Rubber, as elastomer, is difficult to recycle. Today, the main end of life routes of tyres and other rubber products are landfilling, incineration in e.g. cement plants, and grinding to a fine powder, with huge quantities lacking sustainable recycling of this valuable material. Devulcanization, i.e. the breaking up of sulfur bonds by chemical, thermo-physical or biological means, is a promising route that has been investigated for more than 50 years. This review article presents and update on the state-of-the art in rubber devulcanization. This review article addresses established devulcanization technologies and novel processes described in the scientific and patent literatures. It is expected that the public discussion of environmental impacts of thermoplastics will soon spill over to thermosets and elastomers. Therefore, the industry needs to develop and market solutions proactively. Tyre recycling through devulcanization has a huge lever, since approx. 40 million tons of tyres are discarded annually.

With increasing global populations and welfare, consumption has been surging. Polymers—thermoplastics, thermosets, and elastomers—have shown significant growth over more than six decades from the 1950s onwards, with thermoplastics being by far the largest group. In 2018, the production volume has approached 350 million tons. The steady, historic growth rate of 6% per year is expected to flatten considerably in the coming years due to a pressure toward recycling plastics materials. Plastics Europe and other associations have shifted their focus of communication from job and value creation of the industry toward recycling and littering prevention; the circular economy, sustainability, microplastics pollution, and prevention have become common concerns, which the industry is starting to address seriously. Despite the huge efforts put into the recycling of thermoplastics, the achievements have been rather disappointing, apart from selected successful recycling schemes such as PET (polyethylene terephthalate) with bottles of carbonated soft drinks. “Thermal recycling” sounds nice; however, it should only be considered as the last step of a cascaded use, since the incineration to recapture energy is adding little value. Composite materials such as GFRP and CFRP (glass fiber-reinforced plastics and carbon fiber-reinforced plastics) make recycling extremely difficult as well as the variety of applications of plastics and various contaminations such as foodstuffs. PET bottles can be collected and recycled efficiently and effectively, because carbonated soft drinks and bottled water are put almost exclusively into PET containers. Packaging film, on the other hand, is often a multilayer material that is used particularly for perishable food, where recycling becomes virtually impossible. The low value of plastics, compared to other materials, makes recycling challenging, too. Plastics Europe, in one of their recent reports, claims that within the EU28 (Belgium, Bulgaria, Czech Republic, Denmark, Germany, Estonia, Ireland, Greece, Spain, France, Croatia, Italy, Cyprus, Latvia, Lithuania, Luxembourg, Hungary, Malta, Netherlands, Austria, Poland, Portugal, Romania, Slovenia, Slovakia, Finland, Sweden, United Kingdom), Norway, and Switzerland, in 2016, 31.1% of the 27.1 million tons of post-consumer waste collected plastics were recycled, of which 63% were inside the EU, and another 41.6% were sent to energy recovery, with 27.3% remaining for landfilling (the landfilling ban in the EU came into force in 1999). These numbers are misleading, because the total demand was in excess of 50 million tons, and the absolute recycling rates, although they are increasing from year to year driven by landfill restrictions for organic materials, are disappointingly low. Recycled thermoplastics go different routes. Production scrap is recycled most easily; typically, 10%-15% of own material (e.g., sprues in injection molding) can be shredded and added without quality issues. Post-consumer recycled plastics can go into products of lower mechanical properties. Prices of recycled polyolefins, due to consumers’ demand for “green” products, have increased sharply in the last years. Another promising route are bioplastics, which can either be based on renewable raw materials and/or be biodegradable. Currently, their market share is on the order of 1%-2% of global plastics consumption. For polymers (thermoplastics), there are typically two recycling methods: mechanical and thermal (the latter being incineration for energy recovery). Garforth et al. have defined feedstock recycling as a process that “aims to convert waste
polymer into original monomers or other valuable chemicals. Synonyms for feedstock recycling are chemical recycling or tertiary recycling. The main issue was that the original monomers are hard to obtain and that rather a mix of different molecules results. Some authors even understand the production of low-value products such as carbon black as feedstock recycling.

In the case of tires, which are a complex product made from completely different raw materials such as steel, cord, natural and synthetic rubber, additives, etc., full feedstock recycling will not be feasible, i.e., obtaining the original constituents or monomers.

“Feedstock recycling” and “devulcanization” are two terms that are rather not to be used interchangeably, since the ambition is different. The expressions “depolymerization” or “molecular rearrangement” hit the meaning of devulcanization better.

True feedstock recycling can be considered the “holy grail” of plastics recycling in that the monomers are obtained from collected scrap, and then, they are captured and reused. However, this route has not yet been developed sufficiently, and many approaches are still at a low technology readiness level.

For thermosets, recycling as for thermoplastics is not feasible, because the polymer chains have been converted into a rigid network that cannot be dissolved or molten anymore. There are some attempts to e.g., burn off the polymer matrix to recycle fibers from composite materials, which in an energy-efficient process can make sense for high-value materials such as carbon fibers.

For elastomers, recycling options are strongly limited, too, because the polymer is also a network. Elastomers cannot be molten nor be dissolved. One of the huge volume applications of elastomers is tires, in which natural rubber is used next to a mix of synthetic rubbers. By vulcanization or curing, the properties of the natural rubber compounds are finalized (a low sulfur content on the order of 2% yields soft rubber, whereas more sulfur addition gives hard rubber). However, the biodegradability of the raw materials (mostly latex) is thereby lost. Tires are produced (and discarded) on the order of 40 million tons per year on a global basis, and they have become a huge environmental concern.

Whereas waste tire dumps are visible to the public and are of general concern, end-of-life options for tires include incineration in cement plants and grinding them to a fine powder for addition into asphalt or concrete, which are rarely discussed in the general public. The attrition of tires on the roads which leads to microplastics formation is studied and discussed even less, although it bears a strong environmental impact.

In the case of tires and rubber in general, feedstock recycling would be a very beneficial approach. For more than five decades, the devulcanization of rubber has been studied. Different technologies have been developed, and some of them have already made it to the market. This review article provides an update on the state-of-the-art in rubber devulcanization with an outlook on potential future developments.

The literature bears a wealth of information on rubber devulcanization, which can be achieved by thermal, thermochemical, mechanical, and biological means. The process as such has a good environmental performance, since virgin materials and energy are conserved. In addition, it can bring about significant cost savings.

Reuse is better than recycling, and a material recycling path is to be preferred over feedstock recycling due to the lower energy requirements. Energy recovery should be the last step of a cascaded use model. Landfilling in general should be avoided. Although carbon is being sequestered, the burying of organic, reactive materials bears risks, and waste tire dump fires have been reported previously.

Apart from addressing the recycling of large volume rubber product streams such as tires, solutions need to be found to:
1. make raw material manufacturing (i.e., latex/natural rubber) more sustainable
2. make attrition to microplastics particles from tires less harmful, i.e., This might be achieved through suitable bioplastics materials.

Natural rubber today is mainly produced from the latex of the rubber tree or others. The rubber tree is grown in tropical areas, where plantations have often been established on previous rainforest land. Due to its nature to partially crystalize, natural rubber is harder than synthetic rubber, and it will give a longer lifetime to tires. This is also the reason while truck tires, which can run for well over 100,000 km, contain a larger fraction of natural rubber than do passenger car tires. Tire collection needs to be improved, and less environmentally friendly end-of-life options should be discontinued. There is a very strong, scientifically rooted interest in the feedstock recycling of rubber. On the one hand, this route provides a meaningful end-of-life exit for waste tires, and on the other hand, it conserves resources by reducing fresh natural and synthetic rubber demand. The circular economy concept is to be extended to elastomers, in which tires will play a crucial role.

**Keywords**
rubber;recycling;circular economy;devulcanization;environmental technology

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