



Review

# **Crumb Rubber Modifier in Road Asphalt Pavements: State of the Art and Statistics**

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Abstract: Tire rubber recycling for civil engineering applications and products is developing faster, achieving increasingly higher levels of maturation. The improvements in the material circle, where crumb rubber, generated as a by-product of the tire rubber making process, becomes the resource used for the construction of road asphalt pavement, is absolutely necessary for increasing the sustainability of the entire supply chain. The paper reports the results of an accurate data analysis derived from an extensive literature review of existing processes, technologies, and materials within construction of infrastructure. The current position, the direction, and rate of progress of the scientific efforts towards the reuse and recycling of tire rubber worldwide have been shown. Furthermore, an in-depth analysis of a set of important properties of Crumb Rubber Modified Asphalt has been carried out—fabrication parameters, standard properties, high and low-temperature performance, and rheological properties. Statistics over a sample of selected publications have been presented to understand the main processes adopted, rubber particle size, temperatures, and possible further modifications of crumb rubber modified binder.

Keywords: crumb rubber; recycled tire rubber; asphalt rubber

#### 1. Introduction

Roadway infrastructures are ideal targets for effective sustainable design and construction initiatives. Frequently, they are large in project scope and involve considerable amounts of financial resources [1]. Also, highway road pavement construction and maintenance consume significant amounts of materials and energy and produce large amounts of waste, which may have adverse effects on the environment and cause social perturbations [2]. This is further worsened by a project's long construction time and service life, which, ideally, requires maintenance to be performed on a regular basis. Furthermore, while developing countries are still going through a phase of strong investment in the construction of new road infrastructures, the majority of developed countries have just started to experience a change in their investment needs. While the past has seen a considerable amount of financial resources being allocated to the expansion of their road network, the future trend will be towards making the best possible use of the available infrastructures. This is necessary to accommodate growing transport demands, while ensuring that traffic density and the condition of road infrastructures remain at desirable levels, and that the road network is adaptable, automated, and resilient [3]. Failure to make significant progress towards fulfilling road infrastructure investment needs, disregarding sustainability concepts, could prove costly in terms of wasted time, consumption of non-renewable resources, and growing environmental problems, with all the implications this has for living standards and quality of life, or maybe even lead to a permanent and irreversible partial loss of this important asset. One of the main indicators for assessing the sustainability of a project is the "secondary material consumption", i.e., the amount of the recycled materials used in the project as

Coatings 2019, 9, 384 2 of 22

material recovered from previous use or from waste, which is substituted for primary materials. It is usually measured as a percentage of recycled materials used related to the total material consumption. Alternatively, this indicator can be expressed in mass units [4]. This indicator allows the potential benefits arising from the use of reclaimed materials to be quantified, and recently several efforts have been dedicated to increase the percentage of recycled material in road construction. Therefore, pavement engineers are constantly seeking for innovative solutions that allow efficient processing of resources through recycling chains, with a drastic reduction of residual wastes, dissipation of by-products, and consumption of raw materials in road construction. In the last years, the growing popularity of crumb rubber, recycled material coming from End-of-Life Tires (ELTs), has led to several studies being conducted, aiming to improve the material cycle where rubber, generated as a by-product of the scrap tires crushing and sieving, becomes the resource needed in another production process for construction of road pavement layers. The crumb rubber (CR) has different applications in the production of asphalt mixtures. The purpose is to find an alternative to tire disposal and improve the performance of asphalt mixtures. Indeed, tires have a wider range of performance temperatures than bituminous mixtures, because they do not melt in the heat and they do not crack in cold temperatures. The potential advantages of rubberized asphalt mixtures are to be found in their higher resistance to permanent deformation and thermal cracking for a higher durability of the material. Nevertheless, the performance of rubberized materials differs, significantly depending on the processes and technology used. For instance, on one hand certain studies highlight that CR modified mixtures are less susceptible to moisture damage compared with traditional mixtures [5], and the rubber particles contribute to achieving higher performance of both binders and mixtures at high temperatures [6]. On the other hand, if other processes are adopted, certain studies show that the presence of higher volume of air voids caused by the decrease in the compaction of the mixture can lead to higher moisture content. Therefore, these types of CRM mixes are more susceptible to moisture than conventional mixes [7].

The addition of crumb rubber typically results in the increase of the complex modulus of the binder and in the reduction of the phase angle, in particular at high temperatures, providing a favorable effect on the rutting resistance [8]. Nevertheless, these materials are not devoid of limitations. For instance, one of the limits of the industrial production of this material is the instability during storage. The rubber modified bitumen has higher heterogeneity, and the difference in density between bitumen and rubber granulate is significant. For these reasons the rubber particles have the tendency to fall to the bottom, decreasing the storage stability [9].

It is possible to understand from the previous considerations that there are several types of processes and technologies when crumb rubber is used in asphalt mixture applications that result in different performance and durability. With this in mind, this paper provides the scientific community with description and nomenclature of key mechanisms, processes, and technologies linked with the use of crumb rubber in road pavement, together with a pragmatic framework to identify and possibly quantify the key properties through an extensive literature review, the extrapolation of the meaningful data, and the statistical analysis of the information gathered.

#### 2. Tire Rubber Industries

The number of vehicles in the world's major markets is constantly and progressively increasing. This trend is especially evident for China, where it is expected to move from 147 million passenger cars in 2016 to 309 million in 2024 [10]. The increase in Europe is more modest—from 329 million in 2016 to 349 million in 2024, which is probably due to the efforts that Europe devotes to improve public transport. On the other hand, the number of retreated tire sales in Europe is decreasing progressively [10]. That means that the interest in recycling tires to rebuild them is decreasing. This is probably due to the difficult and demanding process of reclaiming and re-vulcanizing the rubber that is used in new tires. Therefore, other solutions for recycling ELTs have to be considered or improved.

Coatings 2019, 9, 384 3 of 22

The most commonly used material in the composition of the car tire compound is the Stirene Butadiene Rubber (SBR) polymer, a synthetic rubber (SR) composed of stirene and butadiene, while the truck tire compound is mainly composed of natural rubber (NR). NR is a crude high molecular polymer whose main constituent is polyisoprene. After proper filtration and solidification from the Hevea brasiliensis, the latex becomes natural rubber, an elastic material with high durability fatigue resistance [11]. Europe has a demand for NR equal to 76% of the total demand for rubber goods. This quantity has been quite stable in the last five years. On the other hand, the demand in Europe for SR for tires is lower, equal to 48% of the total amount required for rubber goods, and has been almost stable in the last five years. However, the biggest consumer of NR and SR is China, with 39% and 29% of consumption, respectively, while Europe and United States are far behind these quantities—Europe consumes 9% of NR and 16% of SR, and the United States consumes 8% of NR and 13% of SR [10].

The crumb rubber used for road construction materials can be obtained from both types of tires (car and truck), generally at 50% each, and therefore can be composed of both SBR and NR, indicated with the abbreviation SBR/NR.

#### 3. History of Reclaimed Rubber in Construction of Infrastructures

The first experiment mixing natural rubber and bitumen to obtain a modified binder took place in the 1840s. A few years later, both synthetic and natural rubber started being used to obtain a modified bitumen and the fabrication process was progressively improved during the 1930s [12,13]. Tires have been used extensively in asphalt binders since the early 1960s.

The U.S. Department of Transportation (DoT) activities in this area have also stimulated the local public agency interest in the use of ELTs in highway applications [14].

Historically, two processes have been adopted for using the CR in asphalt mixtures—the wet and the dry processes, explained in the two subsections below.

#### 3.1. History of the Wet Process

The term "wet process" is used to describe a whole group of technologies that differ from each other for the applied conditions. The following section has the aim of retracing the development steps of these technologies.

In the 1960s, McDonald [15] in the United States developed one of the first commercial binder systems exploiting the principles of the wet process, i.e., the dissolution of the crumb rubber in bitumen as a modifying agent. McDonald's experimental work was conducted with Atlas Rubber, the Arizona DoT, and Sahuaro Petroleum and Asphalt Company. For the first time, he was able to emphasize the beneficial properties of both elements.

Afterward, in the mid-1970s, Arizona Refining Company (ARCO) developed and patented another asphalt-rubber wet process product [14]. Between the 1970s and the 1980s, the Arizona DoT sponsored comprehensive research work for the deep understanding of the asphalt-rubber binder system. The results of this research highlighted that the properties of asphalt-rubber mixtures depend on different variables, such as rubber gradation and concentration, as was definitely proved over the years [14]. Other important factors influencing the properties are rubber and bitumen types, cure time, fabrication time, and temperature. The technology was further refined in the 1980s and continuously developed until 1988, when a definition of rubberized bitumen appeared in the American Society for Testing and Materials (ASTM) in the ASTM D8 and later in ASTM D6114-97 [16].

Between the 1980s and 1990s, the wet process has been split into different categories—McDonald process, continuous blending, and terminal blending. The first method refers to the original wet process, also commonly called Asphalt Rubber (AR) or wet process high-viscosity, developed in the 1960s. The bitumen-rubber blend is fabricated in a blending tank, mixing crumb rubber and bitumen at a high speed for 1–2 min. Afterward, the modified binder is collected in another tank and mixed with augers, where the blend stays for a sufficient period of time (45–60 min) to allow the circulation and consequently the reaction of the blend. The resulting binder is then used to produce bituminous

Coatings 2019, 9, 384 4 of 22

mixtures [16]. The second method, the Continuous Blending (CB), developed in Florida and commonly called "Florida wet process", describes a wet process in a continuous operation rather than a batch procedure [17]. This allowed the concern regarding the McDonald wet process that requires batching and fixed reaction time associated with blending bitumen and rubber to be overcome. Indeed, as previously exposed, this reaction is dependent on a number of factors, including the size and the concentration of CR [18]. The Florida technology differs from the McDonald process in several aspects—the reaction occurs during the blending using a unique unit with agitators, and lower percentages of Crumb Rubber Modifier (CRM) (from 8 to 10 percent rubber) with a smaller size. The continuous blending system requires lower mixing temperature, and shorter reaction time [17]. The third method, commonly called Terminal Blend (TB), was developed and commercialized by NESTE, U.S. Oil, Bitumen, and BAS Recycling BEUGNET [14]. TB is defined as "a form of the wet process where crumb rubber modifier is blended with hot asphalt binder at the refinery or at an asphalt binder storage and distribution terminal and transported to the asphalt concrete mixing plant or job site for use" [19]. No subsequent agitation is reportedly mandatory for such a binder to keep the ground tire rubber particles evenly dispersed in the matrix binder.

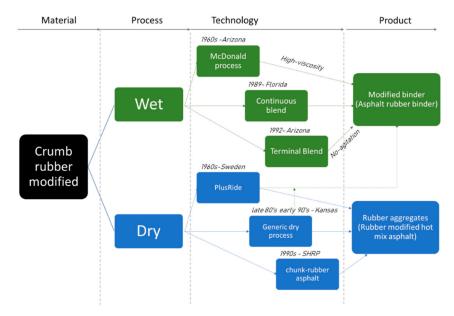
The Rubberized Asphalt Concrete Technical Assistance of California's Department of Resources Recycling and Recovery [20] also defines TB as "CRM modified binders that do not require constant agitation to keep discrete rubber particles uniformly distributed in the hot asphalt cement". The terminal blend system can also be implemented in the field and in the asphalt mixing plant, not only in the binder terminal. Therefore, the term "wet-process-no-agitation" has been proposed to avoid confusion [11]. There are no common technical guidelines and recommendations about rubber and bitumen properties in the Terminal Blend, their evolution during the process, and the final performance [11]. Nevertheless, these properties are strongly correlated with the evolution and the kinetics of the rubber degradation. Indeed, the rubber undergoes a process of devulcanization (described in Section 3.3).

#### 3.2. History of the Dry Process

Parallel to the wet process in the 1960s, scrap tires were processed and also used to replace the virgin aggregates in bituminous mixtures. This type of process is called the "dry process". The dry process, used most frequently in the United States, was developed in Sweden in the late 1960s. Two companies replaced a small portion of aggregates with the same fraction of rubber grains. When crumb rubber is used as a portion of aggregates in hot mix asphalt concrete, the resultant product is sometimes referred to as rubber-modified asphalt concrete (RUMAC). This type of mixture usually contains 1 to 3 percent of rubber by weight of the total aggregate in the mix and target air void content from 2 to 4 percent [17]. In the 1980s, the mix design and the technology was refined, achieving the production of a bituminous mixture commercially called PlusRide [21]. Other dry process techniques include those developed by the Army Corps of Engineers at the Cold Regions Research and Engineering Laboratory (USACRREL, Hanover, NH, USA) [14,22].

In the late 1980s and early 1990s, new concepts related to the dry process were introduced—the generic dry process and chunk-rubber asphalt concrete. Takallou introduced the concept of a generic dry process [23]. Unlike PlusRide, which requires a specific gap gradation for aggregates, the generic dry process, introduced by Takallou, admits "generic" aggregate gradations, hence the definition, generic dry technology [24]. The chunk-rubber asphalt concrete is the result of a modification of the mix design of Plusride proposed by the Cold Regions Research and Engineering Laboratory (CRREL), and it is characterized by an increase of the CR maximum size and the percentage of rubber used in HMA. Figure 1 summarizes the principal materials, processes, technologies, and products for the use of reclaimed rubber in road materials.

Coatings 2019, 9, 384 5 of 22



**Figure 1.** Principal materials, processes, technologies, and products for the use of reclaimed rubber in road materials. Note: Greenbook (2006) proposes three types of production processes: (i) wet process; (ii) dry process; (iii) terminal blend process.

Note that Figure 1 shows a connection between PlusRide and the generic dry process with the modified binder. Indeed, the interaction between rubber and bitumen also occurs in the dry process, even if it is lower because the rubber is not directly used to modify the binder.

One of the main concerns raised by the use of the dry process is that it is not possible to determine the binder properties directly. In order to evaluate the binder properties, it is necessary to extract and recover them, however, these operations alter the characteristics of the CRM bitumen. Even if the contact bitumen-rubber is not direct, as in the case of the wet process, the binder during the dry process is affected by the same parameters. Dry-processed binder systems are partially reacted [14].

It is reasonable to assume that the density of the bitumen does not change significantly in the case of interaction during the dry process, while the density of the bitumen changes significantly in the wet process [25].

#### 3.3. Vulcanized and Devulcanized Rubber for Bituminous Mixtures

The increase of concern to proper managing the ELTs has renewed the interest in the last ten years in another recycling method, called devulcanization [26].

To produce tires, the rubber undergoes a vulcanization process by using sulfur, peroxides, and other substances intended to prevent the tires from cracking and to improve their properties [27]. Vulcanization, therefore, is a process where chemical bonds are created between sulfur and the carbon molecules of rubber. Since the vulcanization is an irreversible process at ambient temperature and atmospheric conditions of pressure, an industrial devulcanization process is necessary to restore the properties of the rubber to be used as virgin rubber [26]. Devulcanization refers to the process in which the crosslink bonds in the vulcanized rubber are selectively broken, cleaving the sulfur–sulfur or carbon–sulfur bonds and shortening the molecular chains [28]. Indeed, the three-dimensional network structure restrains the rubber from melting [29,30]. Therefore, the reclamation of scrap tires is mainly related to shredding and devulcanization of crumb rubber [27]. When rubber is intended for use as an element in wet or dry processes for asphalt mixtures, reclaimed rubber (i.e., devulcanized rubber as a result of the use of chemicals, mechanical, and thermal energy) that has regained its viscosity, as well as the characteristics of the original compound [31], is preferable. In this case, a more homogeneous blend with the binder may be obtained because the rubber acts mainly as a flexible

Coatings 2019, 9, 384 6 of 22

filler [32,33] that can be stored at high temperatures for longer periods without having problems related to its sedimentation [34].

Typically, the performance of asphalt mixtures containing CR depends on multiple factors. They are as follows: (i) the percentage of rubber; (ii) the surface of the rubber particles (criogenic grinding produces smooth particles with relatively lower surface area than ambient grinding [16]); (iii) the dimension of the rubber particles; and (iv) the type of rubber treatment (i.e., vulcanized or devulcanized). Furthermore, in the case of devulcanization, the used method also plays an important role [35] due to the fact that excessive devulcanization may lead to the deterioration of the binder properties.

The devulcanization process encompasses several steps. They can be summarized as follows: (i) shredding the tires to small particles of rubber; (ii) fiber and steel removal through the use of suitable separators; and (iii) further grinding of the rubber to a finer size and then mixing with different reclaiming agents [36]. The most studied devulcanization strategies involve mechanical, chemical, physical, biological, microwave, and ultrasonic processes [27,30,37,38].

The resultant devulcanized rubber might have different properties because they depend on the composition of the vulcanized compound; for instance, truck tires are composed mainly of NR, whereas passenger tires are mainly constituted of SR [39]. Compared to the traditional dense-graded asphalt rubber mixtures, a binder modified with devulcanized rubber allows a lower binder content to be achieved in the asphalt mixture, reducing economic costs. Moreover, it allows the fabrication temperature (approximately 165 °C) to be reduced, therefore reducing engineering risks and energy consumption. All these aspects contribute to simplify the fabrication process, obtaining a binder with high hot storage stability [40,41]. Other authors [42] found that devulcanization resulted in the higher surface activity of the crumb rubber and improved the compatibility with bitumen.

The rubber can be industrially devulcanized before its use in asphalt mixtures (pre-devulcanized) or it can be devulcanized during the production of CR modified bitumen. The crumb rubber particles can be devulcanized and depolymerized in the binder at high temperatures and high shear mix [33]. The devulcanization and depolymerization of rubber is higher when the binder is rich in aromatics [43].

## 4. Overview of the Differences between Processes and Technologies

### 4.1. Types of Bituminous Materials Containing Crumb Rubber: Definitions

The terminology proposed by FHWA publications [22,24] found acceptance among binder users and producers in the 1990s. Nevertheless, the increase of the scientific interest during recent years has led to the proliferation of names to describe similar concepts. This might lead to confusion and overlap. Hence, in this section clear terminologies associated with the definitions will be provided to the reader in order to unequivocally and unambiguously identify the correct technology. The goal is to provide a common ground for the next sections of the paper, as well as possibly provide asphalt technologists with common ground for further investigations. Table 1 reports a summary of the different processes and technologies for recycling crumb rubber in asphalt mixtures.

**Table 1.** Summary of name and definitions of the different processes and technologies.

Process Name and Definition  Wet process: "any method that blends CRM with the asphalt cement before incorporating the binder into the asphalt paving materials. Normally they require agitation, but they can be formulated so as not to require agitation."					
Wet-process-High-Viscosity	"blend of asphalt cement, reclaimed tire rubber, and certain additives, in which the rubber component is at least 15 percent by weight of the total blend and has reacted in the hot asphalt cement sufficiently to cause swelling of the rubber particles"	<ul> <li>McDonald system</li> <li>Recycled Tire Rubber Modified bitumen (RTR-MB)</li> <li>Asphalt Rubber binder</li> <li>Bitumen-rubber binder</li> <li>Crumb rubber binder</li> <li>Batch blending</li> </ul>	[19,22,44–46]		

Coatings 2019, 9, 384 7 of 22

Table 1. Cont.

	Process Name and Defin	ition			
Wet process: "any method that blends CRM with the asphalt cement before incorporating the binder into the asphalt paving materials. Normally they require agitation."					
Technology Included in This Process	Technology Definition	Other Names of Technology	References		
Continuous blend	CRM and the binder are continuously blended during the mix production and then stored in tanks. It has a unique unit with agitators, and the reaction occurs during the blending. It is a continuous production system that uses a finer grind material compared to the McDonald system.	Florida technology	[16,18,21,22]		
Wet-process-no- agitation	A form of the wet process where CRM is blended with hot asphalt binder at the refinery or at an asphalt binder storage and distribution terminal and transported to the asphalt concrete mixing plant or job site for use.	<ul> <li>Terminal blend (produced at the refinery)</li> <li>Field-blends (directly produced in the field)</li> <li>Terminal blend long time</li> <li>Recycled Tire Rubber Modified Bitumen (RTR-MB)</li> <li>Terminal Blend Rubberized Asphalt (TBRA),</li> <li>Rubber Modified Binder, RMB</li> </ul>	[19,47–50]		
	Process Name and Defin	ition			
	mixes the crumb rubber modifier dry with the aggregate l bber aggregate in the paving mixture. This method applie		er. The CRM acts a		
Technology Included in This Process	Technology Definition	Other names of Technology	References		
RUMAC	CRM is used as a rubber aggregate, which is incorporated prior to mixing with binder, producing a rubber modified hot-mix asphalt	PlusRide (commercial name)	[21,51]		

#### concrete. The granulated CR used is between 2-6.3 mm. Similar to the RUMAC technology, however it uses slightly lower percentage of CRM. Generic dry process TAK system [21,23,51] The CRM particle size is lower (finer particles compared to RUMAC, 0.18-2 mm) CRM is used as a rubber aggregate and is incorporated prior to mixing with binder, producing a rubber modified hot-mix asphalt Chunk rubber asphalt [51-53] concrete. The granulated CR is bigger (4.75-9/12.5 mm) and used in higher quantities (3%–12% of the weight of the mixture)

#### 4.2. Wet Process vs. Dry Process in Terms of Performance

Despite the unstable performance of the dry process compared to the wet process, the dry process presents certain advantages, such as the use of rubber particles that are much coarser than the wet process, facilitating industrial production. The amount of rubber can be significantly higher (the dry process can use from 2 to 4 times as much as the wet process), increasing the rate of the recycling material, and the incorporation in the mixture is easier (in the dry process there is no need for special equipment, while in the wet process special mixing chambers, reaction and blending tanks, and oversized pumps are required) [54]. Referring to low noise pavements, according to some authors, when using the dry process in experimental sections, the noise absorption can be significant and higher if compared to the wet process [55,56]; it seems this effect is amplified by the devulcanization treatment [57]. The potential improvement of the acoustic absorption is attributed to the improved chain flexibility of the molecules after the devulcanization treatment [58]. The main problem with CRM dry mixes is the lack of cohesion between the crumb rubber and bitumen. This can cause higher moisture susceptibility and detachment of aggregates [7]. On the contrary, the advantages of the dry process are mainly related to two phenomena: larger CRM particles act as a flexible substitute to the aggregate they replace, and the binder modification that may occur with the addition of the finer CRM particles [59].

The rubberized materials obtained with the wet process present numerous advantages. In this case, the improved properties of the mixture are based on the modified binder that acquired higher

Coatings 2019, 9, 384 8 of 22

performance. When the binder is modified it transfers its properties to the mixture. Therefore, since the binder stiffness increases with the presence of CR, the stiffness of the mixture also increases, offering a higher rutting resistance [59]. The optimum binder quantity is higher for AR mix than for traditional. On one hand, this could be considered a disadvantage for the mixing stability and for the cost implied, while on the other hand the higher amount of binder required provides better aging properties because of the binder film thickness around the aggregates is higher.

The addition of rubber enhances the elasticity of the mixture, improving the resistance to the repeated tensile stress caused by the passages of numerous vehicles. Therefore, fatigue resistance is considerably improved for the rubberized mixtures obtained by the wet process binder modification.

Binder that is less stiff could also help the AR binder to achieve less thermal cracking at low operating temperatures [18]. CRM modified binders also show higher resistance to the effects of high temperature than the traditional binder, and this leads to improved rutting resistance when the CRM mixture is tested [6].

Other studies claim that it is possible to reduce the layer thickness when the CRM mixture is used in asphalt pavement [59]. This led to quantify the potential economic and environmental benefits on a life cycle basis of the wet process compared to the dry process [60–62]. Despite the numerous advantages reported in the case of the wet process, the interactions occurring between crumb rubber and bitumen are not fully understood [63]. There are two important phenomena occurring when the bitumen is modified with crumb rubber [33,63]:

- Absorption of light fractions of binder by crumb rubber;
- Decomposition and depolymerization of crumb rubber in the binder.

These two phenomena contribute to increase the heterogeneity of the rubber modified bitumen and the difference in density between bitumen and rubber granulate becomes significant. For these reasons the rubber particles have a tendency to fall to the bottom, decreasing the storage stability [64]. Several attempts have recently been made to overcome this problem. Recent studies [63] show that, for example, using smaller rubber particles leads to a significant improvement in the stability of CRM bitumen. Even higher improvement can be achieved when chemical compounds that promote the interaction between bitumen and rubber particles are added to the binder [9].

#### 4.3. Wet-Process-High-Viscosity vs. Wet-Process-No-Agitation Techniques

The difference between traditional wet process high viscosity and wet process no-agitation technique is the chemo-physical rubber condition in the blend. The rubber particles swell once in contact with the bitumen at high temperatures due to the absorption within the polymer chain of the lighter parts of the bitumen. This is the key to obtain a gel-like material. At the same time, the bitumen loses the oil fraction, the rubber particles increase in volume, and the distance between the rubber particles is reduced. This entails an increase in viscosity and an increase in thickness around the aggregates due to the gel structure assumed by this modified binder [16]. If the interaction is longer and the temperatures are higher (from 220 to 260 °C), the swelling gives way to the devulcanization and degradation of the rubber particles, the crumb rubber is fully digested into the bitumen, and the storage stability of the final binder is improved [65]. The obtained binder is homogeneous and smooth without visible discrete particles, whereas wet process AR binder shows a rough and granular surface. In the case of AR binder, the presence of bigger rubber particles leads to the increase in thickness of the binder covering the aggregates (up to 36  $\mu$ m). A greater quantity of binder (7%–9%) is required, and this binder is successful when used with open-graded mixtures, i.e., when the percentage of voids is high, and this allows achieving increased coating thicknesses. The increased thickness improves the resistance to oxidation and aging, increasing the durability of the material. The rubber also gives elasticity to the binder. The increase in the bitumen film covering the aggregates, however, leads to an increase in the binder content with a consequent reduction in the benefits in terms of economic and environmental impacts [16]. AR binder is less-used for dense-graded mixtures; this is due to

Coatings 2019, 9, 384 9 of 22

the lower availability of voids. In this case, TB is preferred, which allows lower bitumen thicknesses. The high-temperature rheological properties of TB are lower than AR because the rubber elasticity is completely lost due to the depolymerization process. Nevertheless, with the addition of other modifiers, TB binder can regain considerable high temperature performance [66].

Both of the wet process binders ensure high performance of fatigue resistance and permanent deformation. The main advantages of the wet no-agitation compared to high-viscosity approaches are [16]:

- No agitation or special equipment is required and holding tanks are not needed to store the binder;
- Higher storage stability of the bitumen and higher workability of the bituminous mixtures;
- The asphalt mixture is produced at the same temperature as the polymer modified binder;
- Less bitumen is used to produce rubberized bituminous mixtures (5%–6%);
- Different applications are possible (dense-, gap-, or open-graded);
- This binder can also be used to produce bitumen emulsion.

The main disadvantages of the wet no-agitation approach compared to high-viscosity are [16,66,67]:

- Rubber elasticity is completely lost because of the depolymerization process;
- Lower rutting resistance and lower fatigue resistance;
- To achieve the same performance of AR it is necessary to add modifiers [66];
- Performance of TB is still under evaluation;
- Phase segregation problems are not completely solved.

The nature of the mechanism by which the interaction between bitumen and rubber takes place has not been fully characterized. Moreover, the assessment of the amount of CRM used in rubberized asphalt is not an easy and reliable operation to conduct.

Despite a few studies being available, no clear distinctions were made between AR and TB regarding the performance evaluation [11].

# 4.4. Field Long-Term Performance: Wet Process vs. Dry Process

In Oregon, during the period 1985–1994, several rubber asphalt pavement test sections were paved using wet and dry processes and subsequently monitored. The results showed the lower and unstable performance of the dry process [68]. This is attributed to the rubber "maceration" that leads to a stiffer bitumen being obtained [69]. Indeed, when the rubber is added to the asphalt mixture by means of dry process, the grains of rubber swell up because they absorb part of the volatile parts of bitumen (paraffin and maltenes) [69,70]. This effect has an important impact on the mechanical performance of mixtures containing CR compared to conventional asphalt, especially in the mixture response to plastic deformation [71,72].

Heitzman [24] showed that rubber particles can double their volume after fully absorbing oily parts of bitumen. The mix design of these material should carefully consider certain adaptations or limit expansion of the CR bituminous mixture.

Moreover, the production and paving temperature of rubberized mixtures should be increased to ensure the adequate compaction of rubber asphalt pavement.

During the construction of a test section it was found [24] that the temperature of dry rubber asphalt mixture drops significantly after compaction, therefore, on one hand it is necessary to compact them rapidly to perform an appropriate compaction and to avoid premature road surface damage. On the other hand, if the asphalt mixture is compacted immediately after paving a sticky wheel phenomenon will occur [24].

To overcome the difficulties in the compaction process it is recommended [73] to increase the fabrication temperature by 5–10 °C compared to traditional bituminous mixtures, and reduce the rolling speed during compaction.

It is recommended in a previous study [74] to store the mixture for more than 30 min before paving in order to give to the rubber particles and the bitumen a sufficient interaction time.

Coatings 2019, 9, 384 10 of 22

#### 5. Data Collection and Analysis

In this research work, two stages of analyses have been conducted:

• In the first step, the maturation point of the scientific interest at the global level has been evaluated. All of the publications from the 1970s until now have been collected from an international database (WebOfScience). The frequency distribution per year, country, and funding agencies have been calculated. Therefore, this first step allows "pictures" to be given that illustrate the current position, the direction, and rate of progress of the scientific efforts towards the reuse and recycling of tire rubber. Note that for the purpose of this research, only the Web of Science database has been consulted. Despite the existence of other publications in different languages and non-indexed publications, the authors decided to limit the statistical analysis to the indexed papers in the international database for a more defined tracking of results and for the significance of the related studies.

• In the second step, a deeper analysis has been conducted, focusing on a set of important properties of CRM bitumen. Among the total number of publications, a sample of approximately 100 papers has been selected to conduct an extensive literature review covering articles published in peer-reviewed international journals, reputed conferences, books, reports, guidance documents, and relevant research projects on the use of crumb rubber infrastructure construction. This literature review was meant to build a database of properties distributed by categories (fabrication process, standard properties, and rheological properties) that are intended to be the most relevant for a broader and complete identification of the main characteristics of CRM binder. The information collected was posteriorly used to build the statistics for each parameter or property.

The content of every item was analyzed and the main aspects were highlighted to be used in the two steps. The key information of two selected papers are for instance: materials used, type of grinding, devulcanization method, rubber content, particles size, mixing time and temperature, difference performance evaluated in the paper, Country and year of publication.

### 6. Analysis of Literature Data

#### 6.1. Global Overview of CR Use and Research

The frequency distribution by publication year is shown in Figure 2.

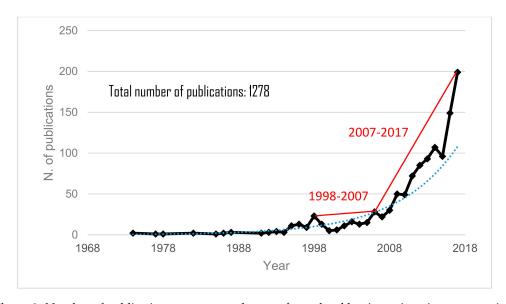


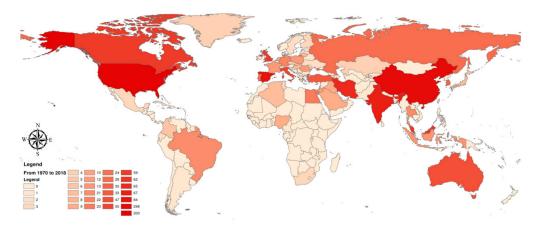
Figure 2. Number of publications per year on the use of crumb rubber in engineering construction.

Coatings 2019, 9, 384 11 of 22

The analysis of Figure 2 shows the increase in the number of publications regarding CR from 1970 up until 2018. Specifically, 1995 marks the real beginning of scientific interest in this field, probably due to the market entry of different products in the years before (Continuous Blend, Terminal Blend, Generic dry process, and chunk-rubber asphalt). The years between 1998 and 2007 register a change in the tendency observed previously, which may suggest that the research interest in the topic may have reached its maturation point. From 2007–2008 up until now, there is clearly a sharp increase (exponential) in the number of publications. This tendency suggests the influence of different aspects; one of these is surely the general increase in the number of publications in the academic domain. Nevertheless, the increase of the scientific interest for this kind of material is also due to the policies and consequently the markets that believe in the feasibility of the use of reclaimed rubber in engineering construction and intensively support the research in this direction.

Figure 3 shows the world map related to the spatial distribution of the scientific interests as the number of publications from the 1970s up until now for each country. The countries are color-scaled, from white, corresponding to zero publications, to red, which corresponds to the country with the highest number of publications related to the use of CR.

Figure 4 shows the number of publications funded by projects, government policies, grants, etc.



**Figure 3.** Color-scale map representing the scientific interest as the number of publications from the 1970s up until 2017 for each Country.

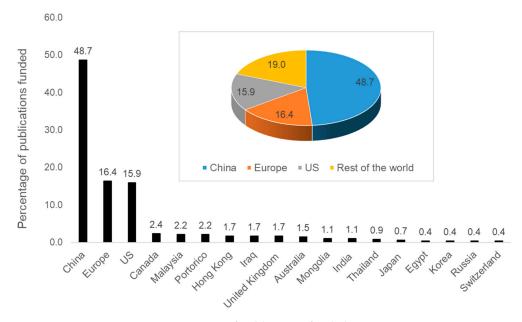


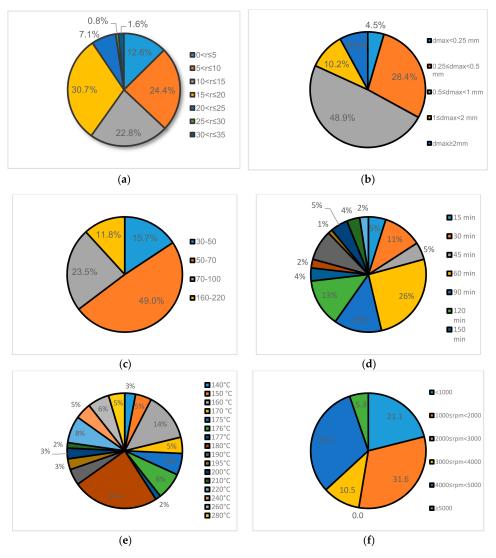
Figure 4. Percentage of publications funded per country.

Coatings 2019, 9, 384 12 of 22

Figure 4 shows that China supports research related to the reuse of reclaimed rubber with almost 50% of the publications. Europe and the United States are far behind, with approximately the same percentage of publications that received funding (16.4% for Europe and 15.9% for the United States, respectively). Note that the results are not exhaustive because in certain regions, academia does not work parallel to the industry. Therefore, there might be some interests in the industry that are not reflected in the number of publications.

The main aspect arising from the analysis of Figures 3 and 4 is that despite the main idea for recycling tire rubber in infrastructure construction being born in the United States in the 1960s and that the main products were patented and commercialized there, China in the last ten years was the undisputed leader of scientific effort and funding dedicated to tire rubber recycling. The publications funded comprise almost half of the entire number of publications, a percentage much higher than the publications that received funding from the United States and Europe. This is probably due to the fact that recycling ELTs is more of a concern considering that the number of Chinese car parks is expected to almost double by 2024 [10].

Figure 5 shows the most common characteristics of the input materials and the fabrication parameters for CRM bitumen.



**Figure 5.** Most common types and percentages of (a) rubber content, (b) maximum size, (c) base bitumen penetration, (d) mixing time, (e) temperature, and (f) shear mix.

Coatings 2019, 9, 384 13 of 22

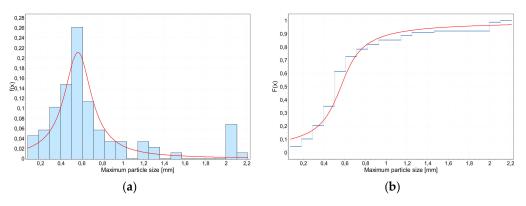
From Figure 5 it is possible to draw certain conclusions:

• The most-used quantity of rubber falls in the interval between 15% and 20%, and a significant number of research works continue using a percentage between 5% and 20% (Figure 5a). Only a few attempts have been made with higher quantities (20%–35%). This means that the "safety point" of rubber used in the modified binder has a threshold of 20%. In the framework of this paper, "safety point" is related to the most-used quantity of rubber for research purposes. Higher quantities of rubber make the binder stiff and inappropriate for road applications. Therefore, higher rubber quantities are still not used.

- Almost 50% of the maximum size of the rubber particles used to modify the binder fall in the range of 0.5–1 mm (Figure 5b). A significant percentage can be observed for the range of 0.25–0.5 mm. The percentage of bigger particles is higher than the percentage of smaller particles  $(d_{\text{max}} < 0.25 \text{ mm})$ . This is probably due to the difficulties in producing smaller rubber particles at the industrial level. Smaller particles facilitate degradation into the modified binder, nevertheless, the production of very fine particles is onerous and expensive. Therefore, the research has still focused on the use of bigger particles, eventually with the addition of additives (polymers and nanomaterials) for improving the binder properties [50,75,76]. See also Figure 6.
- The base bitumen is a standard bitumen with penetration of 50–70 dmm. Less used is the 70–100 dmm, while extremely stiff or soft bitumen are rarely used for this application (Figure 5c). This is probably due to the fact that stiffer or softer binder are less used for the traditional bituminous mixtures, therefore they are also less used for road applications with alternative materials.
- From analyzing the fabrication parameters (Figure 5d–f), it is possible to observe that the most-used mixing time and temperature are, respectively, 60 min and 180 °C (parameters corresponding to the McDonald production process). The most common shear mix adopted to produce the modified binder is between 1000 and 2000 rpm and 4000 and 5000 rpm.

Despite the entry of different products to the market, the most common parameters used for the fabrication process show that the McDonald process is still predominant.

Figure 6 shows the frequency distribution of the CR particle size used for the binder modification.



**Figure 6.** (a) Probability density function and (b) cumulative distribution function of the rubber particle size.

The frequency distribution of rubber particles is properly described with a Cauchy distribution ( $\mu$  = 0.563,  $\sigma$  = 0.161), which shows that the average rubber particle size used for modifying the binder is 0.56 mm, with a small standard deviation (Figure 6a). This means that there is an industrial preference in producing crumb rubber of a certain dimension. Looking at Figure 6b, it is possible to see that the 80% of the rubber particles used are below 0.8 mm diameter size, nevertheless only the 25% are below 0.4 mm diameter size. This can confirm that industries prefer producing medium-size rubber particles, while the production of smaller particles is more energy and cost demanding.

Coatings 2019, 9, 384 14 of 22

Figure 7 shows the low and high temperatures of the Performance Grade (PG) of the base bitumen used for the modification with CR.

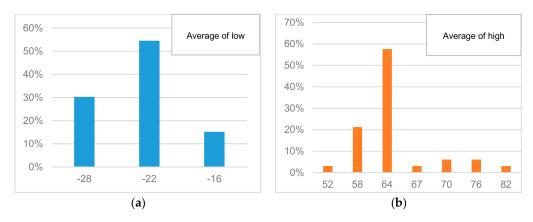


Figure 7. (a) Low and (b) high temperature PG of the bitumen base used for CR modification.

Figures 5c and 7 allow the conclusion to be drawn that the most-used base bitumen for the modification is a standard bitumen of 50/70 dmm penetration and PG64-22.

#### 6.2. Standard Properties, Rheology, and High- and Low-Temperature Properties

The following table (Table 2) provides the legend to read the results of this section. The acronyms are used in the framework of this paper to identify the different technologies. Each acronym has been defined and the corresponding reference listed in Table 2.

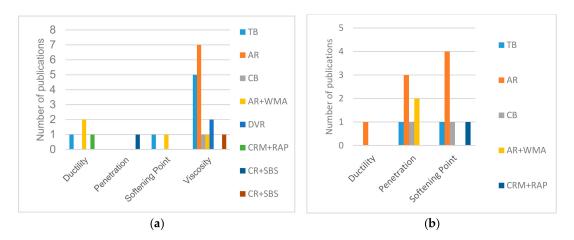
**Table 2.** Principal acronyms of the modification process and technologies found in the literature that are useful for a better understanding of the results.

Acronym	Description	Principal References
Traditional	Neat binder before modification	_
ТВ	Terminal Blend	[49,67,77–80]
AR	Asphalt Rubber (McDonald process)	[64,67,75,79–83]
СВ	Continuous Blend	[77]
AR + WMA	Asphalt Rubber with the further addition of warm mix additives	[42,64,67,84]
DVR	Devulcanized rubber modified binder	[41,75]
GTR or CR	Ground Tire Rubber modified binder or Crumb Rubber	[31,61,85]
CRM + RAP	Binder blend composed of crumb rubber modified bitumen and aged binder extracted and recovered from Reclaimed Asphalt Pavement (RAP)	[85]
CR + SBS	Crumb rubber modified binder with the further addition of STirene Butadiene STirene (SBS)	[83,86]
RAP	Aged binder extracted and recovered from Reclaimed Asphalt Pavement (RAP)	[87]
CR + SBS + sulfur	SBS + sulfur Crumb rubber modified binder with the further addition of SBS and sulfur	
TB + SBS	Terminal Blend modification of the binder with the further addition of SBS	[50,78,88]
TB + PPA	Terminal Blend modification of the binder with the further addition of polyphosphoric acid	[50]

Figures 8–10 show the comparison between different CRM binders (TB, AR, CB, AR + WMA, DVR, CRM + RAP, RAP, CR + SBS, and CR + SBS + sulfur) and the traditional binder for certain standard properties.

Coatings 2019, 9, 384 15 of 22

From Figure 8 it is possible to observe the comparison between different CRM binders with the traditional binder before the modification for different standard properties: viscosity, ductility, softening point and penetration. The results in Figure 8a are to be understood as follows: the histograms represent the number of publications where the results report a higher value for the analyzed characteristic of different CRM binders compared to the traditional binder. Figure 8b, on the other hand, shows the number of the publications reporting a lower value of the considered characteristics for CRM binders compared to the traditional binder.



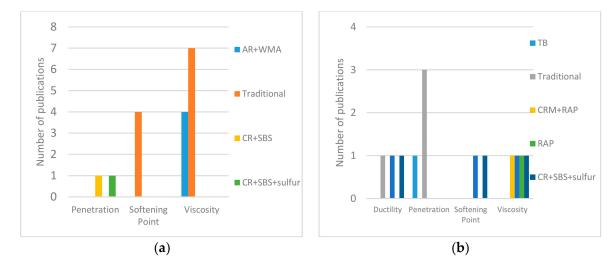
**Figure 8.** (a) Number of publications where the results report a higher value for the analyzed characteristic of different CRM binders compared to the traditional binder, and (b) number of publications reporting a lower value for the considered characteristics for CRM binders compared to the traditional binder. Note that in Figure 8b, the viscosity is not present because in all the results of the papers analyzed, the addition of the rubber always increases the viscosity of the binder.

Figure 8 shows that whatever addition of rubber occurs, with Terminal Blend procedure, Asphalt rubber, and also adding other warm mix additives or SBS to the CRM binder, each modification leads to an increase in the viscosity. In the case of the other standard properties, the situation is more varied. The addition of rubber generally decreases the softening point of the binder, while when a further addition is made, such as a warm mix additive, the softening point increases compared to the traditional binder. The penetration shows a decrease; therefore, the addition of the rubber makes the binder stiffer. Less information is available for the ductility, nevertheless, in the case of AR, the ductility decreases compared to the traditional binder (in one publication), while as soon as the further addition of warm mix additive is considered, the ductility becomes higher compared to the neat binder. It is interesting to note that the majority of the studies, by counting the number of publications, have been conducted on Asphalt Rubber technology. Few studies are available for the evaluation of the effect of the addition of SBS and WMA.

Figure 9 provides the comparison of different CRM binders with the AR.

As for Figure 8, the results shown in Figure 9 have to be understood as follows. The histograms represent the number of publications where the results report a higher value of the analyzed characteristic of AR binders compared to other types of modifications (Figure 9a). Figure 9b, on the other hand, shows the number of publications reporting a lower value of the considered characteristics for AR binders compared to other types of modifications (TB, traditional, CRM + RAP, RAP, CR + SBS + sulfur).

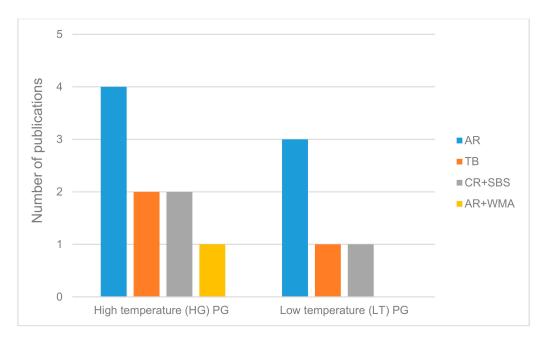
Coatings 2019, 9, 384 16 of 22



**Figure 9.** (a) Number of publications where the results report a higher value of the analyzed characteristic of AR compared to other technologies (AR+WMA, Traditional, CR+SBS, CR + SBS + sulfur), and (b) number of the publications reporting a lower value of the considered characteristics for AR binder compared to the other technologies (TB, traditional, CRM+RAP, RAP, CR + SBS + sulfur).

Figure 9 shows the results of the comparison between AR and other types of CR modifications. According to Figure 9, the ductility of Asphalt Rubber is lower than traditional blend and other blends with the further addition of polymers. The penetration is higher if compared to the same blend with the addition of other polymers, while the softening point is lower. The viscosity is lower than binder further modified with SBS, sulfur, or RAP binder. Instead, when the WMA is added to AR the viscosity decreases.

Figure 10 shows the comparison between different CRM modified binders and the traditional binder before modification for low and high-temperature performance properties (low PG temperature (LT) and high PG temperature (HT)).



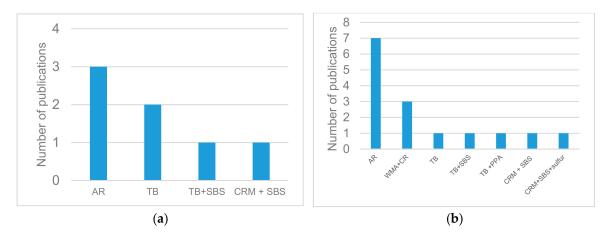
**Figure 10.** Number of publications where the results report a higher value of the analyzed characteristic of CRM binders (AR, TB, CR + SBS and AR + WMA) compared to the traditional binder.

Coatings 2019, 9, 384 17 of 22

The results in Figure 10 show that whatever addition of rubber occurs, with Terminal Blend procedure, Asphalt Rubber, and also using other warm mix additives or SBS, the modification of the binder leads to an extension of the range of the suitable temperatures (PG) of the bitumen. Only in one case it has been found that the high temperature (HG) PG of CRM modified binder decreased in comparison with the neat binder [50]. The authors showed that the PG grades of TB binders decreased with the increase of crumb rubber percentage (5%, 10%, 15%, and 20%) and the PG grades that resulted were lower than that of the traditional binder [50].

Aside from the consideration of the performance, it is interesting noting that the majority of the studies, counting the number of publications, have been conducted on high-temperature properties rather than low-temperature properties. This is probably due to the fact that the high-temperature properties are the cutting edge of the benefits arising from the use of CR and the addition of other additives.

Figure 11 compares the rheological properties (complex modulus and phase angle) of different CRM binders and the traditional binder before its modification.



**Figure 11.** Comparison of rheological properties: (a) the number of studies reporting a higher value of the complex modulus for each CRM binder compared to traditional binder; and (b) the number of studies reporting a lower value of the phase angle for each CRM binder compared to the traditional binder.

The results in Figure 11 show that whatever addition of rubber occurs, with Terminal Blend procedure, Asphalt Rubber, and also using other warm mix additives or SBS, the modification of the binder leads to an increase of the complex modulus and decrease of the phase angle. Therefore, the addition of the rubber, in general, produces a stiffer binder. Phase angle is defined as the time lag between strain and stress under the traffic loading [75]. A smaller phase angle indicates that the asphalt binder is less viscous, which combined with a high complex modulus, enhances the rutting resistance of the binder. This does not seem to be the case for the Terminal Blend, where the rubber is in a state that does not increase the stiffness of the binder (lower complex modulus compared to the traditional reference binder).

Again, the majority of the studies have been conducted on Asphalt Rubber.

#### 7. Conclusions

This work has the intent of providing an overview of the principal processes and technologies for the use of crumb rubber in the asphalt mixture. The paper has two main objectives:

- To illustrate the current position, the direction, and rate of progress of the scientific efforts towards the reuse and recycling of tire rubber worldwide;
- To promote an in-depth analysis of a set of important properties of CRM binders: fabrication parameters, standard properties, high- and low-temperature performance, and rheological properties.

As a result of both analyses, certain main conclusions can be drawn:

Coatings 2019, 9, 384 18 of 22

2007 marks a turning point in the research efforts undertaken for studying the CR application
in civil engineering, evidenced by the exponential increase in the number of publications dedicated
to the use of CR. One of the reasons is that the policies, and consequently the markets, believe
in the feasibility of the use of reclaimed rubber in engineering construction and intensively support
the research in this direction;

- China in the last ten years was the undisputed leader of the scientific effort and funding dedicated to tire rubber recycling. This is probably due to the fact that the number of car parks in China is expected to almost double by 2024, and the necessity of recycling the EOL will become even more important.
- Regarding the statistics for the fabrication parameters, the conclusions are:
- The "safety point" of rubber used in the modified binder has a threshold of 20% of the weight of the binder;
- The average rubber particle size is 0.56 mm, with a small dispersion around this value. Only a small percentage of small particles is used, probably because the production of very fine particles is onerous and expensive;
- The bitumen used for the modification is normally a standard bitumen with 50/70 penetration and PG64-22;
- The McDonald process is still predominant in the research, despite the entry of other products on the market.

In general, few studies are available for the comparison between different technologies of rubber addition in the binder. More often, the comparison is conducted between the CRM binder and the traditional one before the modification. Therefore, the following conclusions regarding the standard properties, the low- and high-temperature properties, and the rheological properties can be drawn:

- Whatever addition of rubber occurs, with Terminal Blend procedure, Asphalt rubber, and also
  using other warm mix additives or SBS, the modification of the binder leads to an increase of the
  viscosity of the binder;
- The addition of rubber decreases the softening point and the ductility, while when a further
  addition is made, such as SBS or warm mix additives, these two values increase compared to the
  traditional binder. In general, the further addition of SBS or warm mix additives significantly
  alters the properties of the CR binder;
- Whatever addition of rubber occurs, with Terminal Blend procedure, Asphalt rubber, and also
  using other warm mix additives or SBS, the modification of the binder leads to the Performance
  Grade extension.
- The addition of the rubber, in general, produces significant effects on the rheological properties of CRM binders. The binder becomes stiffer (higher complex modulus compared to the traditional binder) and less viscous (lower phase angle compared to the traditional binder), indicating higher rutting resistance;
- When the rubber is depolymerized or devulcanized, the effect on the CRM binder properties is reduced because the rubber is evenly dispersed, but loses its elasticity.

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**Conflicts of Interest:** The authors declare that there is no conflict of interest regarding the publication of this paper.

Coatings 2019, 9, 384 19 of 22

#### References

 European Union Road Federation (ERF). ERF 2012 European Road Statistics; European Union Road Federation: Brussels, Belgium, 2012; Available online: https://erf.be/publications/european-road-statistics-2012 (accessed on 13 June 2019).

- 2. Santero, N.; Horvath, A. Global warming potential of pavements. *Environ. Res. Lett.* **2009**, *4*, 034001. [CrossRef]
- 3. European Road Transport Research Advisory Council (ERTRAC). ERTRAC Strategic Research Agenda 2010: Towards a 50% More Efficient Road Transport System by 2030. Available online: https://www.ertrac.org/uploads/documentsearch/id21/ERTRAC\_SRA\_2010.pdf (accessed on 13 June 2013).
- 4. Santos, J.; Bressi, S.; Cerezo, V.; Lo Presti, D. SUP&R DST: Sustainable Pavement & Railways Decision Support Tool. In Proceedings of the Sixth International Symposium on Life-Cycle Civil Engineering, Ghent, Belgium, 28–31 October 2018.
- 5. Palit, S.K.; Reddy, K.S.; Pandey, B.B. Laboratory evaluation of crumb rubber modified asphalt mixes. *J. Mater. Civ. Eng.* **2004**, *16*, 45–53. [CrossRef]
- 6. Wong, C.C.; Wong, W.G. Effect of crumb rubber modifiers on high temperature susceptibility of wearing course mixtures. *Constr. Build. Mater.* **2017**, *21*, 1741–1745. [CrossRef]
- 7. Moreno, F.; Rubio, M.; Martinez-Echevarria, M. Analysis of digestion time and the crumb rubber percentage in dry-process crumb rubber modified hot bituminous mixes. *Constr. Build. Mater.* **2001**, 25, 2323–2334. [CrossRef]
- 8. Bahia, H.U.; Davies, R. Effect of crumb rubber modifiers (CRM) on performance related properties of asphalt binders. *Asph. Paving Technol.* **1994**, *63*, 414.
- 9. Oyuang, C.; Gao, Q.; Shi, Y.; Shan, X. Compatibilizer in waste tire powder and low-density polyethylene blends and the blends modified asphalt. *J. Appl. Polym. Sci.* **2012**, *123*, 485–492. [CrossRef]
- 10. European Tire & Rubber Manufacturer's Association (ETRMA). *The ETRMA Statitics Report*; European Tire & Rubber Manufacturer's Association: Saint-Josse-ten-Noode, Belgium, 2017.
- 11. Han, L.; Zheng, M.; Wang, C. Current status and development of terminal blend Tire rubber modified asphalt. *Constr. Build. Mater.* **2016**, *128*, 399–409. [CrossRef]
- 12. Hanson, D.I.; Foo, K.Y.; Brown, E.R.; Denson, R. Evaluation and characterization of a rubber-modified hot mix asphalt pavement. *Transp. Res. Rec. J. Trans. Res. Board* **1994**, *1436*, 98–107.
- 13. Yildrim, Y. Polymer modified asphalt binder. J. Build. Constr. Mater. 2007, 21, 66–72. [CrossRef]
- 14. Epps, J.A. Uses of Recycled Rubber Tires in Highways; National Academic Press: Washington, DC, USA, 1994.
- 15. McDonald, C.H. Recollections of Early Asphalt-Rubber History. In *National Seminar on Asphalt-Rubber*; US Department of Transportation: Washington, DC, USA, 1981.
- 16. Lo Presti, D. Recycled Tire rubber modified bitumens for road asphalt mixtures: A literature review. *Constr. Build. Mater.* **2013**, *49*, 863–881. [CrossRef]
- 17. Federal Highway Administration (FHWA). *User Guidelines for Waste and Byproduct Materials in Pavement Construction*; Federal Highway Administration: Washington, DC, USA, 1997.
- 18. Heitzman, M. Design and Construction of Asphalt Paving Materials with Crumb Rubber Modifier. *Transp. Res. Record.* **1992**, 1339, 1–8.
- 19. CALTRANS California Department of Transportation, Materials Engineering and Testing Services. *Asphalt Rubber Usage Guide*; Rubber Pavements Association: Sacramento, CA, USA, 2006.
- 20. Technical Assistance and Training. *A Basic Introduction to RAC Usage (RAC-101)*; CalRecycle California's Department of Resources Recycling and Recovery (CalRecycle): Sacramento, CA, USA, 2010.
- 21. Kandhal, P.; Hanson, D. Crumb Rubber Modifier (CRM) Technologies, in Crumb Rubber Modify Work. Available online: https://www.fhwa.dot.gov/pavement/pubs/013377.pdf (accessed on 13 June 2019).
- 22. Federal Highway Administration (FHWA). *A Study of the Use of Recycled Paving Materials-Report to Congress;* US Department of Transportation: Washington, DC, USA, 1993.
- 23. Takallou, H.; McQuillen, I.; Hicks, R.O. *Effect of Mix Ingredients on Performance of Rubber-Modified Asphalt Mixtures*; Transportation Research Institute: Fairbanks, AK, USA, 1985.
- 24. Heitzman, M.A. State of the Practice for the Design and Construction of Asphalt Paving Materials with Crumb Rubber Additive. Report No. FHWA-SA-92-022; Office of Engineering; Pavement Division, Federal Highway Administration: Washington, DC, USA, 1992.

Coatings 2019, 9, 384 20 of 22

25. Peralta, J.; Silva, H.; Machado, A.V.; Pais, J.; Pereira, P.; Sousa, J. Changes in Rubber Due to its Interaction with Bitumen when Producing Asphalt Rubber. *Road Mater. Pavement Des.* **2010**, *11*, 1009–1031. [CrossRef]

- 26. Evaluation of Waste Tire Devulcanization Technologies; CalRecovery Integrated Waste Management Board: Sacramento, CA, USA, 2004.
- 27. Rafique, R.M.U. Life cycle assessment of waste car tires at scandinavian enviro systems. Master Thesis, Chalmers University of Technology, Göteborg, Sweden, December 2012.
- 28. Rader, C.; Baldwin, S.; Cornell, D.; Sadler, G.; Stockel, R. *Plastics, Rubber, and Paper Recycling*; American Chemical Society: Washington, DC, USA, 1995.
- 29. Fukumori, K.; Matsushita, M. Material Recycling Technology of Crosslinked Rubber Waste. *Rev. Toyota* **2003**, 38, 39–47.
- 30. Mangili, I.; Lasagni, M.; Anzano, M.; Collina, E.; Tatangelo, V.; Franzetti, A.; Caracino, P.; Isayev, A.I. Mechanical and rheological properties of natural rubber compounds containing devulcanized ground tire rubber. *Polym. Degrad. Stab.* **2015**, *121*, 369–377. [CrossRef]
- 31. ChemRisk(LLC). *Tire Generic Exposure Scenario End of Life Tire Guidance*; ChemRisk LLC: Pittsburgh, PA, USA, 2009.
- 32. Giavarini, C. Polymer-Modified Bitumen. Asph. Asph. 1994, 1, 381–400.
- 33. Gawel, I.; Stepkowski, R.; Czechowski, F. Molecular interactions between rubber and asphalt. *Ind. Eng. Chem. Res.* **2006**, *4*, 3044–3049. [CrossRef]
- 34. Morrison, G.R.; Van Der Stel, R.; Hesp, S.A.M. Modification of asphalt binders and asphalt concrete mixes with crumb and chemically devulcanized waste rubber. *Transp. Res. Record* **1995**, *1515*, 56–63.
- 35. Navarro, F.J.P.; Partal, F.; Mart'ınez-Boza, C.; Valencia, C.; Gallegos, C. Rheological characteristics of ground tire rubber-modified bitumens. *Chem. Eng. J.* **2002**, *89*, 53–61. [CrossRef]
- 36. Rajan, V.V.; Dierkes, W.K.; Joseph, R.; Noordermeer, J.W.M. Science and technology of rubber reclamation with special attention to NR-based waste latex products. *Prog. Poly. Sci.* **2006**, *31*, 811–834. [CrossRef]
- 37. Adhikari, B.; De, D.; Maiti, S. Reclamation and recycling of waste rubber. *Prog. Polym. Sci.* **2000**, 2, 909–948. [CrossRef]
- 38. Isayev, A.I. Recycling of Rubber. In *The Science and Technology of Rubber*; Mark, J., Erman, B., Roland, M., Eds.; Academic Press: Cambridge, MA, USA, 2013; pp. 697–764.
- 39. Wang, S.; Cheng, D.; Xiao, F. Recent developments in the application of chemical approaches to rubberized asphalt. *Constr. Build. Mater.* **2017**, *131*, 101–113. [CrossRef]
- 40. Dong, R.; Li, J.; Wang, S. Laboratory evaluation of pre-devulcanized crumb rubber—Modified asphalt as a binder in hot-mix asphalt. *J. Mater. Civ. Eng.* **2011**, 23, 1138–1144. [CrossRef]
- 41. Ghasemirad, A.; Asgharzadeh, S.M.; Tabatabaee, N. A comparative evaluation of crumb rubber and devulcanized rubber modified binders. *Pet. Sci. Technol.* **2017**, *35*, 1091–1096. [CrossRef]
- 42. Yu, G.; Li, Z.; Zhou, X.; Li, C. Crumb rubber–modified asphalt: Microwave treatment effects. *Pet. Sci. Technol.* **2011**, 29, 411–417. [CrossRef]
- 43. Billiter, T.C.; Chun, J.S.; Davison, R.R.; Glover, C.J.; Bullin, J.A. Investigation of the curing variables of asphalt-rubber binder. *Pet. Sci. Technol.* **1997**, *15*, 445. [CrossRef]
- 44. *Manual 19. Guidelines for the Design, Manufacture and Construction of Bitumen Rubber Asphalt Wearing Courses;* SABITA: Howard Place, South Africa, 2009.
- 45. Specification Framework for Polymer Modified Binders and Multigrade Bitumens; Austroads Technical Report APT41/06; Austroads: Sydney, Australia, 2006.
- 46. Mturi, G.A.; O'Connell, J.; Zoorob, S.E.; De Beer, M. A study of crumb rubber modified bitumen used in South Africa. *Road Mater. Pavement Des.* **2014**, *15*, 774–790. [CrossRef]
- 47. Lo Presti, D.; Airey, G.; Partal, P. Manufacturing terminal and field bitumen-Tire rubber blends: The importance of processing conditions. In Proceedings of the SIIV—5th International Congress—Sustainability of Road infrastructures, Rome, Italy, 29–31 October 2012.
- 48. PCCAS. Pacific Coast Conference for Asphalt Specifications Minutes; PCCAS: Portland, OR, USA, 2012.
- 49. Wu, C.; Liu, K.; Tang, J.; Li, A. Research on the terminal blend rubberized asphalt with high-volume of rubber crumbs and its gap graded mixture. *Munich Asph. Rubber* **2012**.
- 50. Lin, P.; Huang, W.; Tang, N.; Xiao, F. Performance characteristics of Terminal Blend rubberized asphalt with SBS and polyphosphoric acid. *Constr. Build. Mater.* **2017**, *141*, 171–182. [CrossRef]

Coatings **2019**, 9, 384 21 of 22

51. Nguyen, H.T.; Tran, T. Effects of crumb rubber content and curing time on the properties of asphalt concrete and stone mastic asphalt using dry process. *Int. J. Pavement Res. Technol.* **2018**, *11*, 236–244. [CrossRef]

- 52. Hossain, M.; Sadeq, M.; Funk, L.; Maag, R. A study of chunk rubber from recycled tires as a road construction material. In Proceedings of the 10th Annual Conference on Hazardous Waste Research, New York, NY, USA, 23–24 May 1995.
- 53. Waller, F. Use of Waste Materials in Hot-mix Asphalt; ASTM Publications: Philadelphia, PA, USA, 1993.
- 54. Roberts, F.L.; Kandhal, P.S.; Brown, E.R. Investigation and evaluation of ground tire rubber in hot mix asphalt. *NCAT Rep.* **1989**, *89*, 3.
- 55. Paje, S.E.; Bueno, M.; Terán, F.; Miró, R.; Pérez-Jiménez, F.; Martínez, A. Acoustic field evaluation of asphalt mixtures with crumb rubber. *Appl. Acoust.* **2009**, *71*, 578–582. [CrossRef]
- 56. Paje, S.; Luong, J.; Vázquez, V.F.; Bueno, M.; Mirò, R. Road pavement rehabilitation using a binder with a high content of crumb rubber: Influence on noise reduction. *Constr. Build. Mater.* **2013**, *47*, 789–798. [CrossRef]
- 57. Zhang, X.; Lu, Z.; Tian, D.; Li, H.; Lu, C. Mechanochemical devulcanization of ground tire rubber and its application in acoustic absorbent polyurethane foamed composites. *J. Appl. Polym. Sci.* **2013**, 2013 127, 4006–4014. [CrossRef]
- 58. Liang, T. Continuous Devulcanization of Ground Tire Rubber of Different Particle Sizes Using an Ultrasonic Twin-Screw Extruder. Ph.D. Thesis, The University of Akron, Akron, OH, USA, 2013.
- 59. Buncher, M. Evaluating the Effects of the Wet and Dry Processes for Including Crumb Rubber Modifier in Hot Mix Asphalt. Ph.D. Thesis, National Center of Asphalt Technology; Auburn University, Auburn, AL, USA, 1995.
- 60. Kirk, J.; Holleran, G. Reduced Thickness Asphalt Rubber Concrete Leads to Cost Effective Pavement Rehabilitation. In Proceedings of the International Conference World of Pavements, Sydney, Australia, 20–24 February 2000.
- 61. Farina, A.; Zanetti, M.C.; Santagata, E.; Blengini, G.A. Life cycle assessment applied to bituminous mixtures containing recycled materials: Crumb rubber and reclaimed asphalt pavement. *Resour. Conserv. Recycl.* **2017**, 117, 204–212. [CrossRef]
- 62. Sienkiewicz, M.; Borzędowska-Labuda, K.; Wojtkiewicz, A.; Janik, H. Development of methods improving storage stability of bitumen modified with ground tire rubber: A review. *Fuel Process. Technol.* **2017**, 159, 272–279. [CrossRef]
- 63. Navarro, F.J.; Partal, P.; Martínez-Boza, F.; Gallegos, C. Thermo-rheological behaviour and storage stability of ground tire rubber-modified bitumens. *Fuel* **2004**, *83*, 2041–2049. [CrossRef]
- 64. Bressi, S.; Santos, J.; Giunta, M.; Lo Presti, D. A comparative environmental assessment of asphalt mixtures for railway sub-ballast containing alternative materials. *Resour. Conserv. Recycl.* **2018**, 137, 76–88. [CrossRef]
- 65. Li, B.; Huang, W.; Tang, N.; Hu, J.; Lin, P.; Guan, W.; Xiao, F.; Shan, Z. Evolution of components distribution and its effect on low temperature properties of terminal blend rubberized asphalt binder. *Constr. Build. Mater.* **2017**, *136*, 598–608. [CrossRef]
- 66. Huang, Y.; Bird, R.N.; Heidrich, O. A review of the use of recycled solid waste materials in asphalt pavements. *Resour. Conserv. Recycl.* **2007**, *52*, 58–73. [CrossRef]
- 67. Fontes, L.; Pereira, P.; Pais, J.; Triches, G. Performance of wet process method alternatives: Terminal or continuous blend. In Proceedings of the Asphalt Rubber 2006, Palm Springs, CA, USA, 25–27 October 2006.
- 68. Hunt, E. *Crumb Rubber Modified Asphalt Concrete in Oregon;* No. FHWA-OR-RD-02-13; United States Department of Transportation: Washington, DC, USA, 2002.
- 69. Hernandez-Olivares, F.; Witoszek-Schultz, B.; Alonso-Fernandez, M.; Benito-Moro, C. Rubber-modified hot-mix asphalt pavement by dry process. *Int. J. Pavement Eng.* **2009**, *10*, 277–288. [CrossRef]
- 70. Dong, D.; Huang, X.; Li, X.; Zhang, L. Swelling process of rubber in asphalt and its effect on the structure and properties of rubber and asphalt. *Constr. Build. Mater.* **2012**, *29*, 316–322. [CrossRef]
- 71. Dantas Neto Silvrano, A.; de Farias, M.M.; Luiz Guilherme, R.; Pereira, M.P.; Pais, J.C. The use of crumb rubber in asphalt mixtures using the dry process. In Proceedings of the 2005 International Symposium on Pavement Recycling, São Paulo, Brazil, 14–16 March 2005.
- 72. Moreno, F.; Rubio, M.; Martinez-Echevarria, M.J. The mechanical performance of dry-process crumb rubber modified hot bituminous mixes: The influence of digestion time and crumb rubber percentage. *Constr. Build. Mater.* **2012**, *26*, 466–474. [CrossRef]
- 73. Amirkhanian, S. *Utilization of Crumb Rubber in Asphaltic Concrete Mixtures—South Carolina's Experience*; South Carolina Department of Transportation: Columbia, SC, USA, 2001.

Coatings 2019, 9, 384 22 of 22

74. *The Use of Recycled Crumb Rubber*; APRG Technical Note 10; Austroads Pavement Research Group: Sydney, Australia, 1999.

- 75. Yu, H.; Leng, Z.; Zhou, Z.; Shih, K.; Xiao, F.; Gao, Z. Optimization of preparation procedure of liquid warm mix additive modified asphalt rubber. *J. Clean. Prod.* **2017**, *141*, 336–345. [CrossRef]
- 76. Ge, D.; Yan, K.; You, Z.; Xu, H. Modification mechanism of asphalt binder with waste tire rubber and recycled polyethylene. *Constr. Build. Mater.* **2016**, *126*, 66–76. [CrossRef]
- 77. Fontes, L.; Trichês, G.; Pais, J.; Pereira, P. Evaluating permanent deformation in asphalt rubber mixtures. *Constr. Build. Mater.* **2010**, 24, 1193–1200. [CrossRef]
- 78. Attia, M.; Abdelrahman, M. Enhancing the performance of crumb rubber-modified binders through varying the interaction conditions. *Int. J. Pavement Eng.* **2009**, *10*, 423–434. [CrossRef]
- 79. Thives, L.; Pais, J.; Pereira, P.; Trichês, G.; Amorim, S. Assessment of the digestion time of asphalt rubber binder based on microscopy analysis. *Constr. Build. Mater.* **2013**, *47*, 431–440. [CrossRef]
- 80. Xu, O.; Xiao, F.; Han, S.; Amirkhanian, S.N.; Wang, Z. High temperature rheological properties of crumb rubber modified asphalt binders with various modifiers. *Constr. Build. Mater.* **2016**, *112*, 49–58. [CrossRef]
- 81. Mashaan, N.; Hassan Ali, A.; Rehan Karim, M.; Abdelaziz, M. An overview of crumb rubber modified asphalt. *Int. J. Phys Sci.* **2012**, *72*, 166–170.
- 82. Airey, G.; Rahman, M.; Collop, A. Absorption of bitumen into crumb rubber using the basket drainage method. *Int. J. Pavement Eng.* **2003**, *4*, 105–119. [CrossRef]
- 83. Kök, B.; Çolak, H. Laboratory comparison of the crumb-rubber and SBS modified bitumen and hot mix asphalt. *Constr. Build. Mater.* **2011**, *25*, 3204–3212. [CrossRef]
- 84. Leng, Z.; Yu, H.; Zhang, Z.; Tan, Z. Optimizing the mixing procedure of warm asphalt rubber with wax-based additives through mechanism investigation and performance characterization. *Constr. Build. Mater.* **2017**, 141, 291–299. [CrossRef]
- 85. Akisetty, C.; Xiao, F.; Gandhi, T.; Amirkhanian, S. Estimating correlations between rheological and engineering properties of rubberized asphalt concrete mixtures containing warm mix asphalt additive. *Constr. Build. Mater.* **2011**, 25, 950–956. [CrossRef]
- 86. Mashaan, N.; Karim, M. Investigating the rheological properties of crumb rubber modified bitumen and its correlation with temperature susceptibility. *Mater. Res.* **2013**, *16*, 116–127. [CrossRef]
- 87. Zhang, F.; Hu, C. The research for structural characteristics and modification mechanism of crumb rubber compound modified asphalts. *Constr. Build. Mater.* **2015**, *76*, 330–342. [CrossRef]
- 88. Singh, D.; Sawant, D.; Xiao, F. High and intermediate temperature performance evaluation of crumb rubber modified binders with RAP. *Transp. Geotech.* **2017**, *10*, 13–21. [CrossRef]



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