8. The influence of adhesives on material recycling

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1. Introduction

As a result of both the dramatic increase in world population and the improved living standard of many people after the industrial revolution, we have seen an extreme increase in consumption of natural resources such as wood, water, coal, metal, and oil. The world-wide consumption of natural resources increased from four to ten billion tons between 1963 and 1995 – the consumption of resources rose faster than world population. Experts expect that the consumption of natural resources will quintuple by 2050, if it develops equally to population growth. That is to get one's sums wrong because this is more than planet earth can supply in the long term. Already in 1972 the report of the Club of Rome showed the limitation of growth and opened our eyes to the finiteness of material resources. An increasing consumption would lead to a depletion of important mineral and fossil raw materials in a few decades.

While at the beginning of the Industrial Revolution nobody much thought about the finiteness of natural resources, during the last decades of the twentieth century the call for processes which carefully treat resources became louder and louder. The finiteness of many natural resources made it
necessary to think about how to save them by developing resource-saving technologies. This leads to the idea of Sustainable Development. Sustainable Development was introduced at the environmental summit in Rio de Janeiro in 1992 as a model for future action. The idea is that people living today should be able to satisfy all their needs without leaving a heritage of pollution, climate problems, and used-up resources for future generations. The solution of this task depends upon our ability to use the existing resources of the earth economically and ecologically efficiently. These existing resources are too precious to destroy them after single use. Methods to stop the impending depletion of our natural resources are economical handling of resources, repeated use of sophisticated goods, and consequent recycling on the highest technical level. To execute this recycling successfully, it is necessary to consider this early during the construction and production of goods. The approach is called closed-cycle economy. This is a kind of economy, which was initially introduced in the nineteen nineties and nowadays seems to be not only an ecological but also economical model of success. Avoiding waste is top priority. Waste has to be recycled as far as possible and material should be recovered. Waste does not have to be discarded but can be a raw material very much in demand with lots of opportunities for use. Waste residues would be reduced to a minimum if material circulation could be closed completely. Recipes which make good ecologic sense cannot be free of charge, but the introduction of new sorting and recycling techniques will lower the costs of recycling considerably and will also improve the quality of secondary raw materials. This is a prerequisite for sophisticated products that must hold their ground in the international market. The objective is to save resources and to find renewable raw materials and energy sources with a sustainable economy for all future generations.

Legal regulations reflect this trend in our society. For instance, the introduction of the Packaging Directive in Germany in 1991 was an important step in this direction. The recycling rates and collecting systems predetermined in it have had a strong influence on the recycling of packaging material. On a European level this thought was legally established in the European Packaging Directive (Directive 94/62/EC) dated 20 December 1994. The objective of this directive was to reduce the amount of packaging waste in Europe by 50 % by 2001. At the end of 2001, as a new aim, a recycling rate of 55-70 % by the year 2006 was proposed. The Directive 2004/12/EC of the European parliament and of the council amending Directive 94/62/EC on packaging and packaging waste has set further targets. In order to comply with the objectives of this directive, member states shall take the necessary measures to attain the following targets covering the whole of their territory (Article 6 b), d), e)): No later than 31 December 2008, 60 % as a minimum by weight of packaging waste will be
recovered or incinerated at waste incineration plants with energy recovery. No later than 31 December 2008, between 55 % as a minimum and 80 % as a maximum by weight of packaging waste will be recycled. No later than 31 December 2008, the following minimum recycling targets for materials contained in packaging waste will be attained: 60 % by weight for glass; 60 % by weight for paper and board; 50 % by weight for metals; 22.5 % by weight for plastics, counting exclusively material that is recycled back into plastics; 15 % by weight for wood. Further regulations that demand a recycling of products according to their use are e. g., the EU "End of Life Vehicles" Directive (2000/53/EEC) from 18 September 2000. By 2005 at least 85 % (80 % material recycling) by weight of scrapped cars was to be reused or recycled, and this must rise to 95 % (85 % material recycling) by the year 2015.

To execute the idea of the recycling law it is necessary to think about a material recycling of the used products when designing them. This means that all additives of the process have to be designed in a way that they do not disturb later material recycling. This is especially important for the field of adhesives, because many of today’s products are produced with the help of adhesives.

2. Bonding technologies

Most of the products of everyday life consist of combinations of single components. To guarantee the function of these products, these single components need to be assembled. Joining techniques such as welding, brazing, riveting and screwing are used by industry all over the world on a daily basis, but another method of joining has also proven to be highly successful: adhesive bonding. Known for thousands of years, this method has become as important as the other joining techniques as a result of the pace of developments in recent years. In many areas, this bonding technology has even become a key technology: New, hitherto unrealizable combinations of materials, as well as the need for the highest requirements for connections, have made this flexible joining technique the preferred technology, especially in high-technology areas. In hybrid-joining, the unique advantages of using adhesives are combined with the benefits of other joining techniques.

2.1. Adhesive bonding - joining technology with high potential

Virtually all solid materials can be connected with one another using adhesive – for example glass with metal or paper with plastic. High temperatures are not required in the joining process. That is beneficial for the materials and prevents shape-distortion. Additional functions such as corrosion protection, vibration damping, electrical conductivity and sealing to liquids or gases can be
integrated into the bonded connection. Regarding the designing of adhesive bonded joints, optimum solutions which meet all requirements are determined. Mechanical loads which act on the connections are investigated and also, for example, the effects of liquids, gases, heat, electric current and light. Computer simulations and tests on standard samples give information about the optimum geometric design of the bonded joint, with strength under continuous loads being the most important criteria. The simulation results are then verified by means of load tests on pseudo-components. The successful realization of bonded connections in industrial production requires precise planning of the individual production stages. The joining process must be compatible with previous, subsequent and concurrent production stages. The handling of the parts to be joined is just as important as adapting the technology for applying the adhesive to the use: feeding, mixing, dosing and applying. The combination with other joining techniques is also of key importance. The various options for the hardening step must be taken into account at the planning stage [1].

For high-quality connections, special pre-treatment of the surfaces to be bonded is often necessary. The materials to be bonded are cleaned and activated or modified so that adhesives can adhere better to them. This also gives the surfaces to be joined protection against corrosion. An example is the pre-treatment of aluminium for aircraft construction.

Industrial adhesives are primarily required to join materials in a fast and safe way, and to provide for trouble-free, inexpensive production in existing manufacturing lines (using automatic machinery as far as possible). Furthermore, they must resist the conditions to which the finished product will be exposed when being used later. Price is another important aspect that must not be underestimated when selecting an adhesive. Ecological aspects, too, play an increasingly important role for the selection of an adhesive system. For example, more and more care has been taken that adhesives can be processed in a way that as little waste and wastewater as possible accrue from their production. In addition to that, the impact of adhesives on the reuse (e.g. returnable products) or recycling of bonded products is a growing matter of interest.

As adhesives are important for the production of nearly all goods today, it is increasingly important to understand how adhesive applications influence the idea of the closed-cycle economy. Although adhesives are not recycled per se because of their small amount, they have to be designed in the way that they do not cause troubles when recycling the primary material.

3. Adhesives in recycling processes

Today adhesives play a decisive role in the production of almost every good, especially for mass-produced articles. Bags made of paper, graphic products like
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catalogues or magazines, furniture, cars and airplanes – all these products are manufactured with the help of adhesives today. Hence it is getting more important how adhesive applications influence the idea of closed loop economy. In comparison to the materials being bonded by adhesives, the adhesive quantities used are so small that it is normally not worth recycling the adhesive itself. Even though adhesives due to their small amount will not be recycled alone, it must be considered that they should not disturb the recycling of the primary materials. However, today more and more primary materials are retrieved by recycling. This applies particularly to non-durable materials, such as packages are made of. It is expected of the adhesives used for these products that they do not interfere with the recycling of the primary materials, and not affect the quality of the secondary raw materials. Which properties are required from the adhesives, depends on the applied recycling process.

Adhesives that consist of organic polymers are normally trouble-free in recycling processes that take place at high temperatures e. g., metal or glass recycling, but the choice of adhesive for plastic and paper products is important because of the low temperatures during the recycling process.

4. Adhesives in high temperature recycling processes

High temperature recycling processes are used for the recycling of glass and metals. Temperatures above several hundred degrees are used, and adhesives based on organic material will be burned.

A special recycling process is energy recovery. In the “five step hierarchy” in waste management – prevention, reuse, material recycling, energy recovery, and landfill – the waste-to-energy process is another possibility to save resources.

4.1. Glass recycling

Glass is one of the oldest materials used for many different applications: it has long been used for packaging beverages like wine and other liquids. Due to its strength, tightness, clarity, and tastelessness it is perfectly suitable for this application. To mark glasses and bottles, labels are bonded onto glass packaging. Glass is also used in adhesively-bonded windows in great quantities. These achieve better thermal insulation of buildings and are an important architectural design tool.

Recycling of used glass and bottles has a long tradition. Used glass was already recycled for the production of new glass in the Middle Ages, probably even in ancient Rome. Today recycling of glass is at a very high rate today: At the beginning of the nineteen seventies used glass was systematically brought back to glassworks in large quantities to introduce glass recycling extensively.
In 1974 in Germany 150,000 tons of domestic sales of 2.3 million tons of container glass were reused, corresponding to a recycling rate of 6.5 %. By 2002 the recycling rate of container glass had increased to approximately 90 % [2]. Because of the willingness of the consumer to sort the different colors and because of very exact quality assurance systems in the recycling facilities, a green bottle consists of more than 90 % of used glass, and brown and white bottles have more than 50 % recycle content. The “Duales” System Germany has set up more than 300,000 glass collection containers throughout the republic to collected used glass packaging. All together more than 1.3 million tons of waste glass packaging were collected by the “Duales” System Germany during the year 2007 [3]. In total, in Germany approximately 2.5 million tons of glass were recycled in 2007. In Europe, in 2006 nearly 10.5 million tons of glass packaging were recycled (compared to 9.8 million tons in 2005) [4]. Thanks to glass recycling, waste disposal in Europe has declined (by 13 % between 1996 and 2006) despite an increase in the demand for glass packaging (by 11 % between 1996 and 2006).

In glass recycling the batch is melted at a temperature of about 1550 °C in a tank. Adhesives based on casein or synthetic polymers that are used to label glass bottles, or adhesives based on polysulfide and butyl rubber used for the production of insulating glass, do not disturb the glass recycling process. Because they consist of organic material, they burn at high temperatures without influencing the glass recycling process or the quality of the “new” glass.

4.2. Metal recycling

Adhesives are increasingly used for joining metal-to-metal and for joining metal with other materials. Similarly to glass, the recycling of metals has been a common process for a long time. In contrast to many other materials, in the recycling of metal there are no quality losses. Compared to primary metal extraction, a 95 % savings in energy can be achieved by aluminum recycling and a 75 % saving by steel recycling. Great strides have been made especially in the recycling of packaging made of metal, because the filling of food into aluminium cans or tin plate has increased dramatically during recent decades.

The economic value of aluminium has always been a reason for bringing the material into the loop of metal extraction, processing, use and recovery. Aluminium has been recycled since the days it was first commercially produced and today recycled aluminium accounts for one-third of global aluminium consumption world-wide. In Europe, aluminium enjoys high recycling rates, ranking from 58 % in beverage cans to 85 % in building and construction and 95 % in transportation. With the introduction of the “Duales” Systems Germany
in the packaging field there has been set up an exhaustive system for acquisition, sorting and reuse of aluminium packaging. In fact, there have been various recycling initiatives of the aluminium industry before, like melting down beverage cans, but for logistical reasons these programs did not achieve high recycling rates. The "Duales System" in Germany, however, shows a recycling rate of more than 97 % for aluminium packaging in 2002 [3]. The use of eddy current separators in sorting makes it economically attractive to collect and recycle all packaging containing aluminium and aluminium foil, and this contributes to fulfilling the recycling specifications of the packaging directive. During recycling, the aluminium cans are broken up in a shredder into small pieces, and then are conveyed into a de-lacquering oven to remove the paint, the labels, and residual moisture. The hot shredded aluminium is then passed over a small screen to remove any dirt and contaminants and fed directly into a rotary furnace. Heated to 650 °C, the cans melt and blend in with the molten metal already in the furnace. The reclaimed aluminium can be used as new material without loss of quality, and much less energy and water is used than in the production of new aluminium from bauxite.

Cans or tins made of tin plate consist of 99,8 % steel with a wafer-thin layer of tin. The recycling of used tin plate cans is possible with easy processes resulting from the unique magnetic properties of steel. Tin plate can also be recovered from mixed waste without problems. According to the latest reuse statistics of the "Duales Systems" German, households collected approx. 188,000 tons of tin plate in 2007 [3]. In Europe the recycling rate of tin plate cans was 66 % in 2006 [5].

As a consequence of the high temperatures of all metal recycling processes, adhesives made of organic polymers, used for labeling cans or for bonding any kind of metals in construction and transportation, incinerate and so make possible trouble-free recycling.

4.3. Energy recovery

Burning organic waste is another possibility to save resources. Taking into account that more than 90 % of crude oil is used for energy production, there is a great value in burning organic waste to utilize energy. In principle this can be done with plastic, wood, or paper and board waste. This kind of recycling is especially useful if different kinds of waste are mixed and therefore in material recycling the quality of the secondary material would be worse. Plastics especially continue to decouple growth in demand and material to landfill, both by energy recovery and by material recycling. Adhesives made of organic polymers incinerate and contribute to the energy recovery of waste (cf. Figure 1) [6].
5. Adhesives in low temperature recycling processes

In recycling processes which run at high temperatures, adhesives composed of organic materials show no negative influence. However, in recycling processes that operate at room temperature or just slightly above room temperature, adhesives can cause problems in the process or impact the quality of secondary raw materials. The largest-quantity, most important recycling process that takes place at low temperatures is the recycling of cellulose paper, paperboard and cardboard, and the mechanical recycling of plastics used in packaging.

5.1. Paper recycling

In spite of synthetic packaging materials and the rapid growth of electronic media, paper and board consumption is increasing steadily. While in 1950 about 50 million tons of paper were produced world-wide, in 2005 approximately 366 million tons were produced. In the year 2010 four hundred million tons are projected. More than two thirds of this paper is consumed in Europe, Japan and the USA, where only one fifth of the world population lives. To make this increase in paper production possible and for saving resources at the same time, paper recycling has intensified steadily in the last decades and has now reached a high technical level.

Most products made of paper have a life span of only a few days (eg, newspapers) or a few weeks (eg, packaging). Therefore it is not striking that the thought of recycling has been a firm component of paper production for a long time. As early as the 13th century waste paper was reused. Not only have the
technical conditions for recycling changed in the following centuries, but also the reasons for recycling. At the beginning, suitable raw material for producing writable material was scarce and the use of waste paper was largely determined by economic interest. Especially in countries with not much wood the consumption of wood could be reduced and the forests could be saved. Today the economic advantages are largely fully exploited. The increase of waste paper use in industrialized countries is determined by problems of disposal. In the sense of resource saving, the recycling of used cardboard packaging materials and other papers is a further example in accordance with the idea of sustainable development. It is a prime example for treating all resources including the renewable ones as carefully as possible. Paper recycling in Europe increased markedly throughout the 1990s. The amount of paper collected and recycled at the end of the decade was roughly two thirds more than at the beginning. This means that the recycling rate (percentage of recovered paper use compared to total paper consumption) was 64,5 % in 2007, compared to about 33 % in 1995 according to the “Monitoring Report” of the European Recovered Paper Council. A total of 58,2 million tonnes of paper and board were recycled in Europe in 2006 and more than half of the paper used in Europe today is now made from recovered fibre. Recycling is a significant part of the paper manufacturing process in Europe but also a large industry in its own right, with links to a number of sectors in the global economy. Building on the success of the initial “European Declaration on Paper Recovery” launched in 2000, which was responsible for pushing Europe’s recycling rate to 64,5 %, the new declaration launched in 2007 covers more European countries, more European organizations and has even greater ambition. The European sectors
have now joined forces with the common goal of further increasing Europe’s recycling rate to 66 % by 2010 (cf. Figure 2) [7]. The new target would mean that some two tonnes of paper is recycled in Europe every second. Twelve different sectors in the paper value chain have pledged their support for the declaration covering all paper and board products, and all aim to make sure that correct systems are in place to push European paper recycling rate even higher.

Worldwide, the use of waste paper also increased considerably, from about 85 million tons in 1990 to about 188 million tons in 2005 [8].

### 5.1.1. Paper recycling processes

The prime objective of waste paper recycling is to utilize the fibres contained in pre-or post-consumer waste paper. Non-fibrous components, whether they derive from paper or are added during the processing or the use of paper, should be removed at the highest degree possible to avoid quality defects in the produced papers (specks, holes) and production process faults (e. g., wet web breaking). If you take a closer look at paper and paper products, you notice that even simple papers and boards do not consist solely of cellulose fibres. They contain many additives that guarantee the particular use properties of the papers. In the paper mill many papers are coated to improve surface properties. In processing, most graphic and packaging papers are printed afterwards and then partly varnished or coated. Due to the large number of materials that get into contact with paper during its life cycle, there are very different impurities. Regarding disturbances in production, especially thermoplastic impurities (stickies) must be mentioned. At typical drying temperatures of 80 °C to 120 °C, many thermoplastic substances get soft and sticky, and so lead to problems in the paper machine drying sections. Thermoplastic deposits can also show sticky properties at or only slightly above room temperature, depending on their glass-transition temperature. In principle all non-paper components that can form sufficient adhesion and cohesion can be a source of sticky impurities (eg, resins from wood, coating binders, inkbinders, coatings, impregnation, adhesives). To fulfill both these requirements, stickies have to be liquid or at least must be soft enough to form sufficient adhesion bonds. At the same time the particles must be big enough and must have enough cohesion to achieve noticeable effects. Thermoplastic particles (stickies) big enough to achieve noticeable effects (sufficient adhesion and cohesion) get into the paper machine principally in two ways: particles come in from the waste paper to the drying section already big enough; or they agglomerate during the recycling process, forming large particles from small ones.
5.1.2. Adhesives in paper recycling

Most products made of paper and paperboard are put together then with the help of adhesives to form complex finished products. As adhesives play an important role in these products, it is not surprising that paper and packaging adhesives have a dominant share of the adhesive market (cf. Figure 3) [9]. If we talk about the influence of adhesives in paper recycling, first it is necessary to make a distinction between external and internal paper recycling.

![Figure 3. German adhesives marked.](image-url)

5.1.2.1. External paper recycling

Adhesives have always been a decisive factor in the manufacture of packaging materials made from paper, board or cardboard and for the production for graphic art articles. From the beginning of the 20th century, paper, a natural product, was bonded with adhesives based on natural raw materials such as proteins, starch or cellulose. Today, a wide variety of different materials are used in the packaging industry, and many requirements can only be met by synthetic adhesives. External recycling (post-consumer waste) is the recycling of external accumulated paper waste, primarily from used packages or used newspapers. Whereas printing inks are removed by flotation, the most important removal process for most of the other impurities is sorting. If you look at the post-consumer paper recycling process in detail, you will notice that it is primarily a mechanical process (supported by heat and alkali). In this process one tries to weaken the composite of the cellulose...
fibres by applying mechanical power, so that only single cellulose fibres remain. The mechanical power in a pulper or in refining drums is chosen so that it allows fast but careful fibre isolation. These single fibres can be used for new paper (cf. Figure 4).

Therefore, for external recycling paper recyclers demand: ‘Non-paper components should be dimensioned and mechanically stable in such a way that they survive as large particles, without being comminuted, in the conditions of pulping and allow mechanical separation by means of punched screens, slot screens and centrifugal purifiers. Relevant examples are cover foils, staples, thick adhesive layers, various product samples. Materials applied in very small dimensions or disintegrating into very small parts are unfavourable, because they can not be removed using today’s conventional sorting methods. Recovered paper components which dissolve in the process under standard conditions of deinking (pH=8–10) and reach the process water pose a risk of unintended spreading to all parts of the paper machine. This results in the requirement that recovered paper should contain as few components as possible which dissolve or disperse in a weakly alkaline medium and form sticky residues or cause discoloration [10].’ To fulfill these requirements the paper mills that produce paper or cardboard out of waste paper have lavish cleaning systems (sorting machines and for graphic papers also deinking systems). After defibrating, the suspension passes through several successive cleaning systems in which impurities are separated by their density, size or shape. Today slotted screens with a slot width up to 0.15 mm are considered most effective as far as sticky removal is concerned.

![Figure 4. Paper recycling process.](image-url)
Modern recycling processes in paper mills today allow the sorting of big and compact (>0.2 mm) thermoplastic impurities. In spite of the most modern technology it is not possible to remove water-soluble or very small dispersed particles from the water loops by sorting. Although these particles are so small that you would not expect them to cause any harm, they may agglomerate during the process and so grow to bigger particles (the so-called secondary stickies) that then cause the well-known disturbances in the paper machine or the well-known quality flaws of the finished product.

Whether an adhesive film is mechanically stable enough to withstand undamaged the paper recycling process, so it is not torn into small pieces which pass all sorting facilities, depends on its inherent strength and on its geometry. Furthermore it has to be considered that the cohesion of all materials decreases as temperature rises. The same adhesive will behave completely different when the adhesive film shows different geometry or the recycling process takes place at different temperatures. Thick, compact adhesive films will normally not be broken at moderate temperatures. However the wish for thick adhesive layers during processing of paper and board often conflicts with the wishes of the paper converter. In general one tries to apply as little adhesive as possible, if only for cost reasons. In addition, modern computer-controlled nozzle systems can optimize the adhesive application. There are today many adhesive application systems that apply single points (up to several hundred separate adhesive points per second) and therefore lead to an extreme cost-saving and clean application of adhesive (cf. Figure 5). Next to the saving of adhesive and efficient production, such systems also have ecological advantages. Closed systems need considerably less cleaning expenditure and in the case of water-borne adhesives very little waste or waste water is produced. Even with mechanically stable, non-water-soluble adhesives, there is the danger that these films created out of small droplets are so thin that they cannot be sorted out in the screening machines of the paper recycling mills. Moreover, very thin films and high temperatures reduce the power that can be absorbed by the adhesive film, and it is easily torn into very small parts.

The best classification for adhesives regarding their influence on paper recycling is the general view of adhesive film properties, because in waste paper the adhesive exists as a set or cured film (cf. Figure 6) [11]. In general an adhesive film should have a higher inherent strength than the substrates to be bonded, for example a much higher inherent strength than paper, cardboard or carton or composites (fibre tear), so that it is more easily separable. Non-water-soluble adhesives which are characterized by a high glass transition temperature show enough cohesion in a sufficient layer thickness to survive the pulping process without damage. The problem is
more complex regarding water-soluble or re-dispersible adhesives, as the environment in the recycling process contributes to the weakening of the mechanical stability of the adhesive film. At the predominant pH-values and temperatures of the aqueous environment in the waste paper mills, considerable stress is put on films that contain hydrophilic groups. This can lead to the complete destruction of these films, as their cohesion gets lost completely. By a good choice of raw materials, however, water-borne adhesives can be formulated in the way they are not torn into such tiny particles in the recycling process, that they remain mechanically sortable (cf. Figure 7). Figure 8 shows the adhesive particle distribution of two water-borne pressure sensitive adhesives (PSA) coatings after the repulping of labels. As you can see clearly, adhesive film A is torn into much smaller particles than adhesive film B. The large particles of adhesive film B can be removed from the cellulose suspension nearly completely by mechanical sorting, as tests have shown. The particle sizes given in Figure 8 are not the real size of the three-dimensional adhesive residues, but sizes that were found in a picture-analytical method. These are two-dimensional measurements that were converted into circles of the same size. From these circles the diameters were calculated [12].

Figure 5. Computer-controlled nozzle application systems.
Figure 6. Systematic of adhesives films.
Figure 7. Mechanical separation of adhesives depending on the adhesive film thickness.

Figure 8. Mechanical separation of adhesives.

5.1.2.2. Internal paper recycling

Internal paper recycling (pre-consumer waste) describes the recycling of production waste within a paper mill with a processing line on site. An
example is tissue and towel mills where paper goes directly from the paper machine to rewinders for the production of bathroom tissue or paper toweling. During this processing, adhesives are used for laminating, for the pick-up of the first sheet on the tube, and for the end sheet tiedown, and the waste, or ‘broke’, created here must be returned to the paper mill. Here, in general, only a relatively small amount of rejects are moved back into the paper production process. In contrast to adhesives that get into paper mills by external recycling and have sources varied and unknown, in internal recycling there is only a relatively small amount of adhesive and the types used are known exactly. These mills normally do not have lavish sorting machines. As the additives added to the paper in production cannot be sorted out mechanically, most of the time the additives are required to be completely water-soluble or re-dispersible, even if this pollutes the process water with impurities. Adhesives that are used in this production are normally classified by the European Standard EN 1720 “Adhesives for Paper and Board Packaging and Disposal Sanitary Products Determination of Dispersibility” or by the American TAPPI standard UM 666 “Dispersibility Test for Adhesives”. Today there are many adhesives that fulfill the requirements.

Adhesives based on polymers like polyvinyl alcohol, polyvinylpyrrolidone and its copolymers, polyethyloxazoline, copolyesters containing sulphonated material, hydrophilic polyurethane and polyethyleneoxide, but also adhesives based on starch, dextrin and cellulose show good water-solubility or re-dispersibility [13]. In this connection one has to emphasize that soluble or re-dispersible additives in paper mills can lead to two problems: a contamination of the process waters that leads to more waste water pollution; and future agglomeration of dissolved or re-dispersed additives. If they are thermoplastic materials, so-called secondary stickies can be formed.

5.2. Adhesives in plastic recycling processes

Bonding is an ideal joining system for plastics to themselves or to other materials, and in the field of packaging many examples can be found. The plastic material with the strongest increase in growth rate in packaging is polyethyleneterephthalate (PET). Light weight, transparency and flexibility characterize this perfect packaging material. The success of PET and especially of PET bottles results from the existence of successful recycling systems, some of which allow a closed circulation from bottle to bottle. Depending on the process the recycling material is sorted, crushed, cleaned and separated from secondary products in several steps. Some processes at the end provide a recycled material that is approved by the American
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watchdog FDA for contact with foodstuff and fulfills the greatest demands regarding purity and quality. With this favourable recycled material new preforms can be manufactured for bottle production and so the circuit is closed. In 2007 about 1,200,000 tons of PET bottles were collected for the different recycling processes in Europe [6]. In addition to the production of new bottles (15.2 %), these used bottles also become fibres for clothes, carpets, PET sheets and binders.

Compared to glass and metal can recycling, adhesives that are used for labeling of PET bottles have to be adjusted to the recycling process of PET. Depending on the recycling process, alkali- or water-soluble labeling adhesives are used for disposable bottles as these make possible an easier cleaning of the bottle before recycling. This feature is especially important when selecting a hot melt adhesive, because hot melts normally consist of non-water-soluble thermoplastic polymers. Water- or alkali-soluble or re-dispersible hot melts developed especially for labeling of PET bottles fulfill all recycling requirements. To reduce the effort during the recycling process, it is important to apply only the minimum amount of adhesive necessary. Special application systems for hot melt adhesives exist which spray only a very light quantity of adhesive onto the bottle for the pick up of the label (cf. Figure 9). Hence, when selecting a labeling adhesive the later recycling of the container should be considered right from the beginning.

In recycling, the recycled waste should be as mono-materialistic as possible so that the secondary materials gained are of high quality. Recycling of the plastic base materials is also possible if the quantities and the material of impurities are below a particular level that can be identified very precisely experimentally. Although normally the amount of adhesive applied is very small compared to the parts bonded, a mono-material recycling can be guaranteed even with small amounts by a smart choice of the material of the bonded parts. Examples are pressure sensitive labels used in the automobile industry or electronic industry. These labels are expected to stick to the surfaces of plastic parts safely for decades, but do not disturb the later material recycling of the construction parts. They are made of the same chemical material as the body or some other material that does not cause a quality loss in recycling of the body material. When using such labels, the primary material can be recycled trouble-free without causing quality loss [14].

If plastics undergo a chemical recycling process (decomposition of the polymers into their monomers by depolymerization) small amounts of other organic substances do not disturb, as they can be removed from the monomer stream.
5.3. Recycling of multilayer packaging material

Beverage cartons for short-life goods are made of multilayer layers of cellulose and polyethylene (PE). For long-life goods, there may be an additional aluminium layer between the cellulose and PE layers. These thin materials – about 0,4 mm carton, 0,06 mm PE and 0,0065 mm aluminium – fulfill all requirements for beverage packaging. Recycling of beverage cartons is similar to paper recycling and so can be done without much energy. Not only the high portion of paper (75-80 %), but also the remaining materials polyethylene (21 %) and aluminium (4 % – used only for durable products) are returned into material circulation, which makes good economic sense, too. In 2006, more than 313.000 tons of beverage cartons were collected for recycling in Europe (151.000 tons in Germany). This corresponds to a recycling rate of about 30 % in Europe and 65 % in Germany [15].

In closed pulpers equipped with bars the cartons are crushed, which lets water seep into the carton and makes possible the dispersal of the fibres in a water bath without further additives. In very short time, the cellulose fibres of the carton can be screened out as pulp. Because of their good strength, recycled fibres from beverage cartons are very much in demand. They are used for example for corrugated board and tubular cores, which are needed for rolling up paper, fabrics or carpets. There are several processes to recycle the remaining polyethylene and aluminium foils usefully and at the same time save primary raw materials. These polyethylene and aluminium residues can be used for energy and material utilization in the cement industry. Polyethylene plastic can be used there as a substitute for organic energy sources like oil. Aluminium oxide forms at a temperature of more than 1,450 degrees and can be added to the cement as a necessary loading, so that the cement can solidify faster. In this process, the aluminium residues substitute for some of the primary raw material bauxite. The complete utilization of residue components of beverage cartons - PE and aluminium – can be made with another elaborate process: In a pyrolytic reactor polyethylene is converted at a temperature of 400 °C into process steam and electrical energy. The aluminium is separated as a regained mono-material that can be returned to the aluminium industry in the form of bars.

Adhesives like hot melts, used for attaching pour spouts (fitments), do not disturb this process as they can be sorted out easily and can be utilized for their energy content, too.

6. Disbond on command, adhesives with “switches enclosed”

Durable goods made of plastic or metal are often a mixture of different materials. In recycling, a separation of the elements is useful, and that separation
should be as mono-materialistic as possible so that the secondary material gained are of high quality. Basically almost all bondings can be removed relatively easily, often by the use of high temperatures, but cost and possible damage to the primary material must be considered. Just as time was a factor in the original bonding process, so is time also a factor in their later separation.

One possibility of "dibond-on-command" is to put energy just into the bonded joint to melt, dissolve, or destroy the organic bond line. This might be especially advantageous on pieces where it is not appropriate to heat the entire piece, or on synthetic materials which would be affected by heat.

However, with intelligent solutions systems can be found that combine the advantages of bonding with an easy dismantling or an easy recycling of the primary material. One idea in this direction is the use of nanoparticulate ferrites, which pick up energy from electromagnetic AC-fields, convert it to heat and pass it on to the immediate surroundings of the adhesive layer. This enables a fast, targeted and locally defined energy uptake and thus the "switching" of properties in the surrounding matrix (e.g. polymers). Requirements for a fast debonding with simultaneous intrinsic overheat protection include a tailored combination of ferrite composition, magnetization, solids content, particle size and particle dispersibility in the adhesive [16]. Another possibility to develop adhesives with "switches enclosed", are adhesives based on binders with di- or polysulfide bonds in the polymers. The joints bonded by these adhesives are debonded by activating the debonding components through melting, conformation changing or splitting off the protective groups. The activation may be done through the activity of conductive heating, radiation heating, particle radiation, elec. current, elec. or electromagnetic field, ultrasound, pressure wave, or impulse wave. Adhesive containing room temperature stable, thermally activatable components, which cause swelling of the binder matrix, phase transition, decomposition or generation of gases or water vapor are also suitable for separation of adhesive bonds under heating, which permits complete recycling of the separated parts. As thermally activatable components malonic, oxalic, adipic, citric, glutaric, ascorbic, or benzoic acid, as well as solid diols, or azo components can be used [17].

Conclusions

The recycling of all used products, but especially of mass product articles like packaging or graphic arts, is economically as well as ecologically useful. Legislation in Germany and throughout Europe has contributed to raising to a high level the degree of recycling of all kind of packaging made of glass, metal, plastic, paper and cardboard. To increase this value continuously and to keep recycling more cost-effective, it is necessary for all involved to consider the later recycling of these products.
Since many products are often a mixture of different materials which are bonded with the help of adhesives, it is important to choose adhesives that do not disturb the recycling of the primary materials. In recycling processes which take place at high temperatures the influence of adhesives that are based on organic polymers can be ignored. In low-temperature recycling technologies, the question whether an adhesive is recycling-friendly or not can only be answered by knowing its application and the recycling process. If the recycling processes are known it is easy to choose suitable adhesives. For plastic and paper recycling there are a lot of adhesives today which fulfill the requirements of recyclers.

Concerning the influence of adhesives on the recycling of primary materials there are no special demands for glass, aluminium and tin plate. However, in plastic recycling the requirements of the later recycling processes have to be considered when choosing adhesives. Often water-soluble, lye-soluble or re-dispersible adhesives are demanded. Special developments in the labelling adhesive sector fulfill these requirements. There are systems for beverage carton closing with adhesives that allow trouble-free recycling of the cardboard packaging materials. The right choice of hot melts and a suitable application thickness, or the right choice of adhesive tapes, leads to systems that can be considered recycling-friendly. As demanded by the paper recyclers, an easy and quantitative sorting-out of these systems is possible directly at the beginning of the paper recycling process. Thus adhesives that get into the recycling process do not lead to problems.

In paper recycling a lot of adhesives can be sorted out easily. The ability to sort an adhesive film in the recycling process is not only defined by the properties of the adhesive, especially its cohesion, but also by its geometry. Another influential factor is the fixing of the adhesive film to the substrates. Furthermore, the characteristics of an adhesive film are influenced enormously by the pH-value and the temperature of the water in which the recycling process takes place. To be able to draw a really useful conclusion, the only way is to test the finished paper product. In the finished paper product the adhesive film is what it will be like when the product is later recycled. The influences of geometry and substrates can be tested exactly. Only tests on finished products can indicate whether the finished product or components of the product (e.g., adhesives, but also coatings, inks or other additives) influence recycling or not.

In plastic recycling a lot of adhesives are alkali- or water-soluble and therefore make possible an easier cleaning of plastic articles before actual recycling. Other adhesives may be made of the same chemical material as the plastic article and thus not cause a quality loss in recycling of the plastic material.

To simplify future recycling, adhesives have been developed with “switches enclosed”, which allow the system components to be de-bonded into separate parts after use.
References