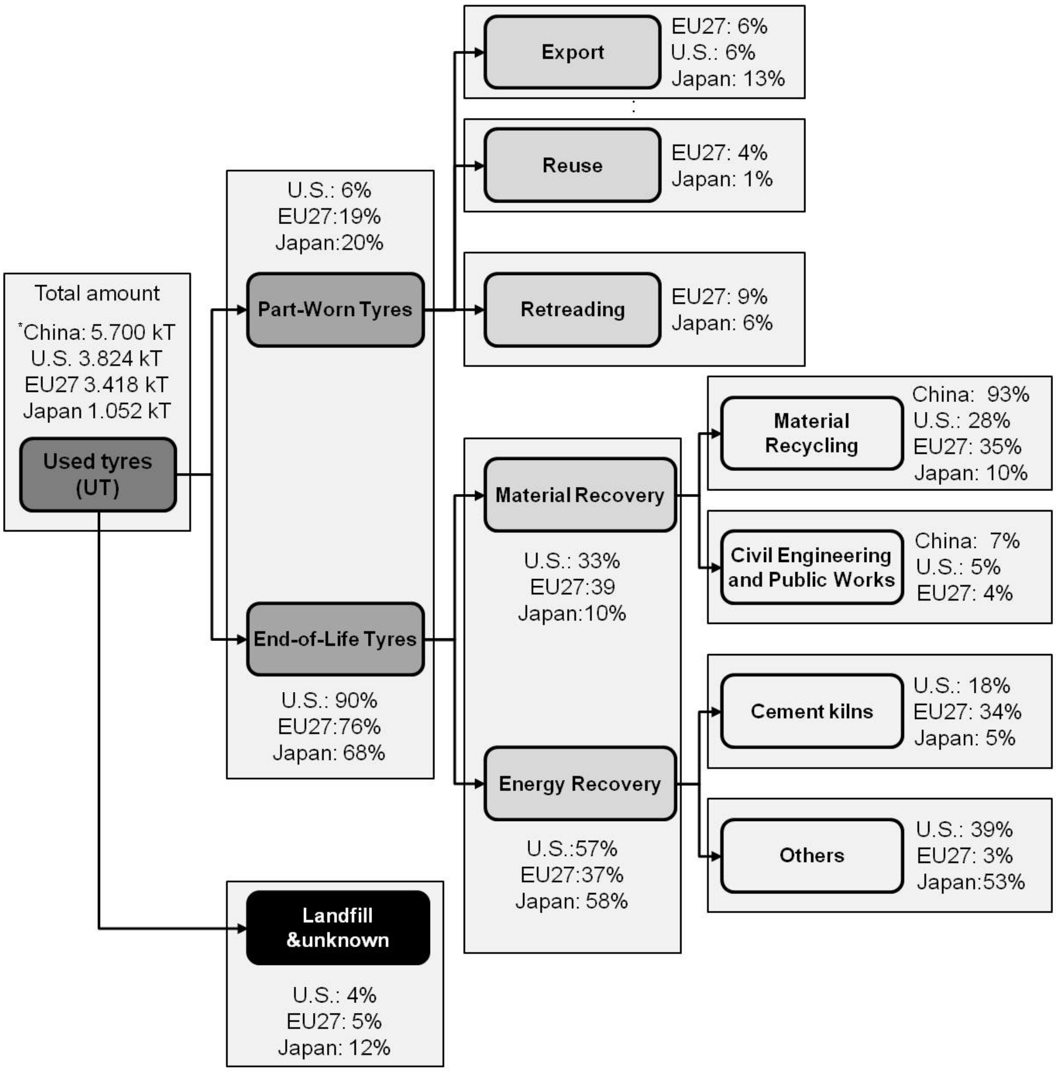
Wang, H.Z., Xu, H., Xuan, X.J., 2009. Review of Waste Tire Reuse& Recycling in China: current situation, problems and countermeasures. Adv. Nat. Sci. 2, 31-39.

WBCSD – World Business Council for Sustainable Development (Switzerland), 2008. The Report Managing End-of-Life Tyres. http://www.wbcsd.org/templates/TemplateWBCSD5/layout.asp?type=p&MenuId=MTYwNg&doOpen=1&ClickMenu=LeftMenu (accessed January 2015).

Williams P.T., 2013. Pyrolysis of waste tyres: A review. Waste Manag. 33, 1714–1728.

Yehia, A.A., Mull, M.A., Ismail, M.N., Hefny, Y.A., Abdel-Bary, E.M., 2003. Effect of Chemically Modified Waste Rubber Powder as a Filler in Natural Rubber Vulcanizates. J. Appl. Polym. Sci. 93, 30–36.

Zhang, X., Zhu, X., Liang, M., Lu, C., 2009. Improvement of the Properties of Ground Tire Rubber (GTR)-Filled Nitrile Rubber Vulcanizates Through Plasma Surface Modification of GTR Powder. J. Appl. Polym. Sci. 114, 1118–1125.



*data include only the total amount of used tyres recovered by recycling

Fig. 1 Comparison of the ways of recovery and flow of used tyres (UT) in China, U.S., Japan and European Union countries (JATMA, 2015; Hu et al., 2014; ETRMA, 2013; RMA, 2013; Li et al., 2010; Wang, 2009).

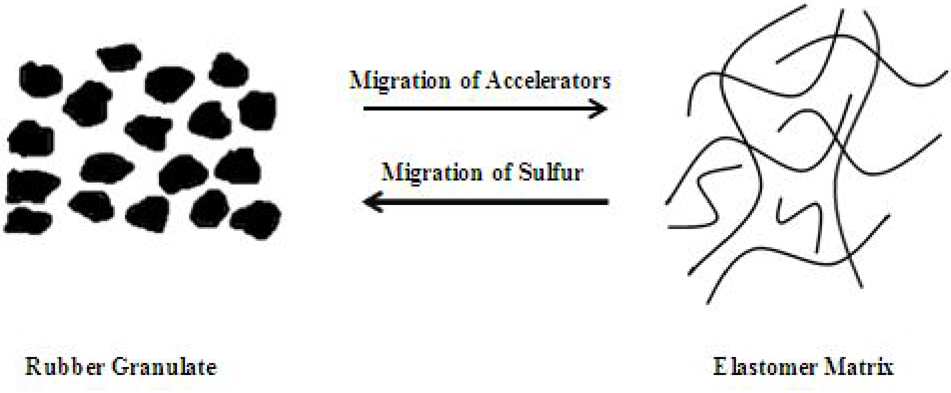


Fig. 2 Direction of migration of sulfur and accelerators in the rubber composites.

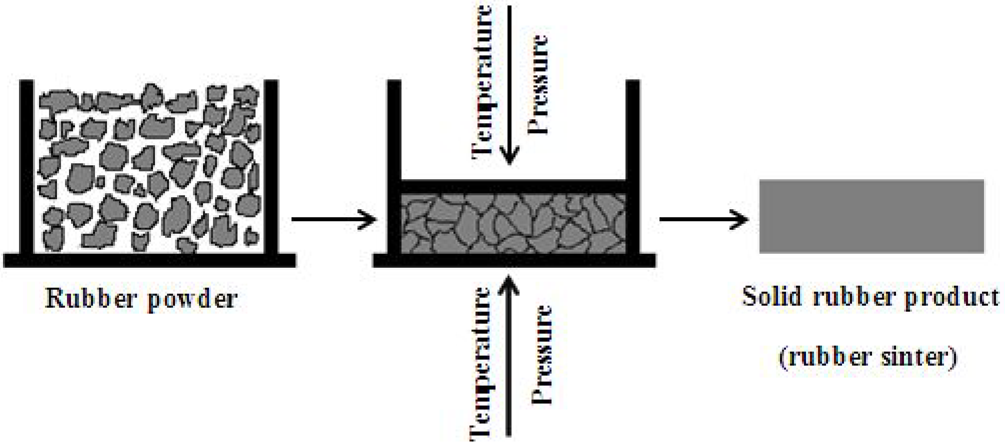


Fig. 3 Process of sintering of rubber granulate.

Table 1. The legislation regulation and models in management of used tyres recovery in EU27, U.S., Japan and China (JATMA, 2015; Hu et al., 2014; ETRMA, 2011; ETRMA, 2013; CRIA,2010; RMA, 2009; Li et al., 2010; Wang, et al.,2009; WBCSD, 2008).

Legal regulations in the management of used tyres	EU27	**Directive on the Landfill of Waste 1000/31/EC (1999)** Ban on the stockpiling whole used tyres in the landfills since July 2003; ban on the stockpiling grinded used tyres in a landfill since July 2006. **End of Life Vehicle Directive 2000/53/EC (2000)** (85% of scrap cars have to be recovered since 2006; tyres have to be dismantled from vehicles before these are scrapped) **Directive on Incineration of Waste - 2000/76/EC** Sets standards of emission for all cement kilns since 2002; since 2008 ban to combustion used tyres in elder cement kilns; since December 2008, new regulations regulating co-incinerating waste tyres in cement kilns (in Europe the cement kilns currently are already complying with this Directive).
	U.S.	**Each state in the U.S. has its own law regulations concerning reuse/recovery and landfilling of used tyres:** (38 states ban on the stockpiling whole tyres in landfills; 35 states allow the stockpiling grinded tyres in landfills; 11 states ban on the stockpiling all tyres in landfills; 21 states allow the stockpiling grinded tyres into monofills (landfill for the disposal of one kind of material); 8 states don't have restrictions on stockpile used tyres in landfills.); 36 states require permits from used tyres haulers; 37 states have state fees for used tyres; 14 states require financial assurances from used tyres haulers; 32 states require financial assurance from recovery companies; the U.S.; Environmental Protection Agency (EPA) reporting that 48 states currently have laws and regulations for management scrap tyres.
	Japan	Ban on the stockpiling whole and grinded used tyres in a landfill; Very restrictive law strictly regulating the collection, transportation and recycling of used tyres – contracts, system require accurate and detailed reporting about tyres recovery.
	China	In China the policy and the legislation on the management of waste tyres are slowly but continuously developed. There are several regulations in China concerning collection, transport, recycling, import and export of used tyres: The Ministry of Industry and Information of China in July 2012 published an ordinance that the recycling companies have to achieve recovery rate of used tyres no less than 10,000 tons per year. Most of the used tyres recyclers in China (approx. 90%) has low level of environment friendly technology and it will be difficult to reach that threshold. The dismantlement and recycling of used tyres should be compatible

		with provisions of relevant laws and regulations. Nowadays, the companies which are registered by industrial and commercial administration do not need special license to collect the tyres; The haulers do not need any license for transport scrap tyres only, if they transport them out of the administrative region of a province, autonomous region or municipality for storage or recovery. The illegally oil refining are strictly prohibited (banned); Only non-vulcanizated rubber scrap or rubber powder can be import.
Model of the management for used tyres	EU27	**Extended Produced Responsibility (EPR)** (for the management of used tyres are responsibility the producers and importers of tyres, valid in Finland, Hungary, Italy, Lithuania, Latvia, Estonia, the Netherlands, Poland, Czech, Romania, Spain and Sweden, Belgium, France and Portugal, Bulgaria, Greece, Italy, Norway) **Tax system** (cost of recovery used tyres is included in their price, valid in Slovakia and Denmark) **Free market** (recovery/recycling of used tyres is cost effective for the recyclers, valid in Ireland, UK, Germany, Switzerland, Austria and Croatia)
	U.S.	**Mixed system** (each state in the U.S. has its own model of used tyres management)
	Japan	**Free market system** (recovery/recycling of used tyres is cost effective for the recyclers)
	China	**Free market system** (recovery/recycling of used tyres is cost effective for the recyclers)

Table 2. The influence of components type and ratio, particle size of rubber granulate, and processing on the properties of polymer-rubber composites obtained from rubber granulate and natural or synthetic rubber.

Composition		Average particle size of rubber granulate	Steps of composition preparation	Comments	Results	References
Components	amount [phr]					
NR	20,6	500 µm	1. Mixing compounds with mixer 2. Mixing on two roll-mill	• RRP used as a partial substitution for carbon black	• Replacing black filler by RRP in amount up to 15 % by weight do not affect in tensile strength, permanent elongation and hardness • Replacing black filler by RRP in amount up to 5 % by weight do not affect in wear resistance	(Dekic et al., 2012)
SBR (styrene-butadiene rubber)	79,4					
RRP (recycled rubber powder)	0-21					
N-220 (carbon black)	53-32					
NR	100	150-250 µm	1. Mixing on two roll-mill	• CB used as a partial substitution for WTD	• Elongation at break increase with increasing amount of WTD • Tensile strength and tensile modulus increase with increasing amount of CB • The highest amount of CB the highest interaction between components	(Ismail et al.,2010)
WTD (waste tyre dust)	30-0					
CB (carbon black)	0-30					
NR (Natural rubber)	100	125-500 µm	1. RP surface oxidation 2. Mixing blends on two roll-mill 3. Vulcanization in a hydraulic press	• Surface oxidation carried out with nitric agents or hydrogen peroxide solution	• Chemically modified RP enhance tensile strength, and aging resistance compared to unmodified RP	(Yehia et al., 2003)
HAF (high-abrasion furnace carbon black)	0-50					
RP (rubber powder)	50-0					

Composition		Average particle size of rubber granulate	Steps of composition preparation	Comments	Results	References
Components	amount [phr]					
NBR (acrylonitrile butadiene rubber)	100	250 µm	1. Mixing on two-roll mill 2. Vulcanization in a compression-molding press	• RG modified by plasma treatment	• Contact angle of RG decrease from 124,7° to 0° after plasma treatment • Plasma treatment improving interfacial adhesion between components • Modified RG improved mechanical properties of composition compared to unmodified RG	(Zhang et al., 2009)
RG	0-20					
NR	100	1. 150-250 µm 2. 450-600 µm 3. 1200-1500 µm	1. Mixing on two-roll mill 2. Vulcanization in a hydraulic press	-	• Tensile strength and elongation at break decreased with amount of WTR increased • Smaller particles of WTR affect enhancement in the mechanical properties • Silica content improved mechanical properties compared to raw NR	(Chandran et al., 2015)
WTR (waste tyre rubber)	0-40					
Silica	20					

Table 3. The influence of type of components, particle size of rubber granulate, components ratio and processing on the properties of polymer-rubber composites obtained from rubber granulate and thermoplastics.

Composition	Percentage ratio of components	Average particle size of rubber granulate	Preparation blends/speciman	Other ingriedients	Results	References
Recycled HDPE/RG (High-density polyethylene/rubber granulate)	100/0-30/70	250 µm	Two roll mill/hot plate press	-	• Mechanical properties decrease with amount of RG increases • Degradation temperature increases with addition of RG	(Crespo et al., 2010)
LLDPE/RG/EOC (Linear low density polyethylene/rubber granulate/ethylene-1-octene copolymers)	50/0/50-50/50/0	500-425 µm (40 mesh)	Twin screw extruder/ Injection molding		• Tensile strength increase with amount of RG decrease • Elongation at break decrease with amount of RG increase • If 5 % by weight of EOC is added, the ultimate tensile properties are obtained	(Rocha et al., 2014)
MAPE/RG (Maleate polyethylene/rubber granulate)	100/0-20/80	300 µm	Twin screw extruder	• UV absorber • Thermal stabilizer • Hindered amine light stabilizer • Antioxidant	• Addition of RG led to degradation of MAPE/RG during thermal ageing at 80ºC • Addition of RG and stabilizers improved stability against weathering	(Kakroodi and Rodrique, 2013a)
MAPE/RG/talc MAPE/RG/wood flour	50/50/0-40/40/20	200 µm		-	• Inclusion of talc and wood flour improved tensile modulus • Addition of talc and wood flour led to reduce tensile strength and strain at break • Thermal stability decrease with increasing amount of wood flour • Talc don’t affect on thermal stability of MAPE/RG	(Kakroodi and Rodrigue, 2013b)
BR/PE/RG (Butyl rubber/Polyethylene/rubber granulate)	0/50/50-30/20/50	1,5 mm	Screw extruder	• Dicumyl peroxide (DCP) • Maleic anhydride (MA)	• Tensile strength and hardness decrease with BR increasing • Elongation at break increase with enhance amount of BR • DCP and MA improved mechanical properties	(Formela et al., 2013)
PVC/RG	100/0-30/70	• 200 µm	Screw	-	• Reducing in tensile strength,	(Orrit et

Composition	Percentage ratio of components	Average particle size of rubber granulate	Preparation blends/speciman	Other ingriedients	Results	References
(Polyvinyl chloride/rubber granulate)		- 200-500 µm - >500 µm	extruder/Press		toughness and stiffness with amount of RG increasing - Addition of RG lead to increases in conductivity, dielectric permittivity, and dielectric loss factor	al., 2011)
PP/SBR/RG	30/40/30-30/0/70	450 µm	Screw extruder/Injection molding	- DCP - N,N'-m-phenyle-bis-maleimide (BMI)	- Ultimate tensile strength and elongation at break - Inclusion BMI improved properties	(Magioli et al., 2010)