

*Original Research*

# Life Cycle Perspective of Liquid Epoxy Resin Use in the Automotive Industry

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## Abstract

Epoxy resins are extensively used as part of the automotive industry, although they are not specifically monitored and recycled. The goal of this study was to summarize the uses and fields of the application of epoxy resins in the automotive industry and show possible ways for its further recovery or energy use at the end of life phase or waste management. To support a general overview of epoxy resins in the automotive industry, lifecycle assessment of these materials was performed and briefly presented. Epoxy resin synthesis contributes to the total carbon footprint of the automotive industry, representing an 84.5% share of the total carbon footprint of the product life cycle. Fossil fuel resource consumption occurs particularly during the phase of LER production.

**Keywords:** liquid epoxy resin, LCA, life cycle assessment, carbon footprint, chemical production

## Introduction

Composite components are frequently used in the automotive industry [1], construction industry [2-4], packaging industry [5-7] and for many technical applications for their practical properties [8-10]. Epoxy resin can positively alternate technical properties of various materials [11], but it also is problematic material in waste management [12-16]. As these materials can cause environmental burdens from their life cycle perspective, the life cycle assessment method is used for evaluation of their potential environmental impacts [17, 18]. European trends regarding waste management emphasize waste reduction, and primarily on decreasing its impacts on the environment. General regulation 2008/98/EC [19] set up a hierarchical regulation for the

handling of wastes. The priority is both to prevent its generation and to enhance recycling of it. In regulation 2000/53/EC [20], requirements are set out for vehicles regarding the recycling of their materials after having completed their operational period [21]. At the present time, the rate of materials reused amounts to 85% of the average vehicle's weight, and the remaining 15% must be stored at waste sites. From 2015 onwards, all vehicles having completed their operational period will be required to recycle and reuse at least 95% of the average vehicle weight. 10% can be used to provide energy, but only 5% of the average vehicle's weight may be stored in waste sites. Within a half year of bringing a new vehicle to market, the producer has to provide information regarding the dismantling and material contents of the vehicle for processing facilities, to an extent which enables them to fulfil these legislative goals. The aforementioned legislation requirements put increased demands on automotive designers and on the choice of materials used for vehicle construction. One

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of the materials used in the automotive, aviation and sailboat industries is epoxy resin [17, 22-24].

Reinforced epoxy resin is used in thermosets [25] in a number of applications [26-30]. These materials are very difficult to recycle [31-33], and research on recycling thermosets is still current [34]. In this paper the possibilities for epoxy resin usage in the automotive industry and its environmental consequences using life cycle assessment are analyzed. Information regarding the average amounts of epoxy resins in vehicles (in the low-mid and mid classes) were acquired in cooperation with the producers of Toyota, Peugeot, Citroën and Škoda Auto. Following the whole life cycle of epoxy resin use in automotive industry forms system boundaries in the study: acquisition of the raw materials, resin production, processing by the vehicle producer, use of the vehicle by the consumer, and dismantling of the vehicle within the waste management system after completion of its operational period. Application of waste materials is also promising in new technologies of LER production [35, 36] and application [37, 38].

Due to LER properties such as physic-chemical resistance, stability, and strength, it is broadly applied in industry engineering designs. Regarding the automotive industry, LER is mostly used as a component of the coating systems that primarily serve as anti-corrosion protection of the vehicle passenger body. Other functions of the coating materials include the smoothing-out and unifying of the surface of the vehicle body, and increasing the attractiveness of the vehicle.

One of the elements of the development strategies among automotive producers involves a focus on improvements in the field of vehicle efficiency, which can in one way be achieved by decreasing the total weight of the vehicle. This is connected with the demands for the use and development of new materials, instead of the conventional metals-based materials. These new materials cannot be welded as is customary – they need to be glued. Compared to welded parts, the surface of the passenger body is smooth, so there is no need for further treatment if LER is used. This brings about economic savings in the car production phase. Layers of glue act as an electrical insulator. Not having the contact of different metals in the construction eliminates the creation of electrolytic corrosion. Mastics are utilized for both sealing and noise insulation. These listed advantages also create requirements for the properties of the adhesives. Epoxy-based glues rank among the highest structural strength adhesives, having excellent shear resistance, which is used especially for the edges of doors and hoods.

## Material and Methods

### Epoxy Resin in the Automotive Industry

Epoxy compounds belong to an important group of polymers and are used in various branches of industry

[39]. The term liquid epoxy resin (hereafter LER) includes compounds containing more than one epoxy (oxiran) group per molecule. Those LERs with low molecular weights are viscous fluids. With growing molecular weight they become solid, fragile, and fusible. Non-reinforced LER has a thermoplastic character [28, 39, 40]. The majority of practical applications of LER is in the form of reinforced epoxy resin [41, 42].

Reinforcement is a process through which chemical reactions transform low-molecular, soluble, and fusible epoxy monomers and oligomers into infusible, insoluble, polymers; mostly having a three-dimensional reticulate structure [39]. After reinforcement, LER belongs to the group of thermosets. Reinforcement of LER can be produced by the polycondensation of hydroxyl groups, the polyaddition of compounds with an active hydrogen atom to the epoxy groups, and by the polymerization of the epoxy groups. During reinforcement, the molecular weight increases. The final properties of reinforced LER can be influenced by the choice of the curing agent. Curing agents with an aromatic ring impart into resins a higher heat resistance than do aliphatic curing agents [40]. After reinforcement, LER achieves its final properties of mechanical strength, dimensional stability, heat resistance, and cohesion, as well as moisture resistance, adhesion to various materials, and excellent electrical properties.

Due to the high stability of epoxy bonds, their chemical degradation is a complex and multistage process, being important, for example, in the construction industry, too [43, 44]. Resins are infusible and insoluble [45], and that causes problems in processing LER during waste management, as represented in product parts having terminated their life cycles [46, 47]. Once it is a part of a product, LER cannot be separated from the product without damaging it. Further research has shown that it is possible to synthesize degradable and decomposable LER, which can also be used as a mold for fiber-reinforced plastics (FRP) [48].

The high requirements upon the properties of composite materials also led to the development of highly functional LERs, which have higher numbers of epoxy groups in their molecules. They replaced the low-heat resistance of aliphatic bonds in the glycidic groups with stable bonds, and the development of curing agents [39].

At present, the most common type of LER products are based on the alkaline condensation of epichlorohydrin and bisphenol A, which produces a diglycidyl ether of bisphenol A (DGEBA). Over 85% of LER of the entire world's production is based on the synthesis of bisphenol A (and its derivatives) with epichlorohydrin [26, 49, 50] having significant environmental consequences [51-53].

LER has excellent physio-chemical and mechanical properties, and is thus used for various applications in varied production fields. In the automotive industry, LER is a component of coating materials, adhesives and glues, and poured materials. Lately, composite materials

containing LER are increasingly used as the mold for fiber-reinforced plastics (FRP).

### *Coating Materials*

An integral part of vehicle production is the external coating. Many different parts of a vehicle are treated with coating systems. The coating is primarily applied to prevent the corrosion of metallic parts, and thus prolonging the life cycle of the treated parts of the vehicle. The coating also improves the visual appearance, and thus increases the attractiveness of the vehicle. A varnished passenger vehicle body, as well as the use of other coating materials, will have to fulfil the following requirements: long-term protection against corrosion, weather, chemical effects (acid rain and other atmospheric pollutants), and solar radiation; as well as providing the best optical properties such as high gloss, homogeneity, and color consistency. The aforementioned requirements are fulfilled by coating systems that may consist of single or multiple layers. The coating materials consist of several compounds. The problematic compounds, from an environmental point of view, are those solvent agents that emit volatile organic compounds (VOCs). Lately, producers prefer water-soluble polishing materials, mostly for reasons related to legislation. Specification 2004/42/EC [54] for the reduction of VOCs emitted from paints, polishes, and materials for vehicle restoration coatings, is the result of an integrated strategy against acidification and surface-level ozone. The amount of coating materials used depends mainly on the model line of the vehicle. On average, there will be a treated surface of between 15-25 m<sup>2</sup>, corresponding to approximately 12-15 liters of coating materials.

The production plants that provide surface treatments usually use a general procedure, but one that differs at individual plants. In the preliminary treatment, a phosphate layer is applied onto the passenger vehicle body, which not only provides long-term corrosion resistance, but also improves paint adhesion. The second phase is an electrophoretic varnishing with the so-called E-coat layer. Next, a primer is applied to furnish several functions. The most important one is the smoothing-out of any surface unevenness, and to thus to form a solid basis for the application of the paint. This has a positive effect on the adhesion of any further layers, ensures anticorrosion protection, and protects the passenger body from damage. Next, comes a varnish base to ensure good optical properties. The last step consists of applying a top coat of transparent varnish, which protects the vehicle from environmental effects.

Cathodoporesis is an electrophoresis technology for varnish deposition. Electrically charged particles of a cathodic coating form a layer with an approximate thickness of 20 μm. Cathodic coatings are water-soluble and prepared based upon epoxy or ultimately acrylate compounds, with a low content of organic solvents. Coating reinforcement is processed at higher

temperatures. Applying 20 μm of an e-coat layer requires approximately 2.9 liters of paint.

The polymer primer layer, usually made based on polyester or polyurethane, forms an approximately 30 μm thick layer. The primer is applied to both the interior and exterior of the vehicle. Primer, based on LER, has the advantage of enabling bonding with galvanized as well as nonferrous metals. The primer is resistant to petrol and diesel, and it is thus possible to be applied in the engine area. Applying a 30 μm primer layer requires approximately 4.3 liters of paint.

The varnish base contains color pigments that provide almost the final appearance. Application of the varnish base is a fully automated process. The coating is applied to the interior and exterior of the vehicle in one or more layers. It forms an approximately 15 μm thick layer. In the case where a metalliferous or pearly varnish is being applied, these particles are contained within the second color compound of the varnish. The basic varnish is usually based on polyurethane, but can also be based on epoxy compounds. The coating can be water-soluble or in a powder. Application of a 15 μm varnish base layer requires approximately 2.1 liters of paint.

After application of the varnished color base, a transparent varnish layer is applied. Transparent varnish is identical for common as well as metalliferous varnishes. The topmost layer is dried in a dryer, which provides reinforcement of the varnish. The topmost varnish is usually based on epoxy compounds and this protects the passenger body from mechanical damage, UV radiation, temperature variations, etc. An approximately 40 μm thick varnish layer requires 5.7 liters of paint.

### *Adhesives*

One of the key strategies of automotive producers is improving their products, leading to an emphasis on decreasing the total weight of the vehicles. Decreasing the weight is connected to the development of new materials, leading to more frequent utilizations of plastic, aluminum, or various composite parts. These materials cannot be welded, only glued. Applying glue provides an advantage, as there is no need for subsequent treatment of the vehicle body in order to provide a faultless appearance. It also decreases production costs.

According to the basic constituent compound, glues can be divided into epoxy, acrylate, and polyurethane, etc.; this division, however, provides only a rough picture. Thermosetting glues can be utilized in many fields of the automotive industry (Table 1) [55]. The content and properties of the glues used in passenger vehicle body construction are closely related to the required function of the joint. Glues can be divided according to their purpose: sealing, reinforcing, or structural. Glues based on LER provide solid joints, strength in the case of impact, and protection from corrosion [55, 56]. Glues used mainly in passenger

Table 1. Function and types of glues used in the automotive industry; table developed based on [55].

Application	Function	Material basis
Structural Bonding	Joining of different substrates	Modified silane (MS)-polymer
	- semistructural	- Elastomer
	- structural	- (Meth)acrylic
	- crash-resistant	- Epoxy and/or Polyurethane
Structural Foams	Local reinforcements	Epoxy and/or Polyurethane
Sealing	Protection against corrosion, water and dust, isolation of noise propagation	PVC
		Elastomer
		EVA
Panel Damping	Reduction of noise, vibrations and harshness (NVH)	PVC
		Epoxy
		Elastomer

vehicle construction have to enable the application of further coatings. Other requirements of the glues are short time intervals for reinforcement of the glued joints, and the glue durability has to be longer than that of the vehicle. Construction glues have to ensure a high structural strength despite a lubricant layer which may appear on construction metal sheets. Unlike other joint technologies, the glued joint does not have maximal strength just after its application. The reinforcement period depends on the adhesive agent used, temperature, and the requirements for joint strength. Strengthening glues are usually reinforced together with the varnish of the passenger vehicle body.

Glues based on epoxy are used to glue various materials, and belong to the class of glues with high adhesion, which ensures very strong joints [57]. Glued joints increase the tenacity of the bearing structure of the passenger vehicle body. Thus, the vehicle can absorb vibrations and inhibit noise inside the vehicle. They also diminish energy in case of an accident [55]. Single-component epoxy glues that are reinforced at high temperatures are mainly used in special applications (flange seams in engine hoods, doors, and vehicle decks). Glues are also used to glue in printed circuits – for example, Škoda's Octavia model, a typical passenger car, uses up to 1.5 kg of glues based on LER.

#### Printed Circuit Boards

Printed circuit boards (PCBs) are used to interconnect electro technical parts. According to the layout, they can be divided into one-sided, two-sided and multilayer boards. Many materials are used for their production; and thus, their thermic and insulation properties differ. The boards are marked according to the materials they are made of and their flame resistance (FR). LER with glass fiber is among the most often used materials for bearing PCBs, and is graded as FR4. It is excellent material from both a price and quality point of view. The bearing board is produced in a manner where

a spun glass textile is impregnated by epoxy agent. This forms a material called *prepreg*. After reinforcement, the board is processed into more layers or according to the specific wishes of the customer [58]. Plastics in PCBs form a maximum of 30% by weight, out of which 4.8% by weight are epoxy compounds [59]. The more the vehicle interior is specially equipped (audio and video facilities, airbags, on-board computer, navigator, safety devices, etc.), the more PCBs it contains, and the more epoxy resin is utilized. Circuit boards including epoxy resins are also a specific topic for waste management [38, 60-63].

#### Composite Compounds

The automotive industry, along with the aircraft industry, are some of the biggest consumers of composite materials. Composite materials are usually heterogeneous, containing two or more compounds of different properties that together have properties that none of the parts have prior to their combination. A composite part contains a mold (matrix) and reinforcement. The part usually contains a filling that provides it with a specific property, for instance fire resistance. The matrix is surrounded by reinforcement, which forms the shape, fixes the filling materials, and transfers mechanical stress to partial fibers. Composites with a polymer or, specifically, a thermoset matrix are the most commonly used group of composites; amounting to over 2/3 of the market [64]. This is mostly due to their low cost, but also for their good properties under dynamic stress. Reinforcement is provided by fibers, usually carbon fibers (carbon fiber reinforced plastics, CFRP) or glass fibers (glass fiber reinforced plastics, GFRP), which provide mechanical properties such as strength and tenacity. Carbon fiber reinforced polymer composites (CFRP) are considered the acme in automotive equipment. They stand out with their low weight, high dynamic stress resistance, and good thermic conductivity. Composites ensure lower weight

Table 2. Material composition of an average European personal vehicle; table developed based on [69].

Material type	% (w/w)
Ferrous Metal	68.3
Plastics	9.1
Light non-Ferrous Metal	6.3
Tires	3.5
Glass	2.9
Fluids	2.1
Rubber	1.6
Heavy non-Ferrous Metal	1.5
Other	1.5
Battery	1.1
Process Polymers	1.1
Electronics	0.7
Carpets	0.4

of the vehicle, which leads to lower fuel consumption. They are also used as a design element. CFRP have a higher specific absorption of energy (SAE) at impact than do conventional materials.

LER is the most commonly used matrix in composite parts. It predominates mainly due to its resistance to material fatigue. Resin has an outstanding adhesion to carbon fibers in CFRP. The manner in which a part is produced depends on if it is designed to be utilized in the interior or exterior. The matrix of the parts consists of 30-70% by weight of composite material. There is a growing demand for the utilization of composites with a thermoplastic matrix, especially due to their easier recyclability [64, 65]. However, thermoset matrices form reticulated structures, and cannot so easily be reused as thermoplastic ones can. This presents a huge challenge to waste management facilities [66].

#### *Epoxy Resin within Vehicle end of Life Phases*

Vehicles with completed operation periods (end of life vehicles, ELVs) are processed in authorized facilities. European Union member states have to organize the processing of these vehicles according to requirements on general specifications of waste handling. The goal of directive 2000/53/EC [20] is to increase materials reuse and limit the utilization of hazardous materials; thus, implementing sustainable vehicle production [67, 68]. In facilities authorized by the state, those vehicles having completed their operational period are dismantled and all environmentally hazardous parts should be removed. Starting from January 1, 2015, 95% of an average vehicle's weight must be materially reused or recycled, and 10% of this should be used for energy production/

recovery. Only 5% of average vehicle weight can be deposited into waste sites.

Wrecked vehicles thus provide a model for drawing attention to the problems connected to the amortization of waste. Vehicles are constructed from various combinations of materials. According to the framework of European legislation, the wrecked vehicles should provide spare parts and secondarily used materials. Such materials should have standardized properties, utilizable by the purchaser of the reused materials, and whose production is certified in accordance with ISO specifications. European specification 2000/53/EC is a challenge to innovate post-shredding technologies [68, 69]. These data are important for following life cycle assessment. After receiving ELVs at an authorized processing facility the vehicle is first dismantled and then broken and crushed in a shredder.

#### *Dismantling of Vehicles*

Dismantling starts by the draining of operational fluids and removal of other hazardous parts of the wreckage (battery, airbags, fuel, oils, cooling fluids, antifreeze, etc.) Dismantling of individual parts depends on the age of the vehicle. Some parts are dismantled in order to provide reusable materials, for instance tires, larger plastic parts (dashboards, battery covers, and bumpers), glass, electronics, or eventually composite parts [36]. Nonferrous material is sold to authorized dealers for further processing. One of the most important tasks of producers is decreasing vehicle weight, which leads to the increasing use of materials such as plastics, aluminum, and composites instead of ferrous metals. Aluminum recycling is technologically easier and economically more feasible than plastics recycling [33]. Car designers consider plastics one of the critical components, as only larger parts get recycled. This is related to efforts toward decreasing the many different kinds of plastics used. Marking their type also makes identification during dismantling easier [33]. After dismantling, the vehicle is broken and crushed in a shredder.

#### *Processing Vehicles and Waste from the Shredder*

The crushing and shredding of vehicle wrecks produces 3 kinds of materials that are separately sorted: 75% of mass consists of ferrous metals, 25% is formed of light and heavy fractions, i.e., residua. The light and heavy fraction is designated as waste from processing vehicles after shredding (ash shredder residue, or ASR). The metal fraction gets sorted via magnetic facilities, and the separated metals are then sold for further utilization [34]. The higher LER fraction that occurs within the vehicle body will become a part of the metal fraction, which will be processed in either metallurgical or steel works. Some parts will probably form in the light fraction of the ASR. This is the result of mechanical damage to the varnish during shredding.

Table 3. Ash shredder residue material compounds, a non-sorted sample; table developed based on [91].

Material compound	w/w
Fines	45%
Foam rubber	18%
Cellulosic	1%
Rubber	7%
Metals	1%
Soft plastic	4%
Rigid plastic	15%
Textiles	9%

The ASR mostly consists of crushed polymers, rubber, textiles, residual metals, and glass (Table 3). Almost half of ASR consists of the light fraction, often contaminated by mineral oils and heavy metals. According to the list of hazardous wastes, ASR is characterized as a hazardous waste in Europe. However, it is usually disposed of in normal waste sites [37-39]. On some physio-chemical parameters, ASR exceeds the limits defined in the legislation for fuels received for waste incineration. In order to be reused for energy, ASR must first be treated prior to any further processing.

To achieve the European directive 2000/53/EC [3] for the 95% reuse and recycling of materials, ASR can be incinerated in cement or metallurgical works. However, it has limitations. Burning ASR enhances the formation of ash and the emissions of hazardous elements. Among the other technologies that can process ASR are pyrolysis or gasification. However, the benefits of processing ASR in these technologies are very little documented [33, 38]. Pyrolytic processing is especially suitable for the heavy fraction of ASR. At a temperature of 500°C the organic fraction is completely decomposed [37]. Precious metals are acquired from the solid fraction by magnetic sieving. Pyrolysis of oil has a high heating value, and can be used as an alternative fuel; however, it must be processed to ensure against the emission of potentially hazardous substances. Oil is also a potential source of materials for the chemical industry [36, 37]. ASRs can,

to a limited extent, be used as a filler in concrete or asphalt.

#### *Processing the Metal Fraction*

LER on the vehicle body is found in the form of coating systems and glues. The metal fraction from the shredder is processed at high temperatures in metallurgical and steel works, in which the resin is burned.

#### *Removing Shredded Residue by Depositing in Waste Sites*

ASR is removed to waste sites. Waste disposal via deposition in waste sites has the lowest priority, in accordance with the European waste treatment hierarchy. In environmental and economic terms, it is the least acceptable method of waste disposal, as it completely loses the material and energetic resources contained within the waste. Currently, among discussed problems is leakage from waste sites. Bisphenol A, from which the LER is synthesized, is one of the endocrine disruptors, specifically between estrogen receptor agonists. Sewage treatment plants are not able to remove bisphenol A completely, which leads to their bioaccumulation in the bodies of animals and humans [70].

#### *Processing of Printed Circuit Boards*

PCBs are highly heterogeneous materials. Processing PCBs in the waste management system is a challenging process. The boards contain some precious metals (silver, platinum, copper) and are therefore a desired commodity by waste processors. Electric waste processors mainly focus on recovery of metals. The nonmetallic fraction (i.e. glass fabric) is burned or deposited into waste sites [71]. Fig. 1 shows a diagram for the mechanical processing of PCBs.

#### *Processing Composite Materials*

Composite parts are heterogeneous materials. At present, composite components that are part of ELVs are disposed of by landfilling or incineration. The driving

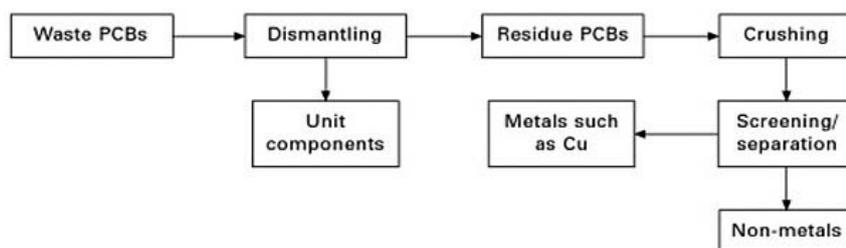


Fig. 1. Scheme of mechanical processing of waste printed circuit boards; scheme developed based on [58].

force behind recycling is the recovery of fibers, mainly those of carbon, which have a high economic value. The recycling of composite materials with a thermoset matrix is limited by technological and especially economic parameters. For recycling FRP with a thermoset matrix, various technologies have been developed, but some have only been tested at the laboratory scale [64]. Currently, they are not commercially used, primarily due to the absence of a recycling market. The production of new composites, which enables easier recycling of the separate parts, is a major challenge for vehicle designers [64]. Suitable recycling technologies, with regard to the recycling of composite matrices, are listed in Table 4. Due to the increasing volume of composite utilization, and its long life cycle, it is necessary to develop new economically and environmentally sustainable technologies that enable their material and energetic reutilization [66]. In recent years, there has been an especially increased production of CFRP. In 2013, worldwide production reached 48,000 tons; but by 2020, it is expected to reach 140,000 tons.

#### *Mechanical Recycling*

Mechanical recycling is a technology used commercially for the recycling of composite parts. Such processing, using mechanical recycling, is suitable for composites that are not too contaminated, by operating fluids for example. Composite scrap is cut, crushed and then ground to a size of less than 50  $\mu\text{m}$ . The resulting powder contains a mixture of fibers, fillers, and polymer matrix. The particles are sorted through a cyclone and screen according to size, into coarse and fine fractions. The coarser fraction consists mainly of reinforcement fibers. The finer fraction has a high content of fillers and polymer from the original composite [66, 72]. Mechanical processing (crushing, grinding) is a simple process technologically. However, fiber lengths get shorter and the overall quality degrades. In the process, the fibers lose their original mechanical properties, and thus reduce the economic return from recycling [73]. The fibers are reused as filler in new composites. Additionally, they are used in the building industry as an additive to asphalt, or as a mineral source in cement. The fine fraction is used as a filler; however, due to its low density, its potential for reuse in composites is quite limited [74, 75]. Due to the low economic value of recycled materials, mechanical recycling is mainly used in the recycling of composites with glass fibers [74].

#### *Thermal Recycling Technologies*

Thermal processes recover and utilize energy from the composite matrix, but also fibers, fillers, and low molecular weight organic compounds out of which the matrix is constructed. Composite materials with a polymer matrix have a high caloric value and

are therefore a good source of energy. The amount of energy depends not only on the matrix used in the composite, but also on the types of reinforcement and fillers. Inorganic fillers reduce the ratio of energy gained; moreover, incineration creates large amounts of ash. On the basis of studies that have been conducted in cooperation with automotive producer Volvo, where the effects the additives in the composites had on analysis after emission from incineration, it was recommended that a mixture of the composite materials be added to a common incineration material, such as municipal waste. A mixture consisting of 10% by weight of composite waste and 90% by weight of municipal waste produces emissions that meet the limits set by European legislation [76].

It is possible to combine both the material and energetic re-utilization of composite wastes in the manufacture of cement, especially in the case of GFRP. The matrix is incinerated and thereby it is reused energetically. Approximately 2/3 of the composite portions, depending on the composition, are used as a raw material for cement. The organic matrix is used as an alternative fuel, and thus decreases the consumption of other (fossil) fuels. Glass fibers are used as an inorganic fraction and replace clay and limestone (fillers) in the cement [74, 77]. The composite portion has to be pre-treated, i.e., reduced to a suitable size. For effective use in the cement industry it is necessary to have a constant supply of composite waste.

Fluid technology is suitable for the recycling of GFRP and CFRP. The composite material is first crushed into smaller pieces and then inserted into the reactor. The organic matrix is used as a source of energy during thermal processing. The optimum temperature in the reactor for a composite with an epoxy matrix is set at 550°C. At this temperature the polymer vaporizes from the composite, releasing the fibers and fillers which are suspended in the gas steam. The renewed fibers are clean. Carbon fibers at 550°C lose 20% of their tenacity [64, 72, 77]. The gasified matrix is burned in the next reactor, releasing further energy. This recycling technology has only been tested on a laboratory scale. An economic analysis of the recovery costs for recycling of GFRP using fluidized burning was evaluated. It was found that 10,000 tons of GFRP had to be processed annually to ensure an economic return with this technology [75]. In the case of processing composites containing carbon fibers, the amount of waste can be lower due to their higher economic value [64].

Other thermal technologies include pyrolysis. This is a process in which the organic material is decomposed at high temperature in the absence of oxygen. Due to the high temperatures the long fibers are released from the composite matrix almost without damage [64]. An important parameter of this technology is the controlled temperature in the reactor and the time period for keeping the composite in the reactor. These process conditions influence the depolymerization of the matrix and the purity of the recovered fibers. Recycling

via pyrolysis is suitable for carbon CFRP and GFRP [64, 77]. During pyrolysis the reticulate polymer matrix is decomposed into low molecular organic substances. They constitute a potential raw material suitable for further chemical processing, it can also be used as a secondary fuel in the form of a condensed liquid product (a mixture of organic compounds with a caloric value on par with fuel oil), or further burned for the generation of electric energy. The gas product can primarily be used as a source of energy for maintaining the temperature in the reactor [64, 77].

In research at the University of Leeds, pyrolytic processing was performed on a composite with an epoxy matrix. The weight percentages of carbon composite pyrolysis products processed at 500°C were as follows: 67.4% solid, 31.3% oil/wax, and 1.2% gas. The pyrolysis gas had a caloric value of 40 MJ/kg, and compared with other matrices it was rich in methane. In the presence of the catalyst, the pyrolysis reactor was maintained at 220°C. The fibers were released from the composite matrix, and the epoxy matrix was also completely decomposed into lower molecular weight hydrocarbons in liquid and gaseous states [77].

#### *Chemical Recycling*

Research into chemical recycling is still ongoing. Via chemical recycling it is possible to obtain the fibers, fillers, and depolymerized matrix. Fibers obtained in this manner achieve the same quality as in the original composite. Chemical recycling is considered the only real recycling technology that makes possible the reuse of the materials the composite is composed of [64]. According to the solvent used, the solvolysis is classified as glycolysis, hydrolysis, etc. To increase the efficiency and speed of the process, it is possible to use supercritical conditions. The solvolysis process can decompose a polymer matrix into its original monomers, and these can be reused in the chemical industry. However, the solvolysis processes were only tested in autoclaves [64], and high-quality carbon fibers from the epoxy matrix without using supercritical conditions in a mixture of hydrogen peroxide and N,N dimethylformamide were obtained [78]. The choice of the solvolysis process depends on the type of composite matrix. Prior to the process, the composite materials need to be properly categorized and separated, which can be a difficult process. Any potential commercial application of chemical recycling would require the research to mainly focus on resolving the potential environmental problems related to the resultant toxic water treatment.

### Methodology of LCA

#### *General Framework of LCA*

Life cycle assessment (LCA) is an analytic method for evaluating potential environmental impacts of products,

services, and technologies [79]. LCA methodology approaches the assessment of the environmental impacts of a product with respect to their entire life cycle, which includes all stages: from acquisition of the raw materials, production of the basic materials, through production of the final product, including its use by the consumer, and its final disposal [80]. Environmental impact assessment of the products are based on evaluating the impacts of the material and energy flows that are exchanged between the observed system and the surrounding ecosystem, i.e., the environment [76, 81, 82].

Individual phases of the life cycle are created by operations transforming the materials and energy inputs into outputs. These processes are interconnected by their material and energy flows. The entire system of processes taking part in a product's life cycle is called the product system. LCA is an interactive methodology, which means that if it is not possible to ensure a consistency of the processed phase with some of the previous phases, the previous phase and all the following phases have to be reprocessed. In order to fulfil the original goal of the study, some aspects regarding its scope may require modification.

#### *LCA Calculations*

The life cycle modelling of liquid epoxy resin life cycle in the automotive industry was performed using GaBi software. GaBi contains a database tool used for general environmental as well as socioeconomic equilibrium modelling. The modular system is constructed in such a way that the database and software are mutually independent units. This enables it to work with separate plans, processes, flows, inventory tables, and environmental impact assessments. Life cycle environmental assessment through GaBi respects ISO 14040 standards [83].

#### *Function of the Assessed System and Functional Unit*

The function of the examined system is to utilize LER as a material in the automotive industry. Epoxides are primarily used as coating materials; whereas their main purpose involves protection against corrosion, as well as chemical and physical agents in the environment. Increased attractiveness of the vehicle passenger body is an equally important function. Epoxides are also utilized as high structural strength glues that provide excellent adhesion to materials. Within this study, examination of LER as a material with a specific function is less substantial. The function of the system simply means the use of LER materials that have a quantified weight.

The functional unit used for performed calculations involves 5 kg of LER, which corresponds with the average amount of LER in one vehicle ranked in the lower-mid and mid-classes of the Toyota, Peugeot, Citroën, and Škoda brands. The quantity of LER,

Table 4. Recycling technologies for composite parts with thermoset plastic matrices; table developed based on [64].

Recycling methods	Technological features	Status of the technology
Mechanical recycling	Shredding - grinding – milling	Commercial operation
Thermal recycling	Combustion/incineration with energy recovery	Promising technology
	Fluidized-bed thermal process for fiber recovery	Only laboratory studies
	Pyrolysis for fiber and matrix recovery	Hindered by the market for recycled fibers
Chemical recycling	Chemical dissolution of matrix	Only laboratory studies
	Solvolysis, potential recovery of resin	Promising technology

expressed in kg, is thus most in accord with the goals of the study. The reference flow involves 5 kilograms of LER, and corresponds with the functional unit.

#### *System Boundary*

The product system encompasses LER production, transport of the product to the car manufacturer, utilization by the user, shredding, transport of the waste, incineration, and landfilling of the waste. The product system does not involve the processing of waste created during LER production nor during the processing of LER by the car manufacturers.

#### *Environmental Impact Assessment*

The characterization model CML 2001–April 2013 was applied in the LCA study. The reason for using the CML characterization model stems from the fact that this model is the one most frequently applied in LCAs within the automotive industry. This is a model which is adjusted for the evaluation of products [84, 85]. The subject of the study involves the following impact categories: global warming, abiotic resource depletion, stratospheric ozone depletion, production of photo-oxidants, acidification, and eutrophication. Ecotoxicity impact categories were not assessed, particularly because emissions from individual processes were replaced by emissions from mixed plastics.

#### *Assumptions Accepted During Development of the Study*

LER, which is one subset of the ELVs, does not have a waste catalogue number. It is taken as an accepted precondition that 70% of the total weight of LER within vehicles are later processed in steel works. The procedure of processing in a steel works has been replaced by the process of plastic incineration. This is to prevent a situation where the potential environmental impacts of elementary flows released from the metalworking would be attributed to the incineration of LER. The remaining 30% of the LER's weight, which is a part of the ASR, is removed to landfills.

Emissions from the incineration are modelled as emissions from the incineration of mixed plastics. Due

to the absence of data for LER incineration emissions, the data from incineration of mixed plastics were used instead. Data for the landfilling of mixed plastics were applied for the emissions of elementary flows from the landfill.

## **Results and Discussion**

### **Life Cycle Inventory and Data Collection**

Information about the average amounts of LER-based coating materials and adhesives in cars of the lower-mid and mid-classes originate from data provided by the manufacturers of Toyota, Peugeot, Citroën, and Škoda cars. The highest amounts of LER in the mid-class vehicles are present in the passenger body of the car. This scenario of the LER life cycle was created by using publicly available data and with the cooperation of LER producers, as well as 4 automotive manufacturers in addition to waste management facilities. The LER life cycle in the automotive industry involves the acquisition of raw materials, LER production, processing of LER by the car's producer, use of the vehicle by the consumer, and end of life of the vehicle within the waste management system. When a vehicle's operational life is completed, it is received by the authorized processing facility, where it is first manually dismantled and then shredded. The metal fraction along with the LER is further processed either in a metallurgical or steel works. The remaining LER, which is part of the ASR, is then landfilled. It is generally assumed that when the ELV is processed in a shredder, 70% of the total LER in the vehicle will be removed, together with the metal fraction, to be processed in metallurgical or steel works. The LER is then incinerated. The remaining amount, i.e., 30% out of the total LER, is present in the ASR and it is taken to a landfill. The passenger body is processed at high temperature in a steel works, where the metal is recovered and the LER is incinerated.

The experimental part of the work focused on identifying those parts of vehicles that contain LER; eventually analyzing further possible epoxy utilization, its advantages in the automotive industry, and subsequently evaluating its life cycle. In order to identify the exact amount of resin and further model

the life cycle scenario, it was necessary to establish cooperation with automotive manufacturers as well as processors involved in waste management.

Data from the processes of epoxy resin production were provided by Plastics Europe. The distance of materials transport was selected as the average difference between European car producers (Toyota, Peugeot, Citroën, Automobile Czech, s.r.o., Kolín; Škoda Auto a.s, Mladá Boleslav; Adam Opel GmbH, Rodgau; Porsche, Stuttgart; PSA, SitedeSevel Nord) and the supplier of the materials. This distance corresponds to 450 km. Trucks with a freight capacity of 12.4 tons were selected for transport, along with a diesel fuel that corresponds with the European average EU 27 Diesel mixture.

The average quantity of LER-based coating materials and adhesives in cars of the lower-mid and mid-classes corresponds with the average amount used by manufacturers of Toyota, Peugeot, Citroën, and Škoda cars. The operational data regarding energy consumption related to technologies for the processing of coating materials (drying, degreasing, varnishing, and reinforcing processes) per unit passenger body was provided by the manufacturer and was 0.402 MWh of electricity per unit and 21.18 m<sup>3</sup> of natural gas per unit. The transport distance for the processed ELVs between individual processors was set at 150 km. A truck with a freight capacity of 12.4 tons was selected for transport, along with a diesel fuel that corresponds to the European average EU 27 diesel mixture.

### Results of Environmental Impact Assessment

Specific environmental problems emerge as a consequence of releasing elementary flows from individual phases of the product life cycle into the environment. Using characterization factors the results from inventory phase were expressed as results of individual impact categories. Assessed impact categories correspond with the evaluated categories that are common for LCA in the automotive industry. Characterization profile or calculations of impacts of individual categories of the product system are provided in Table 5.

The carbon footprint (CFP) represents the total quantity of greenhouse gases (in kilograms of CO<sub>2</sub> equivalent) that are emitted into the environment as a result of the product's life-cycle. The process of LER production has a dominant share in the overall carbon footprint of the LER life cycle within the automotive industry. LER production represents an 84.5% share of the total carbon footprint of the product system.

After the process of resin production, the second-highest CPF in the examined product system pertains to the phase of removing the LER, which is a portion of the ELVs. Removal of resin within ELV waste management represents a 7,8% share of the total carbon footprint of the LER product system. In the modelled product system, the removal of LER in landfills does not have

Table 5. Potential environmental impacts of epoxy resin life cycle in automotive industry expressed as indicator results of CML characterization model; results expressed per functional unit, which was 5 kg of LER (average amount in personal vehicle).

Impact Category	Total	LER production, %	Transport, %	Car Manufacture, %	Shredder, %	Waste Transport, %	Incineration, %	Landfilling, %
Abiotic Depletion (ADP elements) [kg Sb-Equiv.]	0.0002	99.1	0.0	0.5	0.0	0.0	0.3	0.0
Abiotic Depletion (ADP fossil) [MJ]	961	62.6	0.2	34.4	0.2	0.1	0.2	0.4
Acidification Potential (AP) [kg SO <sub>2</sub> -Equiv.]	0.121	87.9	0.6	10.3	0.3	0.2	0.8	0.0
Eutrophication Potential (EP) [kg Phosphate-Equiv.]	0.022	96.1	0.7	2.4	0.2	0.2	0.3	0.0
Global Warming Potential (GWP 100 years) [kg CO <sub>2</sub> -Equiv.]	48.8	84.5	0.3	7.0	0.3	0.1	7.8	0.0
Ozone Layer Depletion Potential (ODP, steady state) [kg R11-Equiv.]	5,83E-09	0.0	0.0	98.2	1.7	0.0	0.1	0.0
Photochemical Ozone Creation Potential (POCP) [kg Ethene-Equiv.]	0.014	80.2	0.5	18.6	0.2	0.2	0.4	0.0

any carbon footprint. The modelled process takes into account the processing of the landfill gases produced.

The resource impact categories represent the influence of product systems on the decreasing availability of renewable and non-renewable resources. The LER life cycle in the automotive industry is particularly a cause for the high consumption of fossil resources.

GWP represents a gauge for potential contributions of elementary emissions to withhold energy from being put into the atmosphere. The highest contribution to GWP pertains to emissions from LER production, and to emissions from LER incineration in metallurgical or steel works in the removal phase of the ELVs. Greenhouse gas emissions also occur during LER processing at car manufacturers.

Acidification of the environment is caused by increased concentrations of acidifying agents into the environment. Acid deposition influences not only the quality and function of the natural ecosystem, but additionally of the anthropogenic landscape. The LER production phase contributes significantly to the production of emissions that have a potential for acidification.

Anthropogenic eutrophication is caused by increased emissions of bioavailable nutrients into the aquatic environment. The eutrophication potential specifies the quantity of nitrogen and phosphorus from the elementary flow emissions that may be released from the process into the environment. During the LER life cycle in the automotive industry, the highest emissions of agents containing potentially bioavailable nitrogen and phosphorus occur in the LER production phase, namely during the waste water cleaning process. These emissions can potentially support eutrophication.

Depletion of stratospheric ozone is caused by emissions of agents that induce the decomposition of stratospheric ozone. Depletion of stratospheric ozone brings about increased quantities of UV radiation penetrating to the Earth's surface. LER processing at car manufacturers induces emissions of agents that have the potential to initiate decomposition of the stratospheric ozone.

Finally, LER production and LER processing at car manufacturers induces a main amount of emissions of agents that contribute to increases in the concentration of tropospheric ozone in the environment.

The following significant issues were formulated on the basis of the inventory and characterization results. The process of resin production in the automotive industry represents the largest share of the total carbon footprint and in the LER life cycle. LER production represents an 84.5% share of the total carbon footprint of the product system.

Removal of LER that is part of the ELVs, and carried out in steel works, has a greater CFP than its removal at landfills. This implies that it is better to utilize LER as a secondary raw material, e.g., as cement or asphalt filler, rather than using it as an energy source.

The LER life cycle in the automotive industry is intensive as regards resources. Fossil fuel resource consumption occurs particularly during the phase of LER production.

To ensure validity of the formulated significant issues, sensitivity analysis was performed. The influence of changes in the total quantity of LER, which are part of the vehicle, on the overall potential environmental impacts were monitored. The total quantity of LER in the product system was increased by 25%, i.e. to 6.25 kg. It was found that environmental impacts within the given range are not dependent on the change of the weight factor. Influences on the monitored impact categories increases linearly with the growing quantity of LER. The change is the equivalent of 25%. Sensitivity analysis was also applied to the accepted presumption, that after the operational period of the vehicle is completed and processed in the shredder, 70% of the total quantity of LER in vehicles (i.e., 3.5 kg) forms the metallic part of the vehicle passenger body, and that this quantity is further processed in either metallurgical or steel works. The remaining LER, which is part of the ASR, is landfilled. This is labelled as an alternative scenario. There was an alternative product system modelled, where on the contrary 70% of the LER out of the total amount is part of the ASR, and it is landfilled. Table 6 illustrates the influence of

Table 6. Percentage change in the characterization profile results of the LER life cycle in alternative scenario according to basement scenario; in alternative scenario 70% of LER as a part of shredder residue is landfilled.

Impact Category	Alternative scenario
Abiotic Depletion (elements) [kg Sb-Equiv.]	+ 4,0 %
Abiotic Depletion (fossil) [MJ]	+ 0,0 %
Acidification Potential [kg SO <sub>2</sub> -Equiv.]	+ 1,0 %
Eutrophication Potential [kg Phosphate-Equiv.]	+ 0,5 %
Global Warming Potential (100 years) [kg CO <sub>2</sub> -Equiv.]	+ 10,4 %
Ozone Layer Depletion Potential (steady state) [kg R11-Equiv.]	+ 0,1 %
Photochem. Ozone Creation Potential [kg Ethene-Equiv.]	+ 0,5 %

an alternative scenario on the characterization profile of the product system. The results imply that removing 70% of the LER, which is part of the ELVs, by landfilling, decreases the overall environmental impact of the product systems on the majority of the assessed impact categories. The potential impact on greenhouse gases is thus decreased by 10.4%. Another, but negative consequence of the alternative scenario can be the release of plastic micronutrients into the environment from landfilled epoxy resins. Unfortunately, there are not characterization factors for such elementary flows published yet, so here the presented results probably underestimate possible environmental impacts of alternative scenarios.

The decrease in potential impacts on greenhouse gas emissions needs to be understood within the context of the type of processing of a car. Emissions from LER processing in steel works were modelled as emissions from the incineration of mixed plastics. Emissions from LER landfilling were modelled as emissions from the landfilling of mixed plastics.

Considering the results of the sensitivity analysis, it is necessary to focus on measuring emissions from LER incineration and LER behavior in landfills. Current discussions encompass landfill leachates that contain Bisphenol A, which acts as an endocrine disruptor [86, 87], which at present cannot be removed in water treatment plants [88, 89]. The degradability of plastics is discussed in general [90]. The modelling of emissions produced by the processes of plastic decomposition in landfills is subject to the assumption that the plastics are stable or even inert. However, recent studies indicate that decomposition may be faster.

As regards the electrotechnical parts of the vehicles, specifically printed circuits, LER can be found in the system boards. LER as a material is often applied for the impregnation of glass fabric. At present, the demand for composite parts is growing continuously. Composite materials reinforced with fibers have incredible potentials for all fields of design. The engineering design sector (wind turbines, aviation industry, etc.) is a majority consumer of fiber-reinforced composites, especially CFRP and GFRP. The automotive industry has a lower market share; nevertheless, it has started to reach the leading position as regards the innovation of materials (61). Fiber-reinforced parts are utilized especially because of their low weight, stability, and capabilities to achieve various shapes. Composites with matrix made from a thermoset polymer, and in particular from the LER, are the ones most frequently used.

If the LER is only present in vehicles as coating materials and glues, the LER is processed within the waste management system together with the passenger body of the vehicle. During the LCI phase, it has been ascertained that a waste catalogue number is not assigned for LER in the case of waste, resulting from the processing of scrapped vehicles. Hence, LER as a material in ELV is officially lost.

## Conclusions

The current trend in the circular economy stipulates requirements for the reuse of materials present in ELVs. This gives rise to requirements for the selection of materials in the course of car design. Composites with a thermoset matrix create major challenges for ELV processing facilities. Thermoset matrices, unlike thermoplastic ones, are reticulated, and cannot be easily reused; for this reason they are hard to recycle. Considering the increasing demand for composite parts and with regard to the legislative requirements, LER manufacturers will participate in the research of recycling technologies that are to be applied for composite materials. Unless the recycling of LER and thermoset matrices of composites, in general, is resolved, we can expect higher utilization of those matrices made from thermoplastics.

LCA study of LER within the life cycle of a passenger vehicle was developed and the following impact categories were used for evaluation: depletion of abiotic resources, potential for global warming, eutrophication potential, acidification, potential for photo-oxidant production, and depletion of stratospheric ozone. Contribution analysis indicated processes involved in LER production as the main source of environmental burden. Removal of LER carried out in steel works has a greater CFP than does its removal into landfills. The life cycle of LER in the automotive industry is a resource-intensive process. Fossil fuel resource consumption occurs especially in the phase of LER production and its processing at the car manufacturer. Elementary flows that are released into the environment during LER production have a potential for the development of GWP, POCP and AP impact categories.

Considering the results of the LCA study, more extensive cooperation with ELV processors should be started in the future. It is necessary to measure which fractions will be produced after shredding, and to what degree LER will become a portion. In addition, it is necessary to concentrate on measuring the emissions from LER incineration and on LER behaviors in landfills. Greater in-depth cooperation should be started with PCB producers in order to identify those segmented processes that are involved in LER processing in PCBs. LER producers should participate in these research projects that are focused on the recycling of thermoset matrices.

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## Conflict of Interest

The authors declare no conflict of interest.

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