

Adhesives in packaging

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The use of adhesives in packaging is described. Both natural product and synthetic adhesives used in the industry are then discussed, with examples of their specific applications and estimates of their market share given. The paper goes on to present examples of packaging systems, detailing the way in which specific adhesives are employed in attaining the design requirements. Finally, future developments for adhesives in the packaging industry are addressed.

(Keywords: adhesives; packaging; overview)

INTRODUCTION

The total annual consumption of adhesives in Europe is approximately 1.2 million tonnes¹. The packaging industry alone accounts for at least 20% of this figure, although world-wide global consumption is reported² to be as high as 35%. Although the actual figures are dependent upon precise definitions of 'packaging', the industry is, most probably, the major single market user of adhesives.

Adhesives are used in packaging in three basic ways.

1. *To form the structure of the packaging material by combination of substrates.* With the possible exception of uncoated glass, packaging materials rarely comprise a single component. In order to achieve the requirements of optimum performance and fitness for purpose at a minimum of cost, packaging is often manufactured by combining basic substrates. These are most commonly:

- metal plate or sheet – tinplate, tin-free steel, aluminium;
- metal foil – mainly aluminium;
- metallized films – mainly aluminium-coated polypropylene and poly(ethylene terephthalate) (PET);
- commodity polymers – polyethylene, polypropylene, ethylene-acrylate ionomers, polystyrene, poly(vinyl chloride) (PVC), PET, polyamides;
- paper and board; and
- glass.

2. *To form the geometric shape of the package.* These processes include carton forming,

seam sealing, case sealing, lidding and heat sealing. Modern filling lines may incorporate form-fill-seal operations where the package is assembled on-line prior to filling or is formed around the contents. These operations are used in the paper and board industry which puts special demands on adhesive performance, in the flexible laminates sector (pouch heat sealing) and in the rigid plastics packaging sector where the container is thermoformed from sheet, filled and the lid heat-sealed in a continuous process.

3. *To apply labels and print and other, miscellaneous operations such as extrusion coating, lacquering and the manufacture of adhesive tapes (which may be used in category 2 operations).* If categories 1 and 2 may be regarded as primary functions for the use of adhesives in packaging, then category 3 is secondary but is still an important area for the use of adhesives in the industry.

ADHESIVES AND ADHESIVE FORMULATIONS

Both natural product and synthetic adhesives are used in packaging. *Table 1* lists the types of adhesives used in the industry with an estimate of their market share³. *Tables 2* and *3* give examples of specific applications for the natural product and synthetic adhesives, respectively⁴. The categories of use as defined above are included.

Adhesives are applied in various forms: as organic solutions (approximately 19% of the total) – these are mainly used in flexible lamination and some labelling operations involving 'difficult' surfaces; as aqueous solutions, dispersions or emulsions (46%) – these are

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Table 1 Packaging adhesives – type and market share

Adhesive type	% of total volume
Starch and dextrin	36
Poly(vinyl acetate) emulsions	27
Hot melts	17
Solvent-based elastomers	6
Rubber latices	6
Poly(vinyl alcohol)	3
Casein	3
Animal glues	1
Acrylic dispersions	1
Total	100

Source: ref. 3

mainly used in the paper and board sectors. Carrier-free adhesives (35%) take the form of hot-melts, powders, tapes, films and reactive liquid systems. The hot-melt adhesives are mainly used for high-speed container formation and some pressure-sensitive tape production whilst reactive liquids are used in the lamination sector.

Natural product adhesives continue to be important due to their relative cost advantage and easy clean-up when compared with their more versatile synthetic counterparts. The main area for use of natural product adhesives is in the paper and board sector. The adhesives are applied via roller, dauber, jet, etc. from aqueous solutions, pastes or emulsions and, depending on the porosity of the substrate, a high degree of tack may be obtained relatively quickly. However, synthetic dispersions based on poly(vinyl acetate) (PVA) and PVA copolymers offer a more versatile range of alternative adhesives for paper and board conversion and can be used on a wider range of surfaces than the natural product adhesives, including the more 'difficult' ones – polyethylene, polypropylene and varnished board.

Production speed is of primary importance in the packaging industry. For example, lamination line speeds of up to a few hundred metres a minute or container assembly rates of several hundred per minute may need to be attained to make a process economic. In the latter application, the carrier-based systems are being superseded by hot-melt adhesives based on ethylene–vinyl acetate copolymers. These adhesives may also be applied by jet and form a sound bond very quickly as heat is lost from the interface. The hot-melt adhesives also have additional advantages in being able to bond 'difficult' surfaces to paper and board, such as aluminium foil, polymer films and varnished board.

Environmental and safety factor awareness is driving the industry towards solventless systems. In the lamination of flexibles, traditionally the major market for organic solvent-based systems, there is increasing use of carrier-free, two- and three-part reactive liquid systems based on polyurethane chemistry. The components may be mixed *in situ* on the application roller and cured by a crosslinking mechanism accelerated by moisture absorption.

Adhesives rarely contain a single component. The

heart of any adhesive is, of course, the binder. However, adhesives are formulated to give the required properties to enable ease of application, control open time and wet tack, and stabilize the bond. Typically, an adhesive formulation may contain two or more of the following:

- binder – the polymer;
- carrier – water, organic solvent (except 100% solids);
- plasticizers – to reduce the glass transition temperature;
- tackifiers – often natural resins, to improve the cohesive strength of the adhesive film before solidification;
- thickeners and fillers – to control viscosity and increase solids content;
- surfactants – to improve wetting;
- biocides and fungicides – for natural product adhesives, paper and board adhesives;
- emulsifiers;
- waxes – rheology modifiers;
- antioxidants.

EXAMPLES OF PACKAGING SYSTEMS

When a package is designed or selected for a particular purpose, various criteria and requirements must be considered. These may include:

- cost;
- consumer appeal;
- content identification (labelling/printing);
- tamper evidence; and
- performance, e.g. mechanical strength, barrier (solvent, microbes, taint and odour, moisture, oxygen, carbon dioxide), temperature resistance (boiling water, sterilization, freezer temperatures and extreme ambient conditions), inertness to product.

Cost is often a primary factor in the selection of a packaging system and lamination technology may be used to provide a composite structure to meet performance requirements with optimum use of materials. One of the most demanding areas is in the manufacture of barrier containers to package food and drink products. The metal can or glass container offers the ultimate barrier but there is a large market for laminated structures which may comprise paper, board, metal foil, commodity and barrier polymers. Adhesives are required to combine these dissimilar materials, to provide a structure that is an optimal combination of cost and performance.

There are many examples of laminated structures on the supermarket shelves.

Chocolate wrapping. The fun-sized sweet packages that are currently popular are illustrative of the constraints of consumer appeal and cost criteria. Taking as an example the packaging of chocolates with a honeycomb centre, the outer containing bag is attractive due to its high gloss. This is typically

Table 2 Natural product adhesives

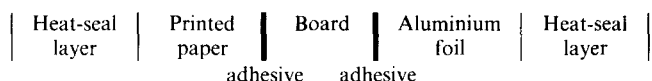
Adhesive type	Form	Applications	Category	Further comments
Animal glue	Solid, jelly, liquid	Cardboard box manufacture; gummed tapes	2 3	Very high tack for heavy-duty application
Casein	Aqueous paste	Wrap-around and patch labels for glass (coated) and metal cans; aluminium foil-to-board lamination	3 1	Starch may be added to give high-tack cream; added to rubber latex adhesives to increase creep resistance
Cellulose derivatives	Organic solution	Paper tissue and towel manufacture	1	Sometimes added to water-based adhesives to increase viscosity
Dextrin (also as synthetic dextrin made from e.g. sucrose)	Paste, latex, gum	Paper-to-board lamination; multi-ply tube winding; envelope side-seams; bag and sack seaming; form-fill-seal operations for bags and cartons; wrap-around label sealing	1 2 2 3	Prepared from maize, wheat or potato starch; may include borax to increase viscosity and consistency
Starch (unmodified)	Aqueous fluid	Corrugated board manufacture	1	Unmodified starch has low cost but is unstable and loses its adhesive qualities with time. Thermal and chemical modification confers long-term stability and a wider range of properties.
Starch (modified)	Aqueous paste	Aluminium foil-to-board lamination; wrap-around and patch labels for glass	1 3	
Starch (pregelatinized)	Water soluble white powder	Solid board lamination; paper sack manufacture;	1 2	Pregelatinized starch is made by rapid drying of pre-cooked starch slurry – to give a stable paste whilst retaining the advantages of unmodified starch. High levels of starch may be added to poly(vinyl acetate) emulsions to produce a high-solids paste for higher processing speed applications
Starch – PVA	Aqueous paste	Aluminium foil-to-board lamination; bonding to paper with difficult surfaces	1 1	
Rosin	Hot melt	Label 'pick-up' adhesive for metal cans	3	Low cost, low operating temperature, good adhesion to tinplate and glass
Rubber (natural)	Aqueous latex	Self-seal envelope flaps; cardboard tray assembly; biscuit and chocolate wrappers	3 2	Blended with acrylic resin to reduce self adhesion to give 'cold-seal' adhesives for chocolate wrappers
Sodium silicate (water glass)	Aqueous solution	Heavy duty tube winding; corrugated board manufacture	1	Gives a stiff but brittle bond
Waxes (natural and synthetic)	Hot melt	Lamination of board (detergent cartons); lamination of paper to aluminium (butter wrappers)	1	Specific use where water and grease barrier properties may be exploited

Table 3 Synthetic adhesives

Adhesive type	Form	Applications	Category	Further comments
Acrylics (acrylates, methacrylates and copolymers)	Aqueous emulsions, organic solutions	Heat-seal coatings for lidding materials; tape adhesives; label adhesive for glass and plastic bottles	1	Versatile range of adhesives available – pressure-sensitive and heat-seal adhesives; u.v. and low temperature resistant; growing application in tape and label markets
			3	
Ethylene-vinyl acetate copolymers	Hot melts	Used in high-speed operations such as case sealing	2	Grades with application temperatures from 100 to 180°C available. Additives include tackifiers and waxes to enable wetting and application. Susceptible to softening by solvents and oils
Polyurethanes	Organic solution, aqueous dispersion, hot melt, 1-, 2- or 3-part liquid	Lamination of flexible films	1	A versatile range of adhesives offering high heat and product resistance depending on the system used. They may be thermoplastic or form crosslinked networks to give maximum thermal stability
			2	
Poly(vinyl acetate)	Aqueous dispersion	Corrugated board manufacture; tube winding; carton forming; carton sealing	1	Not suitable for 'difficult' surfaces such as PE, PP, PVC; requires plasticization. Properties such as moisture resistance and heat resistance may be improved by copolymerization and crosslinking
Poly(vinyl alcohol)	Aqueous solution or filled dispersion	Solid board lamination; tube winding; remoistenable adhesive used for stamps or envelope flaps	3	
Styrene-butadiene and styrene-isoprene block copolymers; styrene-butadiene rubber	Organic solution, aqueous dispersion, hot melt	Pressure-sensitive adhesive for labels and tapes; sealing compounds for metal closures	3	Good creep resistance compared with SBR, u.v. resistance not as good as acrylics
			1	
Vinyl acetate-ethylene copolymers	Aqueous dispersion	Lamination of PP film to printed board; carton sealing; labelling of wet plastic bottles	1	Wide range of copolymers available – versatile adhesive which will bond to many substrates including varnished board
			2	
			3	
Modified polyolefins	Co-extrudable hot melts	Polymer-to-polymer and polymer-to-metal lamination	1	Variety of grades, depending on application – modification generally anhydride or acid type

oriented polypropylene (OPP) film, reverse printed (to display the gloss) and containing an internal heat-seal layer (e.g. ethylene-acrylate copolymer). The heat-seal layer is bonded to the OPP using adhesive lamination, for example, with a polyurethane adhesive. The individual 'fun-size' bags are monolayer, printed OPP. There is no advantage in reverse printing these bags as they do not need to display gloss. The individual bags are sealed using a pressure-sensitive or cold-seal adhesive as heat sealing is usually avoided for chocolate wrapping!

Barrier fruit juice cartons. These cartons derive their mechanical strength from the cardboard and excellent oxygen barrier from aluminium foil. A typical structure may be:



The carton is formed by heat sealing the outer and inner layers which may be extrusion coated on to the paper and aluminium substrates. The heat-seal layers are generally polyolefin-based and may be ionomers. An adhesive lamination process will be used at both of the board interfaces – possibly using a starch-PVA paste.

Barrier lamination processes

Adhesive lamination. The availability of polymers with low gas permeability has extended the development of the barrier laminate market by providing alternatives to aluminium foil and metallized films. Tables 4 and 5 list the oxygen and moisture permeability coefficients of common packaging polymers. The high gas barrier polymers generally used in packaging are poly(vinylidene chloride) (PVDC) and, more usually, ethylene-vinyl alcohol (EVOH) copolymers. These polymers are relatively expensive and therefore are used economically in laminate structures. The polymers offering the best moisture barrier, e.g. polyolefins, are the least expensive and are often used to provide a heat-seal layer and possibly mechanical strength to the laminated structure – which then has an ideal combination of gas and moisture barrier.

Within the last two years, Airco Coating Technology announced the successful development of an economic, silicone-dioxide-coated PET film⁵. The sub-micrometre layer is formed by plasma deposition from an organo-silicon/oxygen/helium gas mixture and gives a barrier performance similar to or better than that of EVOH and PVDC laminates. A heat-seal layer may be applied to the coating which is reported as being exploited as the barrier layer in some milk and fruit juice cartons.

An example of the use of a flexible barrier laminate is the manufacture of a lidding film, a typical structure being:

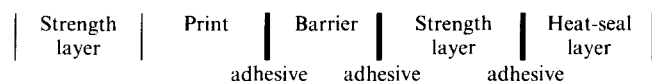


Table 4 Oxygen permeability coefficients of some common polymers

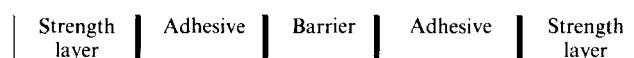
Polymer	Permeability coefficient (cm ³ mm/m ² atm 24 h)
Low density polyethylene	200
High impact polystyrene	150
Polypropylene	70
High density polyethylene	60
Poly(vinyl chloride)	4.3
PET – amorphous	2.9
PET – oriented	1.2
Ethylene-vinyl alcohol copolymer (EVOH) at 100% r.h.	0.50
Poly(acrylonitrile)	0.40
PVDC – VC copolymer	0.060
PVDC latex	0.043
EVOH at 75% r.h.	0.040
PVDC – MA copolymer	0.025
PVOH at 50% r.h.	0.020
EVOH at 50% r.h.	0.009
EVOH at 0% r.h.	0.005
PVOH at 0% r.h.	0.0001

Table 5 Moisture vapour permeability coefficients of some common polymers

Polymer	Moisture vapour permeability coefficient (g mm/m ² 24 h)
Low density polyethylene	0.08–0.4
High density polyethylene	0.01–0.07
Polypropylene	0.03–0.07
PVDC	0.004–0.4
PET	0.2–0.9
Poly(acrylonitrile)	0.5
Nylon 6	2
EVOH	3

The adhesive system would most probably be polyurethane-based to provide product resistance and the ability to withstand a sterilization cycle – heating in an autoclave at a temperature between 121 and 135°C for times up to 1 hour. The strength layers may typically be polyester, the barrier layer may be foil, metallized film or a barrier polymer based on PVDC or more preferably EVOH. The heat-seal layer would be carefully selected to be capable of withstanding the sterilization cycle, yet provide peelability to the consumer.

Co-extrusion processes. Rigid and flexible barrier packaging may also be produced by co-extruding various layers in film, sheet or blow moulding processes. For example, plastic sauce and ketchup bottles comprise a barrier laminate structure made using extrusion-blow moulding technology. The structure of these laminates is generally:



The strength layers are usually polyethylene (PE) or polypropylene (PP). The barrier layer is based on PVDC or, more usually, EVOH. The tie layer polymer would be selected from a wide range of co-extrudable melt adhesives which have been developed by many of

the leading polymer suppliers. The 'Bynel' range from Du Pont is one example of a series based on ethylene-vinyl acetate copolymers modified with carbonyl, acid, acrylate or anhydride groups. An alternative series of adhesives is derived from polypropylene or high, low and linear-low density polyethylenes modified with anhydride groups. Depending on the application, the level of modification ranges from <10 to >20%. The packaging converter selects the grade suitable to his requirements. This class of adhesive may be used to combine almost all types of substrates – paper, metals and polymers.

The PP and PE adhesive grades have higher heat resistance than the EVA-based products and are used in food packaging which requires pasteurization or sterilization.

There is some overlap between the range of flexible laminates produced by adhesive lamination and co-extrusion lamination. Co-extrusion lamination may offer a more cost-effective production route, requiring only one step as all the materials are combined in one stage. As with adhesive lamination, co-extrusion lamination can also be used to bond layers to printed film or paper or to an oriented polymer web.

Adhesives and the metal can

Metal-to-polymer lamination. The use of tinplate containers to package foodstuffs was first commercialized in the early 19th century. Then, food was packed using plain tinplate but over the last 50 years the inertness of the package to some products has been enhanced by applying a lacquer coating to the internal surface of the container. Modern internal lacquers are usually epoxy-based and are applied by coating or spraying the metal surface using a carrier-based system. Recent developments are directed towards reducing the volatile organic components (VOC) content of lacquer formulations and extending the use of water-based systems.

Today, metal cans, trays and components (ends, closures) are manufactured from tinplate, tin-free steel (TFS) or aluminium. Lacquering remains the main method of separating the reactive metal surface from the product, but alternative processes are being developed where the metal substrate is protected by lamination of a polymer film. One such system is called Ferrolite, developed by CarnaudMetalbox Technology⁶.

Ferrolite comprises a steel substrate (tinplate or TFS) laminated on each side with polyester, polypropylene or nylon/polypropylene laminated film, or any combination of these. The films are laminated directly from reels on to the steel coil in a continuous process (Figure 1), by the application of heat and pressure. The adhesion of the films to the substrate is enhanced by the presence of a co-extruded tie or bonding layer. The tie layer is typically 10% of the total film thickness which usually ranges from 12 to 40 μm (Figure 2).

The PP-based film is either a homopolymer or an ethylene-propylene copolymer. The co-extruded

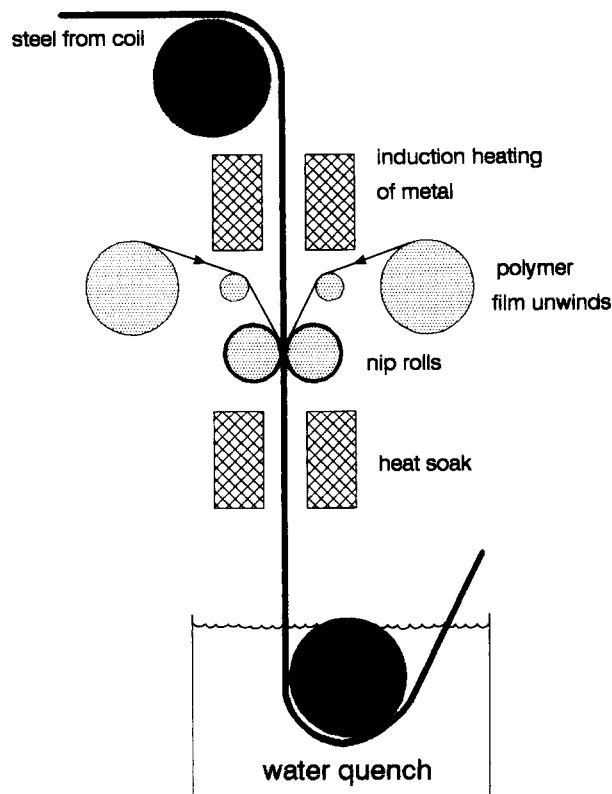


Figure 1 Schematic of metal-polymer lamination process

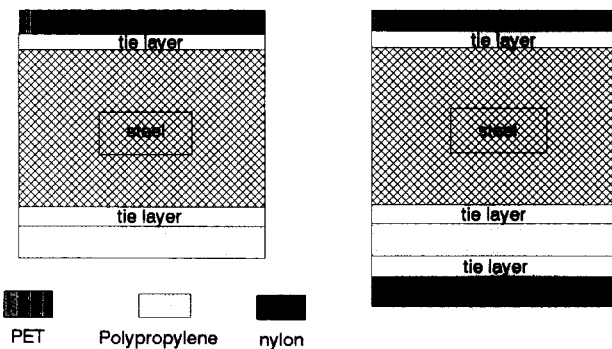


Figure 2 Some Ferrolite laminate structures

bonding resin is an acid- or anhydride-modified polypropylene layer. The polyester-based film comprises a biaxially oriented PET layer co-extruded with a low melting point polyester copolymer to provide a high-tack layer during the thermal lamination process. The nylon/polypropylene film is a four-layer structure with similar tie layers bonding the nylon to the PP as bonds the PP to the steel substrate. The outer nylon layer provides a harder, lower friction surface compared with PP, an advantage in some forming operations.

Metal-polymer laminated products are currently used as components in aerosol cans, food can ends and shallow drawn containers. The main advantages of this technology over traditional lacquering are in the elimination of solvents and carriers, using coil rather than sheet processes and potentially significant energy saving from light-weighting of the metal substrate.

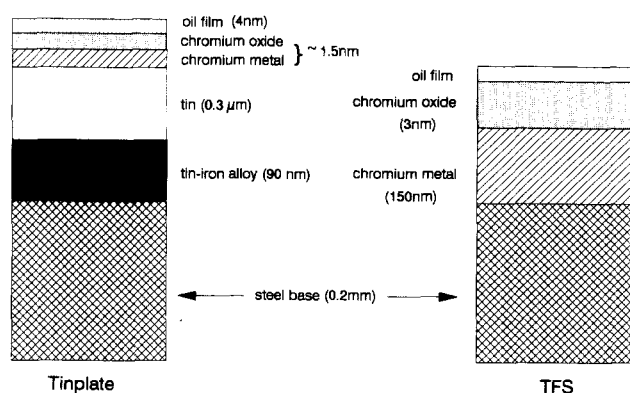


Figure 3 Schematic representation of tinplate and TFS structures

The adhesively bonded can. The majority of food cans are three-piece containers. That is, they are produced from a cylindrical body with a closure seamed on to either end. As mentioned above, the industrial production of three-piece tinplate cans started in the early 19th century and involved soldering the edges of the plate blank to form the cylindrical can body. In the last 20 years, the soldering process has been largely replaced by welding technology – which still requires the use of tinplate to form a high-integrity welded seam. Over a similar time period, the threat of rising tin prices has led to the development of tin-free steel (TFS) for the manufacture of metal cans and components. Figure 3 gives a schematic representation of the structures of tinplate and TFS. The technology to weld TFS is not currently available and these containers are formed or drawn from flat blanks to give the two-piece can – a body with one open end.

Japanese technologists have developed an alternative approach to manufacturing TFS three-piece cans and for the last 20 years have been producing an adhesively bonded TFS can. That is, the can body is formed by using an adhesive to bond the side seam⁷. Initially, these cans were used for beer and soft drinks, where the performance criterion required the container only to withstand pasteurization cycles (heating to 100°C).

Systematic developments of the metal primer system, the adhesive and the TFS surface chemistry have led to a high-speed process producing three-piece cans capable of withstanding food sterilization cycles. The key development has been optimization of the epoxy-based TFS primer system. The adhesive is applied to the primed blank edge as a tape of nylon 12 blended with a polyamide copolymer. The blending gives an optimum combination of the requisite structure and properties – controlled crystallinity, water resistance, adhesive strength and toughness (required to withstand end seaming and body beading operations). The tape is adhered to the primed body blanks by the application and removal of heat and pressure in a highly controlled process. Production speeds of up to 900 can bodies per minute have been claimed – comparable with welding technology.

FUTURE DEVELOPMENTS FOR ADHESIVES IN PACKAGING

Environmental

The immediate motivators for developments of packaging adhesives are environmental legislation and health and safety issues. There is a drive to reduce the need for organic solvent-based systems and to reduce generally the VOC content of water-based systems. For example, adhesive suppliers will not be able to incorporate chlorinated solvents in water-based emulsions from the end of 1994¹. The use of hot-melt adhesives and carrier-free systems is therefore expected to continue to expand in the paper and board, labelling and flexible lamination sectors.

With the continuing requirement to increase line speeds, especially in the paper and board industry, the use of hot-melt adhesives for sealing and some laminating processes may be expected to increase at the expense of aqueous systems. Alternatively, carton blanks may be supplied pre-coated with a heat-seal adhesive, negating the requirement to have on-line applicators at the filling and forming stages¹.

In order to conserve energy, lower melting point (warm melt) adhesives are becoming available and may find special applications where either high (softening) or low (embrittlement) temperature resistance is not a critical requirement.

There is an awareness within the packaging industry of its responsibilities towards the environment beyond reduction in VOC levels. Life-cycle analysis (LCA) is being adopted by leading packaging converters and adhesive suppliers, to evaluate the relative merits of different processes and production routes. Therefore, it is envisaged that there will be a continuing development of metal-to-polymer lamination technology, which has been shown to have environmental advantages over the more traditional lacquering processes for the manufacture of metal containers.

Recycling of packaging materials is included in LCA and synthetic adhesives that offer easier 'clean-up' are being developed¹. These adhesives may be separated from the substrate without leaving a residue or, in the paper and board industry, may be filtered from the pulp in the recycling plant. However, further developments in this area are required to achieve complete and trouble-free regeneration of substrate materials.

An alternative approach to separating the adhesive from the substrate is the development of biodegradable products. One such hot-melt adhesive has been made from microbes and is reportedly used for case sealing².

Labelling

The labelling sector is also undergoing technological developments. Labels are becoming more sophisticated and are being exploited to provide more functions than

merely identification of the package contents. Indicator labels, showing the thermal history of the package (for food), and advanced anti-theft and tamper-evident labels and seals are being developed⁸. The higher specification labels may also be expected to impact on the performance requirements of the label adhesive. Novel application processes, such as in-mould labelling, are providing a developing market for specialized heat-seal coatings capable of withstanding polymer melt processing temperatures.

Testing

The packaging industry has developed many test procedures over the years which are often comparative and specific to end-use requirements⁹. With the exception of the paper and board sector, there has been little effort devoted to standardizing methods, in order to establish bench-marks and specifications. Initiatives have been taken to address this problem in a collaborative EUREKA project involving many industrial sectors and including the packaging industry¹⁰. The result of this study will be the development of a generic model and 'tool kit' which should establish procedures to assure the quality of adhesive bonds.

It may be hoped that the recommendations from this study will point the way to the acceptance of common practices for the evaluation of adhesive bonds in packaging.

CONCLUSIONS

There is little doubt that the packaging industry will continue to be a major consumer of adhesives for the foreseeable future. As confidence grows in the capabilities of adhesive bonding, developing technologies in the engineering and the construction industries may be transferred to the packaging sector, impacting not only on the packaging materials but also on production and handling equipment. In the short term, developments are directed towards meeting exacting environmental demands on packaging and formulating adhesives to enable novel and more rapid manufacturing and assembly processes.

REFERENCES

- 1 *Packaging Week* June 1994, 10(8)
- 2 *Performance Chemicals* December 1993/January 1994
- 3 'MSI Data Report on Adhesives and Sealants: UK', 1992
- 4 Lazarus, D.M. 'Industrial Packaging Adhesives' (ed. K. Booth), Blackie, 1990, Ch. 2
- 5 *Plastics World* August 1993
- 6 Noke, A.C. 'International Tinplate Conference', 1992
- 7 Kobayashi, S. and Miyazawa, T. *Prog. Org. Coat.* 1993, 23, 23
- 8 *Packaging Week* September 1994, 10(16)
- 9 Cochran, M.A. and Allen, K.W. 'DTI MTS Project 3, Environmental Durability of Adhesive Bonds, Report No. 1', January 1994, Ch. 5
- 10 Espie, A.W., Rogerson, J.H. and Ebtehaj, K. *Int. J. Adhes. Adhes.* 1995, 15(2), 81