Review

Recycling of Aseptic Beverage Cartons: A Review

Gordon L. Robertson

School of Agriculture & Food Sciences, University of Queensland, Brisbane, QLD 4072, Australia; g.robertson@uq.edu.au

Abstract: Aseptic beverage cartons are multilayer polymer-coated paperboards with a layer of aluminum foil. Due to their multilayer structure it is commonly assumed that they cannot be recycled. This is not the case and this review details the multifarious processes that are used to recycle aseptic beverage cartons. Hydrapulping to recover the paper fibers that constitute 75% of the carton is the most widespread process, followed by the manufacture of construction materials such as boards and tiles which utilize the complete carton. A range of mechanical, chemical and thermal processes are used to separate the PolyAl (polyethylene and aluminum) residual that remains after the paper fibers have been recovered. The simplest process involves agglutination followed by extrusion to obtain pellets that can then be used in industrial and consumer products or combined with other materials such as lignocellulosic wastes. Chemical approaches involve the solubilization of polyethylene and the removal of aluminum. Various thermal processes have also been investigated and a novel microwave-induced pyrolysis process appears the most commercially viable. It is concluded that the focus in future years is likely to be on recycling cartons into construction materials where there is a theoretical yield of 100% compared with 75% for hydrapulping.

Keywords: aseptic cartons; hydrapulping; construction board; microwave-induced pyrolysis

1. Aseptic Carton Construction

Aseptic LPB (liquid paperboard) beverage cartons are multilayer polymer-coated paperboards with a layer of aluminum foil and play an important role in the delivery of shelf-stable milk and juice products to consumers around the world. The first aseptically filled sterilized cartons containing sterilized milk were sold in Switzerland in October 1961 [1]. Today four companies supply almost all the aseptic LPB beverage cartons to the global food industry: Tetra Pak, SIG Combibloc, Elopak, and Greatview Aseptic Packaging Company.

The typical structure of an aseptic carton is shown in Figure 1. An LDPE (low density polyethylene) outer layer provides a moisture barrier, protects the printing ink layer applied to the paperboard and enables the package flaps to be sealed. The type of paperboard used depends on the product being packed, the regional market where it will be sold and the manufacturing conditions, but it is commonly a duplex or triplex material with a bleached or clay-coated outer layer and often contains CTMP (chemithermomechanical pulp); the paperboard gives the package the required mechanical rigidity and typically represents 75% of the total weight of the package. As there is no contact between the paperboard and the liquid product inside the package, no wet-strength additives are used in the paperboard. The inner side of the paperboard is coated with LDPE to tie it to the aluminum foil (generally 6.35 μm thick and constituting 5% of the total weight of the carton) that provides an odor, light, and gas barrier. Adhesion of the aluminum foil to the innermost plastic layer is achieved through the use of a layer of EMAA (poly(ethylene-co-methacrylic acid) which has rheological properties similar to those of LDPE [2,3]. Finally, an inner layer of LDPE is applied to enable heat sealing of the carton.
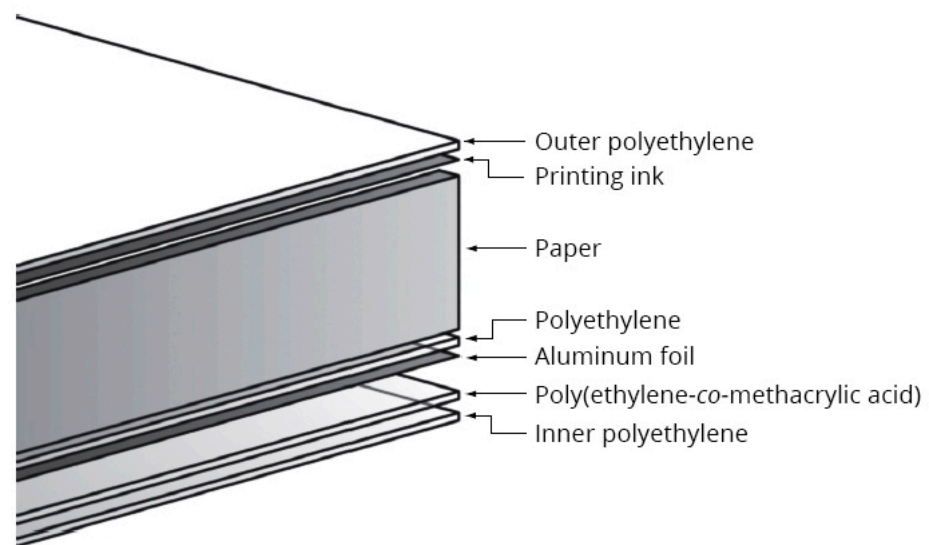
Figure 1. Typical structure of an aseptic carton.

Since 2006 Tetra Pak has used mLLDPE (metallocene-based linear low-density polyethylene) that is 30% thinner than the previous LDPE coating [4]. The roll-fed carton material supplied by Tetra Pak and Great View Aseptic requires a longitudinal seal strip to be applied internally to prevent contact between the aluminum and paperboard edge and the product; this is not required for preformed carton blanks. The strip is composed of a core layer of PET (polyethylene terephthalate), PA (polyamide) or EVOH (ethylene vinyl alcohol copolymer) depending on the oxygen sensitivity of the product, coated with LDPE or LLDPE [5]. The carton may also have an opening device or screw cap made from HDPE (high density polyethylene).

2. Recycling Options

Recycling options can be categorized as primary, secondary, tertiary, and quaternary [6,7]. Primary recycling (sometimes referred to as closed loop recycling) is reprocessing the material back into its original use or comparable products with equivalent quality but is not an option for post-consumer cartons as they cannot be directly converted back into their original use. However, secondary recycling where materials are processed and used in applications not requiring virgin material properties is the most widespread recycling option for aseptic cartons. The paper fibers are separated from the PolyAl (polyethylene & aluminum) and incorporated into paper products; this has been a relatively common process for at least the past 40 years. Another secondary recycling process involves converting the shredded cartons into construction materials. Tertiary recycling involves breaking a product down into its chemical building blocks, and then recycling those chemicals into various products. Quaternary recycling of cartons involves incineration with energy recovery although this process is not considered recycling in many countries. Mechanical, chemical and thermal processes are used to separate the polyethylene and aluminum in the PolyAl residual that remains after the paper fibers have been recovered. The recycling options for aseptic cartons and the PolyAl residual will be considered in this review.

Reliable figures on the recycling rate of beverage cartons are difficult to obtain but a 2019 figure of 51% is given for the 28 countries of the European Union, with some countries such as Belgium and Germany achieving rates of over 70%, according to The Alliance for Beverage Cartons and the Environment [8], an industry group for European beverage carton manufacturers and their paperboard suppliers. The recycling figure is calculated using the method of accounting for the predominant material as specified in 2005/270/EC: Commission Decision of 22 March 2005 establishing the formats relating to the database system. From 2020, a new calculation method came into effect which requires deduction of

non-recycled materials and reporting of the recycling rate for each material separately. In the USA the Carton Council [9] reported a curbside recycling rate of cartons of about 18% in 2019.

2.1. Recovery of the Paper Fibers

As it constitutes 75% of the total weight of the carton, recovery of the paperboard has been the predominant focus of carton recycling approaches. Recycling is accomplished at a paper mill by recovering the paper fibers using a conventional hydrapulper (so named because of the hydraulic forces generated during pulping). Hydrapulpers are large cylindrical vessels with impellers (rotors) at the bottom which break apart the paper fibers and produce a relatively dilute slurry of fibers that can be further processed within the mill. Batch pulpers range in size up to 7.6 m in diameter (up to 68 m^3 stock volume) with capacities up to 14.5 tonnes of dry furnish (cartons) per batch. Typical batch times are 30 to 45 min at ambient temperatures. Continuous pulpers range in size from 3.7 to 8.2 m in diameter (up to 147 m^3 stock volume) with capacities up to 1270 tonnes per day [10].

Contact between the water and the paper layer occurs in the hydrapulper, and the layers separate due to the hydraulic forces inside the pulper. No chemicals are added. The pulper has a drilled plate underneath the rotor, with 9.5 mm holes for fiber/water extraction. There is no automated consistency control, but 12 to 15% is the desired range of operation. Hydrapulpers are generally equipped with a ragger which removes the PolyAl residual, caps, straws and long stringy materials such as baling wire from the slurry [11].

A 1993 patent assigned to Tetra Laval [12] detailed the methods and apparatus for separating paper fiber from mixed waste materials which contain one or more sources of paper fiber, such as beverage cartons, in order to obtain substantially pure paper fiber which can readily be recycled to high grade end uses such as for paperboard having, for example, high brightness and low dirt content, as opposed to low grade end uses such as for tissue paper.

However, recycling a material that results in 25% of residuals (the PolyAl residual and any other polymers including caps, lids and straws) is not usual and is considered a negative by most paper mills. If a batch pulper is used, it must be emptied and cleaned after every batch; blending small quantities of cartons with regular wastepaper will minimize the number of times the pulper will require cleaning but blending is not always a viable option. After removal from the pulper, a perforated rotating cylinder with water showers helps wash the PolyAl residual and recover any entrained fibers (generally 0.5% of fibers are lost with the residuals); the white water is reused. The recovered fibers from the pulper and cylinder are used in the production of paper products such as tissues, towels and writing paper [11].

Recently the recovered fibers were used as a structural filler together with the biobased and biodegradable polymer PBS (poly(butylene succinate)) in the preparation of biocomposites that could be an alternative for conventional fossil polymer-based wood/plastic composites [13]. High filler loading from 10 to 50 wt% of fiber was used in the melt blending process to prepare five composites that were 55 to 75% biobased. The addition of 50 wt% of fiber to PBS gave a 2-fold improvement in the hardness of the composite compared to neat PBS. The Young's modulus increased almost 3-fold, while TS (tensile strength) and ductility decreased correspondingly. Dynamic mechanical analysis showed that the storage modulus and the loss modulus were significantly enhanced in the temperature range of −70 and 70 °C. The composting in soil of PBS and PBS/fiber films showed that the presence of cellulose accelerated the degradation of PBS resulting in decreased degradation times for the composites. It was suggested that the PBS/fiber composites could be used in industries like packaging, furniture and construction.

2.2. Construction Materials

There have been several attempts over the years to utilize the production/factory waste generated in the manufacture of beverage cartons. In 1970 Downs & Schmitt [14] filed

a patent on behalf of the Thilmany Pulp & Paper Company in Kaukauna, Wisconsin for an apparatus and method for continuously forming a composite board from a mass of shredded paper containing thermoplastic material such as that produced in the manufacture of gabletop beverage cartons (paperboard and LDPE). In the mid-1980s Tetra Pak started a project in Lund, Sweden that led to the first production site being built by Tetra Pak in Kenya in 1987 to produce composite board from the local aseptic carton post-production factory waste; a patent for this process was filed by Nilsson in 1989 and assigned to Tetra Pak Finance & Trading [15].

In 1990 EVD (Entwicklungsgesellschaft für Verbundmaterial Diez mbH) was founded in Diez, Germany by Tetra Pak to manufacture a composite material named Tectan® (also the name of fluorocarbon fishing lines sold by Damyl in Germany). In a specially developed, patented process [16], the raw material (mainly post-production beverage carton factory waste) is shredded, dried and then converted into granules. Depending on the designated end use, polyolefins such as LDPE, HDPE or PP (polypropylene) are then combined (according to a formula that delivers the requisite end-product properties) with the paper fibers serving as stabilizers and increasing the rigidity of the plastic matrix. The resulting Tectan® composite material is compression or (since 1997) injection molded to make core plugs and edge protectors used by paperboard manufacturers. One application that created considerable interest from an ecodesign viewpoint was Sony loudspeaker cabinets made from Tectan® that delivered superior acoustic performance [17]. In 1997, Tetra Pak decided not to continue running EVD in its original form, and the employees who had been involved in the development of Tectan® took over the company.

Today composite board (also referred to as panel or particle board) is the most widely produced construction material made from aseptic cartons, and is produced by shredding aseptic cartons into 1 to 5 mm sized particles and thermally compressing them at 170 °C for a time dependent on the board thickness. The composite board obtained is comparatively light and water-resistant with varying levels of flexibility. Lower quality polyolefins are sometimes added since the quantity of plastic in the carton material (20%) is not always sufficient to guarantee the desired mechanical properties. Although post-consumer cartons are used to manufacture this board, the most suitable raw material is post-production factory waste, as contamination from impurities and residual product in post-consumer cartons, coupled with high ambient humidity in many cases, can limit applications for the board. There may also be a high microbial load that might be of concern not only in the final product as a residual odor, but also for the safety of workers when handling this material in the processing line. The time in the hot press varies with the final board thickness but a general rule is to press for 1 min per mm final board thickness (10 mm board = 10 min press time). The board is then transferred to a colder press (>80 °C to allow plastic deformation at a relatively low pressure) to obtain the specified final thickness. It is also important to have cooling of the press and the board temperature should be about 20 °C when leaving the press to avoid creep effects [18].

Many mostly small-scale factories to manufacture composite board from aseptic cartons have been built around the world over the past 30 years with brands including T-PLAK® in Argentina, Reciplak® in Brazil, Tetrabuild® in Chile, Chiptec® in China, Ecoplak® in Colombia, Novapak and PackWall in Czechia, Greenpak in Ecuador, Ecolink in India, Lamiboard in Kenya, saveBOARD in New Zealand, Green Board in Pakistan and Thailand, TeRO® in Poland, Tetra K1® in Slovakia, Maplar® in Spain, Yekpan® in Turkey, and Everboard and Kelly Green Products in the USA. Some process only aseptic cartons while others also include plastic-coated paper products such as cups. Not all of these factories are still operating, and current production volumes are in most cases negligible in comparison to the quantity of aseptic cartons sold in each country [5].

Peña et al. [19] studied the influence of factors such as temperature (140 to 160 °C), pressure (11,000 to 13,800 kPa), heating time (25 to 30 min) and composition on the final properties of boards made from aseptic cartons. They concluded that the use of higher temperatures, times and length of carton pieces accompanied by lower pressures may

improve the TS of the panels. Based on the contribution of this study, the authors concluded that the optimal values of the variables for the commercial production of the panels remain to be found. However, commercial operators, often using a trial-and-error approach, have in most cases already optimized their processes.

Quintero et al. [20] investigated the possibility of using board made from aseptic cartons to create structural solutions for temporary housing and small houses. Hollow column and beam models made from board of thickness about 15 mm helped in understanding theoretical behavior and determining the areas where stresses and deformations occurred. Tests focused mainly on compression and bending with results showing that although the board had low elastic behavior, stresses remained below the ultimate stress. Column failure tended to be brittle compared to failure for the bending resistant elements. Failure loads were similar to those reported for commercial plywood in Colombia.

There have been many reports on the properties of composite boards formed from aseptic cartons, and researchers have attempted to improve board properties using a variety of approaches and additives; these are reviewed below.

2.2.1. Veneers

In order to add value or enhance the appearance of the panels, various veneers have been applied. Ayrilmis et al. [21] evaluated aseptic carton panels overlaid with beech veneer using four types of adhesives: PU (polyurethane), PF (phenol formaldehyde), UF (urea formaldehyde) and MUF (melamine-urea formaldehyde). The panels overlaid with veneer using PU adhesive had the best mechanical properties and water resistance. Wood veneer-faced carton panels had significantly higher mechanical and physical properties than those of wood veneer-faced particleboards, due to a tighter profile structure and higher density of the carton panels. The absence of formaldehyde in carton panels was a positive feature. Based on the findings of this study, it was suggested that carton panels overlaid with veneers could be considered as an alternative raw material with acceptable properties to be used in furniture manufacture, counter tops, flooring, roofing, dividing walls, and kitchen cabinets.

In a later study, Sen et al. [22] reported that wood veneer-faced carton panels had significantly higher antifungal and insecticidal properties than the wood veneer-faced particleboards with the carton panels having a toxic effect on larvae. The results revealed that carton panels with wood veneer could be used in high humidity conditions such as bathroom furniture and roofing material which are exposed to fungus and insect attack.

Rhamin et al. [23] produced "carton board" with a density of 1 g cm^{-3} from aseptic cartons by mixing MUF resin at three levels with two different pressing times (10 and 12 min). Half of the boards were created using walnut veneer layers. Physical and mechanical properties of the boards were evaluated. The different press times and resin levels did not affect the mechanical properties of the board, but the walnut veneer significantly increased the mechanical properties and reduced the physical properties such as internal bonding strength and screw withdrawal resistance. Boards created without using any resin had better physical properties.

2.2.2. Addition of Plastics

Murathan et al. [24] prepared panels of shredded aseptic cartons to which UF or PVA (polyvinyl acetate)-based glue was added in different ratios; panels containing the former exhibited better physical properties than the latter. It was suggested that these materials could be placed behind radiators or electrical heaters to prevent heat loss but they were not considered suitable for use in high humidity environments.

Parada-Soria et al. [25] investigated the thermal and mechanical properties of composites made from multicolored recycled HDPE flakes that had been dry mixed with varying concentrations of aseptic carton flakes and compression molded at 250 °C. Previous studies had shown that the type of pigment utilized in HDPE strongly influenced the mechanical properties of the composites. Moreover, the degree of crystallinity and mechanical modulus

varied among different colored HDPEs suggesting the influence of the pigments to nucleate crystallites. Furthermore, for multicolor HDPE/carton composites, poor adhesion was found between HDPE samples with different colors, adding to the failure mechanism of the composites. Mechanical properties such as Young's modulus, yield stress and ultimate TS were obtained under uniaxial tensile deformation at room temperature. The results indicated that understanding the role of processing parameters would provide the opportunity to generate value-added composite materials from recycled thermoplastics.

Carrillo et al. [26] blended aseptic cartons with HDPE from recycled milk and juice containers. The cartons were first washed and dried before being ground to a sieve size of 3 mm; the HDPE particles were ground to a sieve size of 2 mm. The carton/HDPE (55:45) agglomerated material was manufactured by compression molding and formed the middle layer in a laminate. An LDPE film was applied to each side to provide a moisture barrier and aluminum film was applied over the LDPE films to facilitate removal from the press. The material was then pressed for 10 min while being heated to 150 °C at a pressure of 1.13 MPa, followed by cooling to room temperature at constant pressure for approximately 30 min. The addition of HDPE to the carton material increased the material's capacity to resist flexural loads in situations such as its application in construction and the furniture industry. Screw pull-out strength was also increased in the material, another parameter of vital importance in the aforementioned applications.

In a follow-up study, Chan-Koyoc et al. [27] subjected carton/HDPE (55:45) laminates to aging in an AW (accelerated weathering) chamber with UVB (Ultraviolet B) radiation, temperature and humidity cycles; and NW (natural weathering) in a warm sub-humid environment. The MOR (modulus of rupture) and MOE (modulus of elasticity) decreased by 20% in samples subjected to AW for 2000 h, while the samples exposed to NW required 5200 h to obtain a reduction of 16% in the MOR.

Bekhta et al. [28] evaluated some of the properties of experimental composite panels made from three types of materials: aseptic cartons, food packaging films (recycled stretch wraps), and CPEW (candy polyethylene wrappers) used at different ratios in the panels at a target density of 0.9 g cm^{-3}. The highest MOR value of 15.5 MPa was determined in the samples having 40% cartons and 60% CPEW. MOR values of the panels decreased with decreasing content of CPEW; the increased content of cartons also resulted in a reduction in their strength characteristics and dimensional stability.

In 2020 Aranda-García et al. [29] studied the main processing and formulation factors that affect the performance of panels manufactured with postconsumer aseptic cartons and HDPE (5 to 20 wt%). Their results showed that mechanical strength depended more on the pressing time than the formulation. The specimens tested by immersion in water for 17 days exhibited water absorption of 60 wt% but their structural stability was not compromised.

Guillén-Mallette et al. [30] used two quantities of milled fiber (20 and 30%) from aseptic cartons together with HDPE, mineral fillers calcium carbonate and zinc oxide, a coupling agent and two types of ethylenic copolymer processing aids. The independent variables that had the greatest influence were the type of HDPE, the type of ethylenic copolymer processing aids, and the extrusion speed. The extruded material had a density of 1.11 g cm^{-3}, temperature of fusion of 149 °C, impact resistance of 32 J m^{-1}, TS of 11 MPa, and flexural modulus of 314 MPa.

2.2.3. Blends

Hwang et al. [31] prepared boards of different specific gravities (0.55 to 0.75) by pressing shredded cartons blended with UF resin (6 to 10%) at 180 °C. Mixed particleboard containing wood particles and cartons all having a resin content of 10% and various specific gravities were also prepared. At the same specific gravity, the properties of the boards were affected by their resin content. The MOR, MOE and internal bond strength increased with increasing specific gravity at the same resin content, but thickness swelling of the boards showed the reverse trend. The average MOR of the carton boards approximated

that of the mixed particleboards, and internal bond strength and thickness swelling of the carton boards were smaller than those of the other particleboards. Based on the above observations, it was concluded that aseptic cartons can be made into composite boards with adequate properties either alone or mixed with wood particles.

Moya et al. [32] evaluated the decay resistance, coating and burning properties, and the change of color caused by AW of particleboards manufactured with a combination of three woody species used for commercial reforestation in tropical areas, pineapple leaves, empty fruit bunches of oil palms and aseptic cartons. UF resin (62% solids) was used at 6 to 8% of the total weight of the particleboard. Mixtures containing 50% cartons performed best, followed by mixtures with oil palm components and pineapple leaves. Particleboards with aseptic cartons and oil palm showed the highest resistance to combustion. Mixtures of the three woody species with cartons showed the best performance in AW.

Sun & Zhang [33] studied the optimum hot-pressing process and surface decoration of waste aseptic carton/sawdust composite board made with UF resin. The optimum process variables were resin content 14%, hot-pressing temperature 150 °C, hot-pressing time 7 min and carton/sawdust mass ratio of 4:6. The maximum MOR of the board was 23.1 MPa and the maximum MOE 2917 MPa. The optimum PVC (polyvinyl chloride) surface decoration process was a hot pressing temperature of 50 to 60 °C, hot pressing time of 10 to 20 s, and hot pressing pressure greater than 100 kPa.

Ebadi et al. [34] blended LDPE (60%) and poplar timber powder (0 to 40%) with shredded aseptic cartons (0 to 30%); in some trials 3% MAPE (malic anhydride-grafted polyethylene) replaced 3% LDPE. Analysis of the injection molded samples showed that the composites containing 30% cartons and 3% MAPE had the highest strength and tensile modulus as well as the highest impact resistance. In a 2017 follow-up study, Ebadi et al. [35] investigated the physical and mechanical properties of the wood-plastic samples with results showing that adding cartons and MAPE to samples increased the flexural strength and MOE and reduced 24 h water absorption and thickness swelling.

Hassanin & Candan [36] manufactured composite boards from shredded 100% aseptic cartons (target thickness of 5 mm and density of 0.83 g cm^{-3}) and five sandwich structures with a core of 100% cartons and skins of jute woven fabric, glass woven fabric, or PP nonwoven spunbonded fabric; and a layer of PP between the cartons and the jute or glass fiber. The MOR of the boards with different skin materials was higher than that for commercial particleboard (thickness of 10 mm and an average density of 0.66 g cm^{-3}) which was higher than the carton board without skins. The internal bond strength values of the carton board were higher than the commercial particleboard but lower than the boards with skins. Adding a jute skin layer increased the water absorption and thickness swelling of the board due to the hydrophilic nature of the jute. The thermal insulation provided by the glass fabric affected the melting of PE and PP during the hot pressing and resulted in weak internal adhesion which directly led to increased water absorption. It was concluded that the boards containing aseptic cartons had a very high potential to compete with standard commercial particleboard due to higher mechanical and physical properties and lower cost.

Nassef et al. [37] reported in 2018 that composites made from 100% aseptic cartons (target density of 0.8 g cm^{-3} and a thickness of 10 mm) and a skin of woven glass fiber fabric possessed higher dynamic characteristics in terms of damping ratio compared to commercial particleboard and medium density fiberboard. Bending tests showed competitive MOR values of both composites, with the addition of glass fiber fabric enhancing the MOR values. The 100% aseptic carton panels showed low stiffness values based on MOE results, and the natural frequencies of both composites were close to those for fiberboard and particleboard indicating comparable dynamic stiffness behavior.

Hassanin et al. [38] fabricated thermal insulation composite panels from aseptic cartons and wool fiber. Introducing the wool waste enhanced the thermal insulation properties of the final samples as air was trapped within the wool fibers. Panels made with glass fiber skin showed an 8.5% reduction in thermal conductivity compared to a 6.5% reduction

when jute fabric was used. The results indicated that aseptic carton waste can be combined with wool fiber to manufacture insulation for buildings.

Mohareb et al. [39] evaluated commercial particleboard specimens and composite specimens made of ground 100% aseptic cartons blended with different percentages of natural short fiber wool. Water absorption and thickness swelling showed a significant improvement compared to the commercial particleboard. Increasing the percentage of wool waste in the developed composites lead to a significant improvement in sample durability against fungal decay. The termite resistance of carton or carton/wool panels was enhanced compared to the pine and beech wood reference samples.

In a 2019 follow-up study, Hamouda et al. [40] reported the physical behavior, fungal decay and termite attack under laboratory conditions of composites developed from aseptic cartons and wool yarn wastes and compared the results to standard wood products. The shredded cartons were pressed with wool yarn at 190 to 200 °C for 2 min at 500 kPa and then for a further 3 min at 1000 kPa. The hybrid panel densities varied between 0.73 and 0.82 g cm^{-3}. When wool yarn wastes were increased to 15%, the MOR reached a higher value than that of commercial particleboards. The highest internal bonding strength was found for hybrid composites with 10% wool yarn content. Moreover, thickness swelling and water absorption of the fabricated hybrid composites were found to be better than commercial particleboards and they also met the minimum strength requirements of the relevant British Standards. It was concluded that aseptic cartons and wool yarn could be utilized as a promising alternative source of raw materials to manufacture value-added, eco-friendly, advanced and sustainable structural applications such as wood panels.

2.2.4. Fire Performance

Figen et al. [41] shredded aseptic cartons and then pressed them at 180 °C and a pressure of 1200 kPa for 12 min to form panels of 18 mm thickness and density of 1.09 g cm^{-3}. Samples were heated in a nitrogen atmosphere at different heating rates and the results showed that thermal degradation consisted of three distinct steps after moisture evaporation. The first step was accepted as the main thermal degradation phase (200 to 400 °C) and corresponded to the degradation of paperboard. The second step occurred between 400 and 461 °C and corresponded to the degradation of paperboard and LDPE. The last step was associated with the decomposition of the remaining paperboard and LDPE layer and the residue consisted of char and aluminum foil after the thermal degradation.

Yilgor et al. [42] evaluated some chemical, physical and biological properties, weathering and fire performance of panel board made from shredded aseptic cartons with and without zinc borate ($Zn_3B_2O_6$). Results showed that fungal degradation (mainly of cellulose) occurred in the natural polymer of the panel board but caused no changes to the LDPE. However, the LDPE seemed more sensitive to weathering than the cellulose. Incorporation of 1% (*w*/*w*) zinc borate did not improve fire performance of the panels, but a loading of 10% did using test parameters such as mass loss, ignition time and peak heat release rate.

In an effort to expand the potential applications of composite board, Xu et al. [43] developed flameproof composites using post-consumer aseptic cartons and HDPE with the addition of APP (ammonium polyphosphate) and MEL (melamine) as intumescent FRs (flame retardants). The cartons were washed with water, air dried, shredded, ground into powder (380 to 830 μm) and dried to 2 to 3% moisture. The powder was then mixed with HDPE (70:30) and up to 30% FR before extrusion and injection molding to produce test specimens for flammability and mechanical tests. While the FRs positively affected the fire retardancy of the composites, there was a deterioration in TS when the FR content exceeded 30 wt%. This process does not appear to have been commercialized.

2.2.5. Roofing Tiles

Araújo et al. [44] evaluated the mechanical and physical-chemistry properties of tiles made from aseptic cartons and compared them with conventional fibrocement tiles. The results showed that the TS of the carton tiles was greater (14.4 MPa) than the conventional

ones (5.2 MPa), as was the flexural strength (1.5 and 0.8 MPa, respectively). The fibrement tiles showed greater density and water absorption than the cartons ones. They concluded that tiles produced from cartons could easily be used as substitutes for conventional tiles.

2.2.6. Wall Cladding

In 2019 Foti et al. [45] examined the compressive strength and microstructure of gypsum-bonded wastepaper-based composites. Recycled wastepaper of various types including aseptic cartons were shredded to short length strips of about 4×18 mm and used as filling materials in natural gypsum in a ratio of 1:3 (*v/v*), and water was added to the mix. Seven different types of composites were produced depending on the material used. The densities ranged between 1.26 and 1.34 g cm^{-3}, and compressive strength was the lowest (4480 kPa) in the gypsum–magazine paper composites and the highest (6460 kPa) in the gypsum–carton composites. Since the samples exhibited adequate compressive strength, the products could be suitable for such applications as interior walls in building constructions. SEM (Scanning Electron Microscopy) indicated good adhesion between the hydrophobic matrix and the lignocellulosic fibers.

2.2.7. Layering of Cartons

While the preceding processes used shredded or ground aseptic cartons, Gallego et al. [46] prepared post-consumer aseptic cartons in three ways: stacked in 5 layers without cutting; cutting into 10 mm $\times$ 15 mm slices; and cutting into 5 mm $\times$ 5 mm slices. Several cycles of pressure and temperature were applied in a manual hydraulic press with hot plates to obtain the composites which the authors concluded could be applied for non-structural purposes in the building industry.

In a similar approach, Antón et al. [47] described a manufacturing process for binding 6 to 10 sheets of aseptic carton material (85 mm $\times$ 115 mm) at temperatures of 170 to 190 °C for 30 to 45 min at pressures of 3 MPa and 5 MPa. Density ranged from 0.74 to 0.81 g cm^{-3} which was lower than the theoretical value (0.981 g cm^{-3}) mainly due to the entrapment of bubbles during the fabrication process. The flexural strength values obtained for samples of 10 sheets ranged from 28 to 32 MPa and for the flexural modulus 5116 to 5790 MPa. Corresponding values for six sheets were 27 to 36 MPa and 7318 to 9215 MPa. The most influential factors in the process (for the range of chosen conditions) were the pressure and the time, with the temperature being a secondary factor. The results obtained for this type of material were very similar to those for wood-based materials and presented good reliability from a strength point of view.

Olfos et al. [48] developed insulating, self-supporting and multilayer panels composed entirely of aseptic cartons thermofused by mechanical means. Cartons were washed and opened using guillotines or paper cutters and three layers arranged in an overlapping manner; the two exterior faces had a reflective appearance, achieved by exposing the aluminum side of the cartons. The core of the panel consisted of unopened cartons arranged edge to edge, thus minimizing the preparation steps by omitting any cutting. A second design consisted of a series of prisms formed by cartons transversely cut and arranged next to each other, perpendicular to the base attached to two 'flat boards' followed by a central core. Finally, another 'flat board' that closed and formed the final insulation panel was added. This arrangement sought to limit the air movement within the cylinders and generate reflection on both aluminum faces. A third design consisted of a 'flat board' as the first layer, followed by a core based on a wavy 'flat board' created via thermoforming, and another 'flat board' layer. It was concluded that the three proposed panels could compete on equal terms with mineral and glass wools, two of the traditional insulators of moderate price and high performance available in the market.

Pons & Abt [49] analyzed the mechanical and fire properties of four household waste packages (HPDE, PET, PS (polystyrene) and aseptic cartons) as part of a broader research project developing new low-cost solar control devices for school façades reusing household waste. The aseptic cartons exhibited anisotropic mechanical behavior due to their

production process; parallel to the short edges of the carton it was stiffer and stronger but also more brittle compared to the other direction. Aseptic cartons also had a high specific strength and a TS per surface weight similar to steel and aluminum panels. Regarding the fire properties, the four waste containers had similar behavior to synthetic polymers and therefore should not be put close to flames. The aseptic cartons should have the polyethylene/aluminum layer on the outside (i.e., external) surface. It was concluded that because post-consumer waste packages were not designed for façade purposes, they should be introduced cautiously in the construction sector. In a subsequent paper, Habibi et al. [50] reported the degradation behavior and mechanical properties of aged samples of household waste containers including aseptic cartons under real environmental conditions. The TS of the cartons decreased due to UV aging but could be used in exterior conditions if their aluminum faces were exposed or painted with TiO_2 (titanium dioxide).

2.2.8. Irradiation

Gamma radiation has been used in order to improve the performance of various building products incorporating aseptic cartons. Martinez-Barrera et al. [51] studied the effects of paperboard from aseptic cartons and gamma radiation on the mechanical properties of cement concrete. Concrete specimens were prepared with paperboard (0.5 mm) at concentrations of 3, 5, and 7 wt% and irradiated at 200, 250, and 300 kGy. The highest improvement in mechanical properties was for concrete with 3 wt% of paperboard and irradiated at 300 kGy. Martinez-Barrera et al. [52] evaluated the effects of lamellae (1.5 to 3.0 mm) from aseptic cartons (up to 30 wt%), as well as of gamma rays (200 and 300 kGy), on the mechanical properties of cement concrete. Improvements of up to 39% for compressive strength and 30% for the elasticity modulus were obtained for concrete produced with 10% of cartons and irradiated at 300 kGy.

Martínez-López et al. [53] partially substituted silica sand in polyester resin composites used as polymer mortar with cut aseptic cartons (rectangular shapes with average sizes of 2 to 4 mm) at 1, 2, 4 and 6 wt% and then irradiated the modified composites with gamma rays at doses from 100 to 500 kGy. There was an improvement of 15% in the compressive strength and 16% in the flexural strength when 1% of carton particles and a 100 kGy irradiation dose were used. Mechanical properties decreased considerably at 2% and 200 kGy. Martinez-Barrera et al. [54] prepared polyester-based composites (80% polyester resin and 20% silica sand) and partially replaced the silica sand with paperboard from aseptic cartons at concentrations of 1, 2, 4 and 6 wt% and then irradiated them. Improvements were found in the mechanical properties (compressive and flexural strength as well as MOE) of the composites when they were irradiated at 100 and 200 kGy.

2.2.9. Novel Application

Xu et al. [55] developed a novel EMI (electromagnetic interference) shielding board using waste aseptic cartons with added iron fibers. The fiber loading level, fiber length and number of iron fiber layers significantly affected the EMI shielding properties. The shielding effectiveness increased with increasing fiber loading, fiber length and number of fiber layers, while the volume resistivity showed the opposite tendency. The boards had excellent total EMI shielding performance in the range of 9000 Hz to 200 MHz and 600 to 1500 MHz, and it was suggested that this value-added product could be used in packaging, construction and other application fields as it exhibited both environmental and economic advantages.

3. Pyrolysis

Pyrolysis is one of the alternative routes for treatment of aseptic cartons and involves thermochemical breakdown at elevated temperatures of 300–900 °C in an oxygen-free environment, resulting in a mixture of gas, oil, tar and char as the solid residue [56]. The gas can be used as fuel, frequently for heating the pyrolysis reactor, and the oil can either be used as fuel or as raw material for the production of chemicals.

Korkmaz et al. [57] investigated the pyrolysis of aseptic cartons in a nitrogen atmosphere at different temperatures (400 to 600 °C) using one- and two-stage pyrolysis modes. The char obtained was suitable to use as a solid fuel because of its high calorific value and low ash content. The gas produced was mostly formed from degradation of the paperboard and consisted largely of CO (40 mol%) and CO_2 (55 mol%). The first stage (below 400 °C) was characterized by primary degradation of paperboard, while the degradation of PE was significant in the second stage with the wax obtained having fewer impurities such as tarry compounds from cellulose degradation. The yield of Al from the second stage was 6.8 wt%. They concluded that aseptic cartons are a useful recycling resource and pyrolysis may be recognized as an attractive approach. However, because of the high quality of the paperboard in aseptic cartons, recovery of the paper fibers by hydrapulping is considered a more economically viable process than pyrolysis.

In 2021 Zúñiga-Muro et al. [58] provided a detailed analysis of the recycling of aseptic cartons via pyrolysis to recover the most valuable solid products (char and Al), and the subsequent application of the char in the adsorption of mercury in aqueous solutions. The chars showed outstanding mercury adsorption properties. Pyrolysis allowed an effective recovery of Al at pyrolysis temperatures of 600 and 800 °C; the Al recovered represented ~25% of the total solid product and exhibited an Al content >79.9 wt%. Higher temperatures, up to 1000 °C, promoted its oxidation to aluminum oxide and hydroxide. These findings may contribute to the development of new adsorbents with promising performance for water treatment and the associated recovery of Al for further industrial applications.

In pyrolysis, catalysts are usually used for two reasons: to decrease the pyrolysis temperature or for secondary tar decomposition when the aim is maximization of the gas produced. Haydary et al. [59] used a laboratory scale pyrolysis unit to study the pyrolysis of aseptic cartons at temperatures ranging from 650 to 850 °C using two catalysts (dolomite and red clay) and obtained a char with a carbon content of 52.0 to 68.0%. The aim was to maximize the amount of gas produced and reduce its tar content. Al foil was easily separated from the solid residue, and at temperatures below 750 °C, the Al foil obtained had no visible structural or chemical changes.

Tekin et al. [60] subjected waste aseptic cartons and waste diesel motor oil to co-pyrolysis at 500 °C in a fixed bed reactor. The fuel properties of the oils obtained from co-pyrolysis were found to be similar to those of commercial diesel. Co-pyrolysis oils can be used as liquid fuels after the upgrading process, and the higher heating values of solid residues from coprocessing were close to those of sub-bituminous coal.

Among the available thermal processes, HTL (hydrothermal liquefaction) provides an alternative way for the conversion of aseptic cartons into high-value energy fuel. Lokahita et al. [61] investigated the possibility of HTL (temperatures up to 240 °C and holding times up to 60 min) for processing post-consumer aseptic cartons. The HTL process degrades the paperboard into hydrochar, a coal-like material with a high calorific value (25.22 MJ kg^{-1}) that can be used as a co-combustion material with other solid fuels, especially coal. In addition, the PolyAl residual has potential to be used as rigid board material or in Al refining. A hard, robust, Al-rich material (20% to 25%) was formed during the hydrothermal treatment. In addition to its simplicity, hydrothermal treatment requires less capital cost but its industrial application as an aseptic carton recycling process still needs further investigation.

Wang et al. [62] evaluated HTL of aseptic cartons using SCW (sub/supercritical water: T_c = 374 °C and P_c = 22.1 MPa) in micro-batch reactors. The influence of temperature (300 to 420 °C), pressure (16 to 24 MPa), residence time (5 to 60 min) and feed concentration (5 to 40 wt%) on bio-oil yield, HHV (high heating value), and functional groups in bio-oil were investigated. The results showed that bio-oil yield firstly increased with increasing temperature and then decreased when the temperature exceeded 360 °C. Reaction times longer than 30 min gave a negative effect on bio-oil yield while the influence of pressure on bio-oil yield increased markedly from 16 MPa to 22 MPa, and then stabilized. Maximum bio-oil yield of 35.55% was found at 360 °C, 22 MPa, 30 min and a feed concentration of

20 wt%. HHV and energy recovery efficiency increased significantly with temperature, and a maximum HHV of 48.747 MJ kg^{-1} and energy recovery efficiency of 46.49% were found at 420 °C, 20 MPa, 30 min and feed concentration of 20 wt%. The main compounds in bio-oil were ketones, phenolics, esters, and alcohols, formed by the liquefaction of carbohydrates and lignin. Possible liquefaction pathways of aseptic cartons were proposed but the economic viability of this process has yet to be established.

4. Recycling the Polyethylene and Aluminum (PolyAl) Residual

After the paper fibers have been removed by hydrapulping, a residual of polyethylene and aluminum (PolyAl) remains. Lopes & Felisberti [3] characterized the PolyAl residual and compared its properties with those of pure LDPE and EMAA, the polymers that constitute the residual. PolyAl is around 15% aluminum particles of different shapes and sizes. The residual had higher thermooxidative stability, higher crystallinity, lower impact resistance, and higher tensile strength than the pure olefin polymers.

A variety of recycling approaches have been reported and several techniques on a laboratory or pilot plant scale have been developed for processing the PolyAl residual; they can be grouped into mechanical, chemical and thermal processes.

4.1. Mechanical

A process to recycle the PolyAl residue was described by von Zuben and Neves [63,64]. It consisted of washing to remove remaining paper fibers, agglutinating the material followed by extrusion to obtain pellets, and injection molding to produce products such as hangers, pens, brooms, notepads, clip holders, flowerpots, etc. For some products virgin LDPE was added to the PolyAl pellets. Since 2016 Recon Polymers BV, located in Roosendaal, Netherlands has processed PolyAl into granulate, which is then injection molded to make products such as gravel grids, plug boxes and bird feed containers. Investal in Tambov, Russia recycles PolyAl into pellets that can then be used in industrial and consumer products such as composite panels, benches, crates and pens.

The PolyAl residual has also been used to make laminated boards for the building industry [65]. However, during extrusion the fiber residues can burn generating carbon and water vapor which compromises quality. Hidalgo [66] investigated the feasibility of manufacturing composite rigid board from the PolyAl residual using a hot press. The composite board had low water absorption and acceptable TS and could be used to manufacture cable reels. Hidalgo-Salazar et al. [67] illustrated a number of products from PolyAl residual using techniques including compression molding, extrusion, injection molding, rotational molding, fabrication of fiber-reinforced composites, and blends with other polymers.

Later Hidalgo-Salazar et al. [68] reported the dynamic mechanical response and performance to short-term creep of composites made from mats of fique (a strong durable fiber obtained from the leaves of a succulent tropical American plant of the genus Furcraea and traditionally used for the manufacture of packaging and cordages) and PolyAl residual from aseptic cartons. Chemical treatments such as alkalinization with NaOH, silanization and impregnation treatments with LDPE, were applied to improve the compatibility of the fiber matrix; their effects on the creep response and mechanical properties of PolyAl/Fique were investigated. A later report by Hidalgo-Salazar et al. [69] included details on the maximum strength, Young's modulus and flexural properties which increased as a direct function of the amount of reinforcement contained in the material. A reduction in density from the generation of voids at the interface between the PolyAl and fique fibers was identified, and there was also greater water absorption due to the weak interphase fiber-matrix and the hydrophilic nature of the fibers. Muñoz-Vélez et al. [70] manufactured panels with 10%, 20%, and 30% *v/v* of surface-treated fique fiber by hot compression molding; pre-impregnation with LDPE promoted a significant increase in the tensile and flexural properties, and panels with 30% fibers showed a 53.15% decrease in water absorption capacity compared to panels made with 30% untreated fique fibers. Increases

in the fiber content caused mainly better mechanical performances, which increased as a direct function of the amount of fique incorporated.

Ayrilmis et al. [71] investigated the feasibility of lignocellulosic wastes (rice husk particles and beech sawdust) as a filler in the PolyAl residual. The injection-molded composites were prepared from PolyAl with and without 3 wt% MAPE at 40, 50, and 60 wt% of sawdust or rice husk flour. The sawdust flour composites had better flexural and tensile properties than those made with rice husk flour. The strength and modulus values of the filled composites were significantly higher than the unfilled composites. The TS values of the filled PolyAl composites increased with increasing filler content up to 50 wt%. The tensile elongation at break values declined sharply with the addition of the filler. It was suggested that the reinforced thermoplastic composites with their low cost and high performance could be efficiently used in automotive interiors and outdoor decking applications.

In 2020 Ovchinnikov et al. [72] defined the chemical composition, thermal, mechanical and technological properties of a composite consisting of PolyAl (61 wt%), PE/paper (23 wt%) and plastic caps (16 wt%) after the paper had been removed from aseptic cartons. The TS and MOE were higher compared to pure LDPE and the composite had sufficient mechanical strength and technological characteristics to be used for different unspecified applications. The properties of the composite could be improved by using additives to increase the elasticity and melt flow index.

Valim et al. [73] studied the properties and behavior of low weight PolyAl roofing tiles using optical microscopy, SEM and X-ray dispersive energy spectroscopy; the thermal and mechanical behavior of the tiles after they had been subjected to hygrothermal air conditioning and UV radiation was also reported. It was concluded that although the weathering involving humidity and UV radiation influenced the thermal and mechanical performance of the PolyAl tiles, it was not enough to make their use unfeasible in civil construction.

Sánchez-Alvareza et al. [74] reported the results of an optical non-destructive comparative study of the surface deformation of PolyAl and clay roof tiles exposed to heat radiation. The PolyAl tile showed a greater deformation profile compared with the classic clay tile, but its thermal insulation properties were better (thermal conductivity was $0.22 \text{ W m}^{-1} \text{ K}^{-1}$ compared to $1 \text{ W m}^{-1} \text{ K}^{-1}$ for the clay roof tile). In addition to being good thermal insulators and waterproof, the PolyAl tiles do not generate fungi or bacteria, are good acoustic insulators, highly unbreakable, and have high durability, making them an excellent alternative as long term and low-cost construction elements.

By eliminating the impurities and foreign polymers from the PolyAl residual, Italian company Ecoplasteam S.p.a. based in Alessandria, Italy produces an added-value product (trade name EcoAllene® AA00 BASE) that can be injected, extruded, blended and compounded like a normal polymer, and further recycled. Ecoplasteam licenses the production process technology from Swiss company REPLAN (Recycling Planet) Global that holds a 2008 patent [75]. It involves a pulping process followed by settling and then centrifuging to obtain a solid PolyAl fraction, shredding, drying to <2% moisture content, compacting, extruding and subdividing into granules. The final material is 85% LDPE and about 15% aluminum with <2% cellulose.

In 2020 Cravero & Frache [76] evaluated and improved the flammability and combustion behavior of the EcoAllene® PolyAl residue to widen its use to applications where these properties are required. Thermogravimetric analysis showed an enhancement in the main degradation step temperature (from 385 °C to 421 °C) due to the presence of the aluminum flakes. Two FR approaches were tested: an intumescent system made of APP and pentaerythritol, and magnesium hydroxide. For all the materials tested, the temperature of the main weight loss step increased, the flammability rating improved and the fire hazard decreased. Full compatibility was found in the PolyAl–magnesium hydroxide compound, while the PolyAl-intumescent appeared as a heterogeneous system.

4.2. Chemical

Chemical approaches involve the solubilization of LDPE, removal of the aluminum and evaporation of the solvent or the addition of an anti-solvent to recover the LDPE.

A 1992 patent assigned to Tetra Laval [77] for separating PolyAl involved treatment with an organic acid or a mixture of organic acids selected from among formic, acetic, propanoic, butyric and similar volatile organic acids with acetic acid alone being preferred. Very good separation efficiency was achieved using an aqueous solution containing approximately 80 wt% acetic acid at 80 °C (close to the flash point). However, the high concentration of acetic acid is very aggressive and will attack the aluminum, leading to the formation of H_2, as well as loss in the quantity of aluminum recovered in the process [78].

Aluminum-plastic laminates have also been separated using a hydrothermal process where only superheated water was used in the separation [79]. However, the results suggest that separation of Al and plastic was incomplete which, in addition to the considerable energy demand for keeping water at high temperatures, makes this hydrothermal process inefficient.

Mu'min et al. [80] employed a wet torrefaction (hydrothermal) process at 170 °C and an acetic acid concentration of 3% to treat PolyAl residual. The highest Al yields were 14.68% for a 30-min holding time and 15.78% for a 60-min holding time, close to the theoretical maximum Al yield of 15.85%. The recovered plastic had a calorific value of more than 30 MJ kg^{-1}, sufficient to be used as a fuel although it is likely to be too expensive compared to alternative fuels.

Zhang et al. [81] separated PolyAl using aqueous solutions of organic acids with formic acid performing best at 60 °C with a separating time of 25 min and 4.73% Al loss. Delamination using an organic solvent blend of benzene, ethanol and water was investigated by Zhang et al. [82]. Delamination occurred due to the swelling of PE and although some material loss was observed, it was lower than that obtained with separation using organic acids. A blend of benzene-ethanol-water (30:20:50 volume ratio) for 5 min at 60 °C was used to separate PolyAl layers; the bright spots of irregular particles with scattered distribution detected by SEM on the surfaces of the PE and Al were Al, O and C [83].

Yan et al. [84] studied a wet process technique on PolyAl and compared the separation efficiency achieved using different separation reagents (hydrochloric, methanoic and acetic acids). The impacts of a range of parameters such as the reagent concentration, temperature, and liquid–solid ratio, on the separation time and aluminum loss ratio were determined. Methanoic acid was found to be the optimal separation reagent at a concentration of 2 to 4 mol L^{-1}, a temperature of 60 to 80 °C, and a liquid–solid ratio of 30 L kg^{-1}. These conditions allowed Al and PE to be separated in less than 30 min, with less than 3% aluminum loss. A demonstration facility with a capacity of 50 t d^{-1} was built in China and achieved recovery rates of more than 98% for PE and 72% for Al.

Rodríguez-Gómez et al. [85] proposed a separation process for the PolyAl residual using a washing process with waste cooking oil and various solvents (ethanol, chloroform and isopropyl alcohol). The bench scale results showed that up to 85% and 80%, respectively of Al and PE could be recovered.

However, the use of vegetable oil, while more environmentally friendly than organic solvents such as toluene and xylene, required chloroform or alcohols to remove oily contaminants from both the PE and Al. The quality and purity of the Al and PE strongly affect their commercial value, and the harsh conditions of some chemical processes can hamper the extrudability of the recovered PE or the melting of Al flakes [86].

Solvent-based PE extraction methods have been studied for the separation of PolyAl residual, where PE is dissolved in an organic solvent and separated from the undissolved Al [78]. Pappa et al. [87] evaluated the SDP (Selective Dissolution-Precipitation) process for the separation/recycling of LDPE/PP mixtures. The SDP process relies on controlling and adjusting the polymer's solubility by changing solvent and/or dissolution conditions. The basic steps of SDP for recovering a certain polymer with high purity involve dissolution of

the polymer, removal of the non-dissolved material through filtration and addition of a non-solvent or anti-solvent so that the polymer precipitates again [88].

In 2021 Georgiopoulou et al. [89] developed a SDP process to recover PE and Al. Xylene at 85 °C for 2 h was utilized as the solvent for PE dissolution, and isopropanol as the antisolvent to precipitate it. PE coming from the inner layers was recovered as a white powder of high purity with thermal properties similar to those of pure LDPE. PE from the outer plastic layer still contained impurities such as printing inks, suggesting that further purification may be needed depending on the application for which it is destined.

A 2017 Italian patent filed by Tagliavini et al. [90] described a new technology of SHS (switchable hydrophilic solvents) for the separation and recovery of PE and Al from shredded polylaminate food packaging such as aseptic cartons. It allows the recovery of both materials in high quantities (>99% for Al and > 80% for PE) and good quality ($\geq$86% of non-oxidized Al). In a follow-up publication, Samori et al., [86] reported that treatment with DMCHA (*N*,*N*-dimethylcyclohexylamine), a lipophilic tertiary amine, allowed very high material recovery (>99% for Al and >80% for PE), without compromising the quality in terms of oxidation or polymer degradation. The polarity of DMCHA can be tuned through CO_2 addition and removal, switching from a neutral solvent (suitable for PE solubilization) to an ionic liquid (in which PE is no longer soluble and thus can be recovered). Results from a simplified and preliminary life cycle analysis confirmed the potential environmental benefits of the SHS approach compared with other treatment and disposal scenarios. Mumladze et al. [91] reported the use of DMCHA for the recycling of six different multilayer flexible packaging materials all of which contained Al foil and various plastic polymers. Al was recovered in the form of flakes with an average size of 100 μm and could be used in powder metallurgy applications.

Nieminen et al. [92] reported in 2020 the separation of Al and polymeric layers of waste pharmaceutical blisters by exploitation of a DES (deep eutectic solvent) consisting of lactic acid and choline chloride (in a molar ratio of 1:9), and pure lactic acid, both of which are considered green and environmentally friendly solvents due to their nonflammability, nontoxicity and low vapor pressure. After the separation by pure DES, the recovered Al fraction was corroded, containing 65 wt% of Al and 23 wt% of oxygen resulting from the formation of aluminium lactate, whereas after lactic acid treatment, Al surfaces contained about 95% of Al (Al foil contains 96% of Al). The results showed that the DES used and lactic acid can offer viable green separation methodologies for Al and plastic from blister packages and is a possible approach for recycling the PolyAl residual. However, methods for purifying the used solvent and recovering the dissolved Al need to be developed prior to implementing the technology in industry.

4.3. Thermal

Siddiqui et al. [93] reported thermal and catalytic pyrolysis of aluminum/plastic laminates (coffee capsules) over zeolite catalysts to produce high quality oil. Yin et al. [94] reported the efficient recovery of high purity Al from aluminum/plastic laminates (face masks) using thermo-delamination. Both approaches could possibly be adopted to recover Al from PolyAl residuals, but a systematic approach for the interactions between the components during pyrolysis would be necessary prior to commercialization, not only for Al recovery but also for efficient energy production.

Plasma pyrolysis integrates conventional pyrolysis with the thermochemical properties of plasma to transform plastic waste into syngas (synthesis gas). The process temperatures are very high, ranging between 1730 and 9730 °C, and the waste plastics are decomposed into monomers. The process is extremely fast, lasting between 0.01 and 0.5 sec, depending on process temperature and type of waste [95]. The resulting syngas is composed mainly of CO, H_2 and small amounts of higher hydrocarbons.

Hepworth et al. [96] developed a new type of plasma device, SSP (Sustained Shockwave Plasma), for treatment of electric arc furnace dust on a laboratory scale to produce a non-hazardous slag and metallic zinc and lead to recycle. It was then adapted to pro-

cess aseptic cartons. A plasma sustained by discreet pulses applied to weakly ionized gas, known as a SSP, was developed by Refranco Corporation in collaboration with Tetra Laval. A pilot scale 500-kW arc torch was constructed in Singapore in 1996 for treatment of aseptic cartons and municipal solid waste in general. The feed particles entered a "free-fall" chamber and enhanced and stabilized the plasma. The particles themselves served as the constricting nozzle, in contrast to the electrodes in a DC (direct current) torch and the magnetic field in an ICP (inductively coupled plasma) torch. The main benefits were high electrical efficiency and operational flexibility. The frequency, amplitude, and width of the pulses were variable and could be adjusted to suit a particular condition. Despite several years of trials, the plant never operated as envisaged.

In 2001 Corenso, jointly owned by Stora Enso and UPM-Kymmene, opened a 85,000 tonnes/year carton recycling plant adjacent to their paper mill at Varkaus, Finland at a total cost of €45 million. To process the PolyAl residue the plant used atmospheric-pressure bubbling fluidised-bed gasification (BFB) technology developed by VTT (Technical Research Centre of Finland) at a temperature of 800 °C. The gas produced from the plastic was combusted in a steam boiler, replacing fuel oil consumption in the power plant in Varkaus. The aluminum was removed from the gas as a fine powder but contained coal-like particles and required further refining before it could be used. The plant closed in 2010. The quality of the aluminum can be hampered by a high level of oxidation and/or char residue.

Alcoa Aluminio, in a joint venture with Tetra Pak, Klabin and TSL Ambiental called EET (Edging Environment Technology) established a recycling plant in Brazil in 2005 at a cost of $40 million. It used electrical energy to produce a jet of plasma at 1500 °C that ionized the PolyAl residual; the plastic was transformed into oil and the aluminum melted but was contaminated with residual paper fibers. The plant ceased operation in 2010.

Stora Enso opened a carton recycling plant in Barcelona, Spain in 2010 that included a pyrolysis component costing €6.5 million developed in partnership with Alucha Recycling Technologies. The PolyAl residue from hydrapulping was dried and broken down into small pieces prior to being heated to 450 °C in an oxygen-free rotary kiln where the plastic pyrolyzed with the products used to heat the kiln and produce steam for the paper mill, while the aluminum remained as small flakes that were cooled, compressed into briquettes and remelted to make new aluminum products. Stora Enso and Alucha Recycling Technologies were granted the 'Best of the Best' LIFE Environment Project award for 2010 by the European Commission [97]. In 2016 the paper mill was sold and the pyrolysis plant closed.

Microwave-assisted pyrolysis of waste plastics involves mixing plastics with a highly microwave-absorbent dielectric material with the heat absorbed from the microwaves being transferred to the plastics by conduction. The sources of microwave radiation allow very high temperatures and heating rates and reach high conversion efficiencies of electrical energy into heat [56]. Ludlow-Palafox & Chase [98] evaluated a novel microwave-induced pyrolysis process and recovered clean aluminum from Al/plastic toothpaste tubing (used as an example of a laminated material) together with hydrocarbons.

In 2005 Enval, a UK-based company spun off from the University of Cambridge, was established in England and built a pilot plant to demonstrate the viability of microwave-induced pyrolysis of Al/plastic including the PolyAl residue from carton recycling. Using patented technology [99] a bed of carbon is heated using microwave energy in a reactor chamber, and the reactor is purged with nitrogen. At a temperature of typically 500 to 600 °C, laminate material is dropped into and mixed with the carbon bed. With continued microwave irradiation of the carbon bed, the organic content of the laminate is heated by conduction and pyrolyzes to a gaseous fraction that can be recovered by condensation to form an oily or waxy hydrocarbon product, together with a non-condensable gaseous fraction. The aluminum can be separated from the carbon bed by coarse sieving and recovered as a solid. About 75% of the plastic condenses into an oil and the company is now collaborating with various plastics producers to use this oil to manufacture new plastics; the other 25% of what was plastic remains as a gas and is fed into an electricity

generator that then feeds the microwaves to produce the energy needed for the process. In 2015 a commercial unit that can recycle up to 2000 tonnes a year began operation at Alconbury, near Huntingdon and a second plant is being planned for the north of England.

5. Conclusions

As is clear from this review, there are a number of recycling options for aseptic beverage cartons, ranging from construction materials that utilize the whole carton, to hydra- pulping to recover the paper fibers, and various processes to process the PolyAl residual that remains after hydrapulping. While there are many paper mills in the world that have hydrapulpers that could recycle aseptic cartons, the fact that the maximum theoretical yield is just 75% compared to 85% or more for other paper packaging is a disincentive, as is the challenge of economically processing the PolyAl residual. Therefore, it is suggested that the focus in future years is likely to be on recycling cartons into construction materials where there is a theoretical yield of 100%. For this to be successful, considerable investment in new factories will be required as well as the development of economically viable markets for the materials. However, the major barrier to the expansion of aseptic carton recycling is not recycling capacity but the collection and sorting infrastructure which is still less than adequate in many countries. Overriding all of the above is economics: unless the collection, sorting and recycling is profitable or subsidized then aseptic carton recycling will not expand.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The author declares no conflict of interest.

References

1. Robertson, G.L. The paper beverage carton: Past and future. *Food Technol.* **2002**, *56*, 46–51.
2. Pieper, G.; Petersen, K. Free fatty acids from orange juice absorption into laminated cartons and their effects on adhesion. *J. Food Sci.* **1995**, *60*, 1088–1091. [CrossRef]
3. Lopes, C.M.A.; Felisberti, M.I. Composite of low-density polyethylene and aluminum obtained from the recycling of postconsumer aseptic packaging. *J. Appl. Polym. Sci.* **2006**, *101*, 3183–3191. [CrossRef]
4. Colvin, R. Tetra Pak Makes Materials Change for Liquids Packaging. Plastics Today 9th February 2006. Available online: www.plasticstoday.com/tetra-pak-makes-materials-change-liquids-packaging/ (accessed on 23 November 2020).
5. Zawadiak, J.; Wojciechowski, S.; Piotrowski, T.; Krypa, A. Tetra Pak recycling—current trends and new developments. *Am. J. Chem. Eng.* **2017**, *5*, 37–42. [CrossRef]
6. Henshaw, J.M.; Han, W. An overview of recycling issues for composite materials. *J. Thermoplast. Compos. Mater.* **1996**, *9*, 4–20. [CrossRef]
7. Singh, N.; Hui, D.; Singh, R.; Ahuja, I.; Feo, L.; Fraternali, F. Recycling of plastic solid waste: A state of art review and future applications. *Compos. Part B* **2017**, *115*, 409–422. [CrossRef]
8. Alliance for Beverage Cartons and the Environment. Available online: www.beveragecarton.eu/wp-content/uploads/2020/06/ACE_best_practices_BC_recycling_202006.pdf (accessed on 11 February 2021).
9. Carton Council. Available online: www.recyclecartons.com/about/ (accessed on 11 February 2021).
10. Bajpai, P. Stock preparation. In *Biermann's Handbook of Pulp and Paper: Paper and Board Making*, 3rd ed.; Elsevier: Amsterdam, The Netherlands, 2018; Volume 2, pp. 65–76.
11. Abreu, M. *Recycling the Fibres on Tetra Pak Cartons*; Tetra Pak Canada Inc.: Markham, ON, Canada, 2000; p. 44.
12. Hyder, A.; Nyström, T. Method and Apparatus for Separating Paper Fiber and Plastics from Mixed Waste Materials and Products Obtained Thereby. U.S. Patent US5390860A, 14 April 1993.
13. Platnieks, O.; Barkane, A.; Ijudina, N.; Gaidukova, G.; Thakur, V.K.; Gaidukovs, S. Sustainable tetra pak recycled cellulose/Poly(Butylene succinate) based woody-like composites for a circular economy. *J. Clean. Prod.* **2020**, *270*, 122321. [CrossRef]
14. Downs, M.L.; Schmitt, M.G. Composite Board and Method of Manufacture. U.S. Patent 3718536, 22 April 1970.
15. Nilsson, I. A Method for the Manufacture of Fibreboards. European Patent 0358119A1, 1 September 1989.
16. Heuser, H.; Knorr, N. Process for the Production of a Composite Particle Object and Object so Obtained. European Patent 0588023A1, 22 July 1993.

17. Günther, J.; Ridder, C. SONY: Addressing different stages of the product's life cycle. *UNEP Ind. Environ.* **1997**, *20*, 28.
18. Nyström, T. *Production of Panel Board—A World Overview*; Tetra Pak Carton Packaging Division: Lund, Sweden, 2000; p. 11.
19. Peña, M.; Méndez, M.; Chacón, R. Multifactorial analysis for the characterization and prototyping of recyclable cardboard panels of short and long term. *Artículo Investig. Rev. Cienc. Ing.* **2016**, *37*, 177–186.
20. Quintero, M.; Rodríguez, P.; Rubio, J.; Jaramillo, L.; Nuñez-Moreno, F. Bending and compression characterization of hollow structural elements made of recycled Tetra Pak®-Based Boards (RTPBB) and an approximated calculation of the carbon footprint involved in their production. *Rev. Ing. Construcción* **2018**, *32*, 131–148. [CrossRef]
21. Ayrilmis, N.; Candan, Z.; Hiziroglu, S. Physical and mechanical properties of cardboard panels made from used beverage carton with veneer overlay. *Mater. Des.* **2008**, *29*, 1897–1903. [CrossRef]
22. Sen, S.; Ayrilmis, N.; Candan, Z. Fungicide and insecticide properties of cardboard panels made from used beverage carton with veneer overlay. *Afr. J. Agric. Res.* **2010**, *5*, 159–165.
23. Rhamin, H.; Madhoushi, M.; Ebrahimi, A.; Faraji, F. Effect of resin content, press time and overlaying on physical and mechanical properties of carton board made from recycled beverage carton and MUF resin. *Life Sci. J.* **2013**, *10*, 613–619.
24. Murathan, A.; Murathan, A.S.; Gürü, M.; Balbas, M. Manufacturing low density boards from waste cardboards containing aluminium. *Mater. Des.* **2007**, *28*, 2215–2217. [CrossRef]
25. Parada-Soria, A.; Yao, H.F.; Alvarado-Tenorio, B.; Sanchez-Cadena, L.; Romo-Uribe, A. Recycled HDPE-tetrapack composites. Isothermal crystallization light scattering and mechanical properties. *MRS Online Proc. Library* **2012**, *1485*, 77–82. [CrossRef]
26. Carrillo, J.G.; Ventura, D.A.P.; Gamboa, R.A.; Cruz-Estrada, R.H. Improvement on mechanical properties of a particle board made of recycled material based on Tetra Brik®. *Mater. Res. Soc. Symp. Proc.* **2014**, *1611*, 13–18. [CrossRef]
27. Chan-Koyoc, M.C.; Cruz-Estrada, R.H.; Cruz-Delgado, V.J.; Carrillo, J.G. Effect of natural and accelerated aging on the mechanical performance of a composite based on recycled multilayer carton. *J. Polym. Environ.* **2019**, *27*, 2509–2522. [CrossRef]
28. Bekhta, P.; Lyutyy, P.; Hiziroglu, S.; Ortynska, G. Properties of composite panels made from Tetra Pak and polyethylene waste material. *J. Polym. Environ.* **2016**, *24*, 159–165. [CrossRef]
29. Aranda-García, F.J.; González-Pérez, M.M.; Robledo-Ortíz, J.R.; Sedano-de la Rosa, C.; Espinoza, K.; Ramírez-Arreola, D.E. Influence of processing time on physical and mechanical properties of composite boards made of recycled multilayer containers and HDPE. *J. Mater. Cycles Waste Manag.* **2020**, *22*, 2020–2028. [CrossRef]
30. Guillén-Mallette, J.; Carrillo-Baeza, J.; Aranda-Ayala, A.; Rivero- Ayala, M. Optimization of processability and physical and mechanical properties of extruded polyethylene—Tetra Pak cartons composites by experimental design. *J. Thermoplast. Compos. Mater.* **2019**, 1–26. [CrossRef]
31. Hwang, G.-S.; Wang, E.-C.; Su, Y.-C. Preparation of composite board using foil-laminated and plastic-laminated liquid packaging paperboard as raw materials. *J. Wood Sci.* **2006**, *52*, 230–235. [CrossRef]
32. Moya, R.; Camacho, D.; Mata, J.; Fallas, R.S. Fungal decay, coating, burning properties and change of color of particleboards manufactured with woody biomass, agricultural wastes and Tetra Pak residues. *J. Biomater. Nanobiotech.* **2013**, *4*, 334–342. [CrossRef]
33. Sun, X.; Zhang, Q.-H. Study on the optimum hot-pressing process and surface decoration of waste Tetra Pak/sawdust composite board. *Adv. Mater. Res.* **2013**, *710*, 147–151. [CrossRef]
34. Ebadi, M.; Farsi, M.; Narchin, P.; Madhoushi, M. The effect of beverage storage packets (Tetra Pak™) waste on mechanical properties of wood–plastic composites. *J. Thermoplast. Compos. Mater.* **2016**, *29*, 1601–1610. [CrossRef]
35. Ebadi, M.; Farsi, M.; Narchin, P. Some of the physical and mechanical properties of composites made from Tetra Pak™/LDPE. *J. Thermoplast. Compos. Mater.* **2017**, *31*, 1054–1065. [CrossRef]
36. Hassanin, A.H.; Candan, Z. Novel Bio-Based Composites Panels from Tetra Pak Waste. *Key Eng. Mater.* **2016**, *689*, 138–142. [CrossRef]
37. Nassef, M.G.; El-Galy, I.; Hassanin, A. Mechanical and dynamic characterization of sustainable composites based on food packaging waste. *MATEC Web Conf.* **2018**, *249*, 01002. [CrossRef]
38. Hassanin, A.H.; Candan, Z.; Demirkir, C.; Hamouda, T. Thermal insulation properties of hybrid textile reinforced biocomposites from food packaging waste. *J. Ind. Text.* **2018**, *47*, 1024–1037. [CrossRef]
39. Mohareb, A.O.S.; Hassanin, A.H.; Candelier, K.; Thévenon, M.F.; Candan, Z. Developing biocomposites panels from food packaging and textiles wastes: Physical and biological performance. *J. Polym. Environ.* **2017**, *25*, 126–135. [CrossRef]
40. Hamouda, T.; Hassanin, A.H.; Saba, N.; Demirelli, M.; Kilic, A.; Candan, Z.; Jawaid, M. Evaluation of mechanical and physical properties of hybrid composites from food packaging and textiles wastes. *J. Polym. Environ.* **2019**, *27*, 489–497. [CrossRef]
41. Figen, A.K.; Terzi, E.; Yilgör, N.; Kartal, S.N.; Piskin, S. Thermal degradation characteristic of Tetra Pak panel boards under inert atmosphere. *Korean J. Chem. Eng.* **2013**, *30*, 878–890. [CrossRef]
42. Yilgor, N.; Köse, C.; Terzi, E.; Figen, A.K.; Ibach, R.; Kartal, S.N.; Piskin, S. Degradation behavior and accelerated weathering of composite boards produced from waste Tetra Pak® packaging materials. *BioResources* **2014**, *9*, 4784–4807. [CrossRef]
43. Xu, C.; Jian, W.; Xing, C.; Zhou, H.; Zhao, Y.; Pan, H.; Xiong, X. Flame retardancy and mechanical properties of thermal plastic composite panels made from Tetra Pak waste and high-density polyethylene. *Polym. Compos.* **2016**, *37*, 1797–1804. [CrossRef]
44. Araújo, D.C.; Morais, C.R.S.; Altides, M.E.D. Avaliação mecânica e físico-química entre telhas convencionais e alternativas usadas em habitações populares. Mechanical and physical-chemical evaluation between conventional and alternative tiles used in popular housing. *J. Mater. Process.* **2008**, *3*, 50–56.

45. Foti, D.; Adamopoulos, S.; Voulgaridis, E.; Passialis, C.; Amiandamhen, S.O.; Daniel, G. Microstructure and compressive strength of gypsum-bonded composites with papers, paperboards and Tetra Pak recycled materials. *J. Wood Sci.* **2019**, *65*, 42–50. [CrossRef]
46. Gallego, S.M.; Aponte, A.G.; Sierra, R.B.; Marín, J.F. Evaluation of mechanical properties of composites manufactured from recycled Tetra Pak containers. *Tecnura* **2020**, *24*, 36–46. [CrossRef]
47. Antón, N.; González-Fernández, Á.; Villarino, A. Reliability and mechanical properties of materials recycled from multilayer flexible packages. *Materials* **2020**, *13*, 3992. [CrossRef]
48. Olfos, L.; Valdes, F.; Sarabia, G.; Barros, P. Thermal insulating panels based on recycled Tetra-Pak® packaging materials. *IOP Conf. Ser. Earth and Environ. Sci.* **2020**, *503*, 012076. [CrossRef]
49. Pons, O.; Abt, T. Evaluation of household waste materials for façade components in primary educational workshops. *Mechanical and fire properties. J. Build. Eng.* **2020**, *29*, 101202. [CrossRef]
50. Habibi, S.; Pons, O.; Abt, T. Evaluation of household waste materials for façade components in primary educational workshops. Degradation behavior and mechanical properties of aged samples. *J. Build. Eng.* **2021**, *33*, 101573. [CrossRef]
51. Martínez-Barrera, G.; Barrera-Díaz, C.E.; Cuevas-Yañez, E.; Varela-Guerrero, V.; Vigueras-Santiago, E.; Ávila-Córdoba, L.; Martínez-López, M. Waste cellulose from Tetra Pak packages as reinforcement of cement concrete. *Adv. Mater. Sci. Eng.* **2015**, 682926. [CrossRef]
52. Martínez-Barrera, G.; del Coz-Diaz, J.J.; Alonso-Martínez, M.; Martínez-López, M. Lamellae of waste beverage packaging (Tetra Pak) and gamma radiation as tools for improvement of concrete. *Case Stud. Constr. Mater.* **2020**, *12*, e00315. [CrossRef]
53. Martínez-López, M.; Martínez-Barrera, G.; Barrera-Díaz, C.; Ureña-Núñez, F.; Laredo dos Reis, J.M. Waste Tetra Pak particles from beverage containers as reinforcements in polymer mortar: Effect of gamma irradiation as an interfacial coupling factor. *Constr. Build. Mater.* **2016**, *121*, 1–8. [CrossRef]
54. Martínez-Barrera, G.; Martínez-López, M.; González-Rivas, N.; del Coz-Diaz, J.J.; Ávila-Córdoba, L.; Laredo dos Reis, J.M.; Gencel, O. Recycled cellulose from Tetra Pak packaging as reinforcement of polyester based composites. *Constr. Build. Mater.* **2017**, *157*, 1018–1023. [CrossRef]
55. Xu, C.; Liu, J.; Zhu, X.; Zhu, Y.; Xiong, X.; Cheng, X. Electromagnetic interference shielding boards produced using Tetra Paks waste and iron fiber. *J. Mater. Cycles Waste Manag.* **2015**, *17*, 391–398. [CrossRef]
56. Arshad, H.; Sulaiman, S.A.; Hussain, Z.; Naz, Y.; Basrawi, F. Microwave assisted pyrolysis of plastic waste for production of fuels: A review. *MATEC Web Conf.* **2017**, *131*, 02005. [CrossRef]
57. Korkmaz, A.; Yanik, J.; Brebu, M.; Vasile, C. Pyrolysis of the tetra pak. *Waste Manag.* **2009**, *29*, 2836–2841. [CrossRef] [PubMed]
58. Zúñiga-Muro, N.M.; Bonilla-Petriciolet, A.; Mendoza-Castillo, D.I.; Duran-Valle, C.J.; Silvestre-Albero, J.; Reynel-Ávila, H.E.; Tapia-Picazo, J.C. Recycling of Tetra pak wastes via pyrolysis: Characterization of solid products and application of the resulting char in the adsorption of mercury from water. *J. Clean. Prod.* **2021**. [CrossRef]
59. Haydary, J.; Dudáš, D.S.J. Pyrolysis of aseptic packages (tetrapak) in a laboratory screw type reactor and secondary thermal/catalytic tar decomposition. *Waste Manag.* **2013**, *33*, 1136–1141. [CrossRef]
60. Tekin, K.; Ucar, S.; Karagöz, S. Influence of co-pyrolysis of waste Tetra Pak with waste motor oil on product distribution and properties for fuel application. *Energy Fuels* **2019**, *33*, 11101–11112. [CrossRef]
61. Lokahita, B.; Aziz, M.; Yoshikawa, K.; Takahashi, F. Energy and resource recovery from Tetra Pak waste using hydrothermal treatment. *Appl. Energy* **2017**, *207*, 107–113. [CrossRef]
62. Wang, Y.; Zhu, Y.; Liu, Z.; Su, J.; Fang, C.; Xu, D.; Song, W.; Wang, S. Influences of operating parameters on liquefaction performances of Tetra Pak in sub-/supercritical water. *J. Environ. Manag.* **2019**, *237*, 545–551. [CrossRef]
63. Von Zuben, F.; Neves, F.L. *Recycling of Aluminum and Polyethylene Present in Tetra Pak Carton Packaging*; International Aluminum Recycling Seminar: São Paulo, Brazil, 1999; pp. 96–109.
64. Von Zuben, F.; Neves, F.L. Recycling of aluminum and polyethylene from Tetra Pak carton packages as plastic composite. In Proceedings of the GPEC 2003—Global Plastics Environmental Council, Detroit, MI, USA, 26–27 February 2003.
65. Von Zuben, F.; Orsato, R.J.; Van Wassenhove, L. Turning waste into wealth. *Int. Commer. Rev.* **2007**, *7*, 116–123. [CrossRef]
66. Hidalgo, M. Manufacturing rigid board by packaging waste containing aluminum and polyethylene. *J. Sci. Ind. Res.* **2011**, *70*, 232–234.
67. Hidalgo-Salazar, M.A.; Neves, F.L.; Baena, E. Possibilities of fabrication of aluminium polyethylene obtained from multilayer package recycling. *Inf. Técnico* **2013**, *77*, 100–109.
68. Hidalgo-Salazar, M.A.; Mina, J.H.; Herrera-Franco, P.J. The effect of interfacial adhesion on the creep behaviour of LDPE–Al–Fique composite materials. *Compos. Part B* **2013**, *55*, 345–351. [CrossRef]
69. Hidalgo-Salazar, M.A.; Muñoz, M.F.; Mina, J.H. Influence of incorporation of natural fibers on the physical, mechanical, and thermal properties of composites LDPE-Al reinforced with fique fibers. *Int. J. Polym. Sci.* **2015**, 1–8. [CrossRef]
70. Muñoz-Vélez, M.F.; Hidalgo-Salazar, M.A.; Mina-Hernández, J.H. Effect of content and surface modification of fique fibers on the properties of a low-density polyethylene (LDPE)-Al/Fique composite. *Polymers* **2018**, *10*, 1050. [CrossRef] [PubMed]
71. Ayrilmis, N.; Kaymakci, A.; Akbulut, T.; Elmas, G.M. Mechanical performance of composites based on wastes of polyethylene aluminum and lignocellulosics. *Compos. Part B* **2013**, *47*, 150–154. [CrossRef]
72. Ovchinnikov, V.; Mastalygina, E.E.; Pantyukhov, P. Investigation of novel polymer composites based on recycled multilayer combined packaging materials. *Solid State Phenom.* **2020**, *299*, 94–99. [CrossRef]

73. Valim, F.C.F.; Silveira, D.C.; Costa, M.L.; Pereira, M.S.; Botelho, E.C. Study of the morphological, thermal and mechanical properties of recycled aluminum and low density polyethylene particulate composite. *Matéria* **2015**, *20*, 852–865. [CrossRef]

74. Sánchez-Alvareza, A.; De la Torre Ibarrab, M.H.; Santoyo, F.M. Thermal performance comparison of recycled and clay roof tiles due heat radiation by digital holographic interferometry. *Rev. Mex. Física* **2019**, *65*, 678–683. [CrossRef]

75. Lecce, R.; Cerutti, S. Method for Recycling Composite Material. European Patent 2123418A1, 21 May 2008.

76. Cravero, F.; Frache, A. Improving fire performances of PEAL: More second-life options for recycled Tetra Pak®. *Polymers* **2020**, *12*, 2357. [CrossRef]

77. Johansson, H.; Ackermann, P.W. Method of Recovering Individual Component Parts from Packaging Material Waste. U.S. Patent 5421526, 27 May 1992.

78. Kaiser, K.; Schmid, M.; Schlummer, M. Recycling of polymer-based multilayer packaging: A review. *Recycling* **2018**, *3*, 1. [CrossRef]

79. Prawisudha, P.; Mu'min, G.F.; Yoshikawa, K.; Pasek, A.D. Experimental study on separation of metal layer in aluminum-plastic packaging by employing hydrothermal process. In Proceedings of the Advancement in Technology and Management for Tomorrow; 2014. Available online: https://www.semanticscholar.org/paper/Experimental-Study-on-Separation-of-Metal-Layer-in-Prawisudha-Mu%E2%80%99min/73dce3b6c9c000728e6920f6437f42a067eabd39 (accessed on 24 January 2021).

80. Mu'min, G.F.; Prawisudha, P.; Zaini, I.N.; Aziz, M.; Pasek, A.D. Municipal solid waste processing and separation employing wet torrefaction for alternative fuel production and aluminum reclamation. *Waste Manag.* **2017**, *67*, 106–120. [CrossRef] [PubMed]

81. Zhang, J.F.; Yan, D.H.; Li, Z.H. The recycling of the Tetra-Pak packages: Research on the wet process separation conditions of aluminum and polythene in the Tetra-Pak packages. In Proceedings of the 2009 3rd International Conference on Bioinformatics and Biomedical Engineering, Beijing, China, 11–13 June 2009.

82. Zhang, S.F.; Zhang, L.L.; Luo, K.; Sun, Z.X.; Mei, X.X. Separation properties of aluminium–plastic laminates in post-consumer Tetra Pak with mixed organic solvent. *Waste Manag. Res.* **2014**, *32*, 317–322. [CrossRef] [PubMed]

83. Zhang, S.; Luo, K.; Zhang, L.; Mei, X.; Cao, S.; Wang, B. Interfacial separation and characterization of Al–PE composites during delamination of post-consumer Tetra Pak materials. *J. Chem. Technol. Biotechnol.* **2015**, *90*, 1152–1159. [CrossRef]

84. Yan, D.; Peng, Z.; Liu, Y.; Huang, Q.; Xie, M.; Wang, Q. Optimizing and developing a continuous separation system for the wet process separation of aluminum and polyethylene in aseptic composite packaging waste. *Waste Manag.* **2015**, *35*, 21–28. [CrossRef]

85. Rodríguez-Gómez, J.E.; Silva-Reynoso, Y.Q.; Varela-Guerrero, V.; Núñez-Pineda, A.; Barrera-Díaz, C.E. Development of a process using waste vegetable oil for separation of aluminum and polyethylene from Tetra Pak. *Fuel* **2015**, *149*, 90–94. [CrossRef]

86. Samorì, C.; Cespi, D.; Blair, P.; Galletti, P.; Malferrari, D.; Passarini, F.; Vassura, I.; Tagliavini, E. Application of switchable hydrophilicity solvents for recycling multilayer packaging materials. *Green Chem.* **2017**, *19*, 1714–1720. [CrossRef]

87. Pappa, G.; Boukouvalas, C.; Giannaris, C.; Ntaras, N.; Zografos, V.; Magoulas, K.; Lygeros, A.; Tassios, D. The selective dissolution/precipitation technique for polymer recycling: A pilot unit application. *Resour. Conserv. Recycl.* **2001**, *34*, 33–44. [CrossRef]

88. Zhao, Y.B.; Lv, X.D.; Ni, H.G. Solvent-based separation and recycling of waste plastics: A review. *Chemosphere* **2018**, *209*, 707–720. [CrossRef]

89. Georgiopoulou, I.; Pappa, G.D.; Vouyiouka, S.N.; Magoulas, K. Recycling of post-consumer multilayer Tetra Pak® packaging with the Selective Dissolution-Precipitation process. *Resour. Conserv. Recycl.* **2021**, *165*, 105268. [CrossRef]

90. Tagliavini, E.; Galletti, P.; Samori, C.; Malferrari, D. Method for Separating and Recovering Polyethylene and Aluminum from a Polylaminate Material. World Patent WO 2018/087653 A2, 7 November 2017.

91. Mumladze, T.; Yousef, S.; Tatariants, M.; Kriūkienė, R.; Makarevicius, V.; Lukošiūtė, S.-I.; Bendikieneb, R.; Denafasa, G. Sustainable approach to recycling of multilayer flexible packaging using switchable hydrophilicity solvents. *Green Chem.* **2018**, *20*, 3604–3618. [CrossRef]

92. Nieminen, J.; Anugwoma, I.; Kallioinen, M.; Mänttäri, M. Green solvents in recovery of aluminium and plastic from waste pharmaceutical blister packaging. *Waste Manag.* **2020**, *107*, 20–27. [CrossRef] [PubMed]

93. Siddiqui, M.Z.; Park, Y.K.; Kang, Y.; Watanabe, A.; Kim, S.; Kim, Y.M. Effective use of aluminum-plastic laminate as a feedstock for catalytic pyrolysis over micro and mesoporous catalysts. *J. Clean. Prod.* **2019**, *229*, 1093–1101. [CrossRef]

94. Yin, S.; Rajarao, R.; Gong, B.; Wang, Y.; Kong, C.; Sahajwalla, V. Thermodelamination of metallised composite plastic: An innovative approach to generate aluminium from packaging plastic waste. *J. Clean. Prod.* **2019**, *211*, 321–329. [CrossRef]

95. Dave, P.N.; Joshi, A.K. Plasma pyrolysis and gasification of plastics waste—A review. *J. Sci. Ind. Res.* **2010**, *69*, 177–179.

96. Hepworth, M.T.; Tylko, J.K.; Han, H. Treatment of electric arc furnace dust with a sustained shockwave plasma reactor. *Waste Manag. Res.* **1993**, *11*, 415–427. [CrossRef]

97. European Commission. LIFE Program—Project CLEAN: Converting Laminates into Energy and Aluminium for the Benefit of Nature, LIFE06 ENV/E/000010. 2010. Available online: https://ec.europa.eu/environment/life/project/Projects/index.cfm?fuseaction=home.getProjects&themeID=70&projectList (accessed on 17 December 2020).

98. Ludlow-Palafox, C.; Chase, H.A. Microwave-induced pyrolysis of plastic wastes. *Ind. Eng. Chem. Res.* **2001**, *40*, 4749–4756. [CrossRef]

99. Ludlow-Palafox, C.; Chase, H.A. Microwave Induced Pyrolysis Reactor and Method. U.S. Patent 7951270 B2, 31 May 2011.