EB Technology for Pressure-Sensitive Adhesive Applications

By Stephen C. Lapin, Ph.D.

Initial reports on the use of ultraviolet (UV) and electron beam (EB) technology for pressure-sensitive adhesives (PSAs) began to appear in the late 1960s. It has been an active area of interest indicating continued belief in the technology. The main advantages are:

1. Improved shear, and heat- and chemical-resistance properties of crosslinked adhesives.
2. Application benefits such as pattern printing and elimination of pot life issues for chemical crosslinking systems.
3. Environmental benefits such as the elimination of volatile organic compounds and reduced energy usage for drying.[1]

Explosive growth in UV/EB PSA technology was predicted but never materialized. Accurate data on the use of UV/EB technology has been difficult to obtain since most companies that convert UV/EB PSAs also manufacture their own adhesives (captive production). A 2003 study by ChemQuest estimated that 1.5 million pounds of UV PSAs were being processed annually. It was estimated that 97% of this volume was captive. The same study estimates that nine million pounds of PSAs were processed using EB technology. Nearly 100% of EB adhesive production was captive.[2]

Much of the effort to introduce UV/EB PSAs to the merchant marketplace was focused on UV technology. This was likely due in part to the lower capital cost of UV equipment compared to EB equipment. In addition, PSA suppliers often had UV equipment available but had limited access to EB equipment for development purposes. In most cases, UV PSAs did not meet the expectations of the marketplace. Adhesive performance often did not justify higher applied cost. Consistent performance may also have been an issue due to the need to UV cure within a small process window.[3]

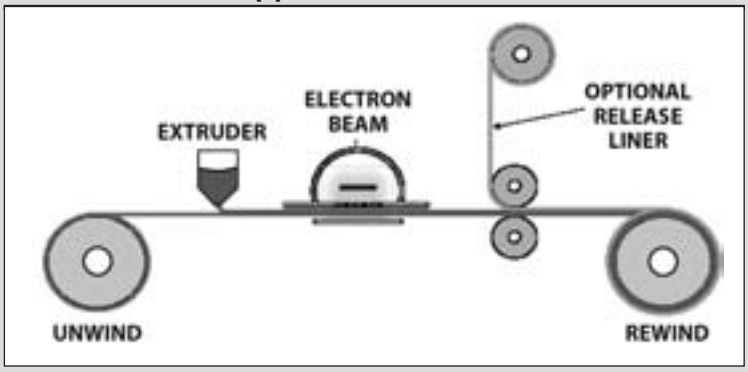
EB technology is quite different from UV. The accelerated electrons will directly ionize polymers without the need for photoinitiators. This provides the potential to crosslink compositions that are similar to conventional PSAs. This helps reduce the applied cost of the adhesive. Another advantage of EB is its very consistent output. This includes uniform energy deposition through the thickness of the adhesive as well as across the web. EB output is also very constant with time.

EB PSA Technology

EB processing of PSAs may include curing and crosslinking. Curing involves the polymerization of low-molecular weight polymer precursors (monomers and oligomers) with reactive (acrylate) end groups. Crosslinking involves the creation of covalent bonds between existing

FIGURE 1

EB hot-melt PSA application



high-molecular weight polymer chains. EB PSA systems are also known that combine curing and crosslinking. Examples include multifunctional acrylate monomers blended with adhesive polymers and tackifiers.[4]

EB PSAs may exist in a variety of forms, including syrups, solvent-based, water-based and hot- or warm-melt based systems.

Syrups are solvent- and water-free liquid systems based on formulated monomers, oligomers, tackifying resins and additives. The specialty nature of the raw materials can result in a relatively high applied cost. Syrups cure upon EB exposure to give a pressure-sensitive material. Syrups may be formulated for properties ranging from high tack/low shear to low tack/high shear. The properties of these adhesive are highly dependent on the degree of cure. High-performance properties are not expected with syrups. The main

TABLE 1

EB crosslinked PSA patents

U.S. Patent No.	Chemistry Type	Summary
4,133,731 (now expired)	A-B-A styrenic block copolymer blended with tackifier and multifunctional acrylate monomers	EB irradiation greatly improves stripability temperature limit
4,151,057 (now expired)	A-B-A styrenic block copolymer containing hydrogenated diene blocks blended with tackifier and multifunctional acrylate monomers	EB irradiation greatly improves high temperature performance
4,432,848 (now expired)	A-B (polystyrene-polyisoprene) copolymer blended with tackifier and multifunctional acrylate monomers	EB irradiation greatly improves solvent and heat resistance
5,066,728 (now expired)	Phenylbutadiene multi-block copolymer blend with tackifier	EB irradiation improves SAFT and shear with no loss of adhesive properties
5,104,921 (now expired)	Branched block copolymer with low unsaturation index	Greatly improved SAFT and shear upon EB irradiation
5,296,547	Block copolymers with mixed molecular weight end-blocks	Greatly improved shear properties upon EB irradiation
5,714,548	Star block copolymers	Greatly improved shear properties upon EB irradiation
6,630,531	Polymodyl asymmetric elastomeric block copolymers	Transfer and foam tapes with enhanced bonding to low energy surfaces
6,887,919	Star block copolymers blended with tackifier	EB irradiation enhances removability by stretching of the adhesive

TABLE 2

PSA performance as a function of EB irradiation

EB Dose (kGy)	SAFT (°C)	95°C Holding Power (min)
0	73	1
19	114	166
29	118	378
48	117	>1,000

advantage is for pattern printing (flexo or screen) which may be integrated with other printing or converting processes. UV-curable syrups are fairly well known;[5] however, EB-curable syrups have limited commercial use.

EB crosslinking of solvent- and water-based, pressure-sensitive adhesives is known. The process involves application of the PSA (direct or transfer coated on a release liner). The solvent or water is then evaporated using a conventional thermal dryer. This is followed by EB crosslinking of the resulting adhesive layer. The advantages of these systems are improved process control and pot life compared to chemical crosslinking systems. EB also has the potential to provide improved bonding to the substrate by a grafting mechanism. There are limited commercial applications of solvent- or water-based EB PSAs.

Crosslinked hot-melt adhesives are the most common type of EB PSA. In its simplest form, the process involves extrusion of the hot-melt or warm-melt adhesive followed by in-line EB irradiation (Figure 1). Variations include EB irradiation through a release liner and EB irradiation on a transfer drum.[6] Crosslinking of hot-melt PSAs provides improved shear, heat and solvent resistance. There are many patents on EB crosslinked, hot-melt adhesive systems. A sample of representative U.S. patents is given in Table 1.

Table 2 shows data from U.S. Patent 5,104,921. The shear adhesion failure temperature (SAFT) of this adhesive system increased upon EB irradiation at 19 kGy and then shows little additional increase at 29 and 48 kGy. The holding power increased dramatically upon EB irradiation and then continues to increase to more than 1,000 minutes after 48 kGy.

Table 3 shows data from U.S. Patent 5,296,547. The adhesive performance of four different polymers (blended with tackifier and plasticizer) are shown with and without EB irradiation. The peel strength of all four polymers is only slightly affected while the lap shear (LSP) shows a dramatic enhancement as a result of EB irradiation.

Figure 2 is from U.S. Patent 4,133,731. The adhesive systems in this case are blended with multifunctional (meth)acrylate monomers (coupling agents). The contour plot shows the stripability temperature limit (STL). The region above the contour is where no adhesive residue remains upon peeling at 400°F. The results show that lower EB doses are effective in maintaining good STL as the amount of coupling agent is increased. A comparison of hexanediol diacrylate (HDODA) and hexanediol methacrylate (HDODM) coupling agents shows that the acrylate is more effective than the corresponding methacrylate.

A more recent study showed that the lower end (<125 kV) of the EB energy spectrum was effective for crosslinking relatively thin (1.6 mil) layers of pressure-sensitive adhesives. The study also showed that the EB conditions (including dose, voltage, foil material/thickness and air gap) could be adjusted to give optimum adhesive performance while minimizing the effect on the underlying substrate. The conditions were predicted by Monte Carlo depth/dose calculations

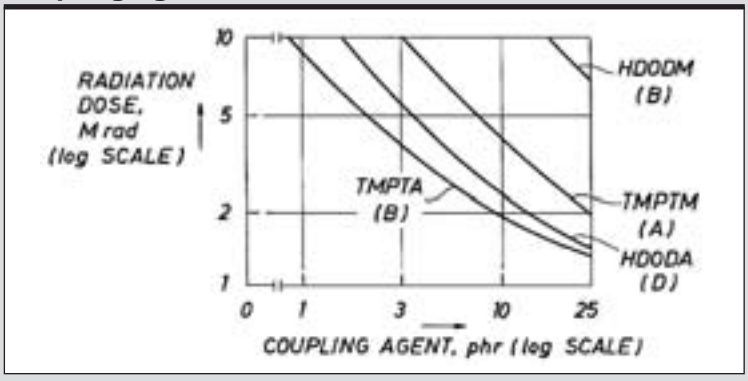
TABLE 3

Adhesive polymer enhancement with EB irradiation

Polymer	EB Dose (kGy)	Peel Force (N/m)	LSP (min)
A	0	490	193
A	50	280	4,000
B	0	595	399
B	50	385	6,000
C	0	595	546
C	50	332	5,000
D	0	472	3.2
D	50	770	10,000

FIGURE 2

400°F Stripability as a function of EB dose and coupling agent concentration



and confirmed by adhesive and substrate testing.[7]

Advanced research in EB processing showed the possibility of forming pressure-sensitive materials by irradiation of monomers directly on the substrate. Pulsed electron beams were used to enhance the structure of the polymer that was formed. This resulted in improved adhesive properties compared to polymers formed by continuous EB irradiation.[8]

The enhancement in pressure-sensitive adhesive performance resulting from EB irradiation is clear in these references. These performance enhancements help drive the use of EB technology.

EB Equipment

Commercial self-shielded EB processors have been available for more than 30 years. This EB equipment has proven long-term reliability in many industrial applications. EB processors generate accelerated electrons using electrically operated filaments within a vacuum chamber. Electrons are accelerated though a thin metal foil window and impinge on a moving web at atmospheric pressure.

EB processes are characterized by the dose, throughput and accelerating voltage of the systems. A typical EB dose range for curing or crosslinking

FIGURE 3

Depth/dose profiles for new-generation EB processors

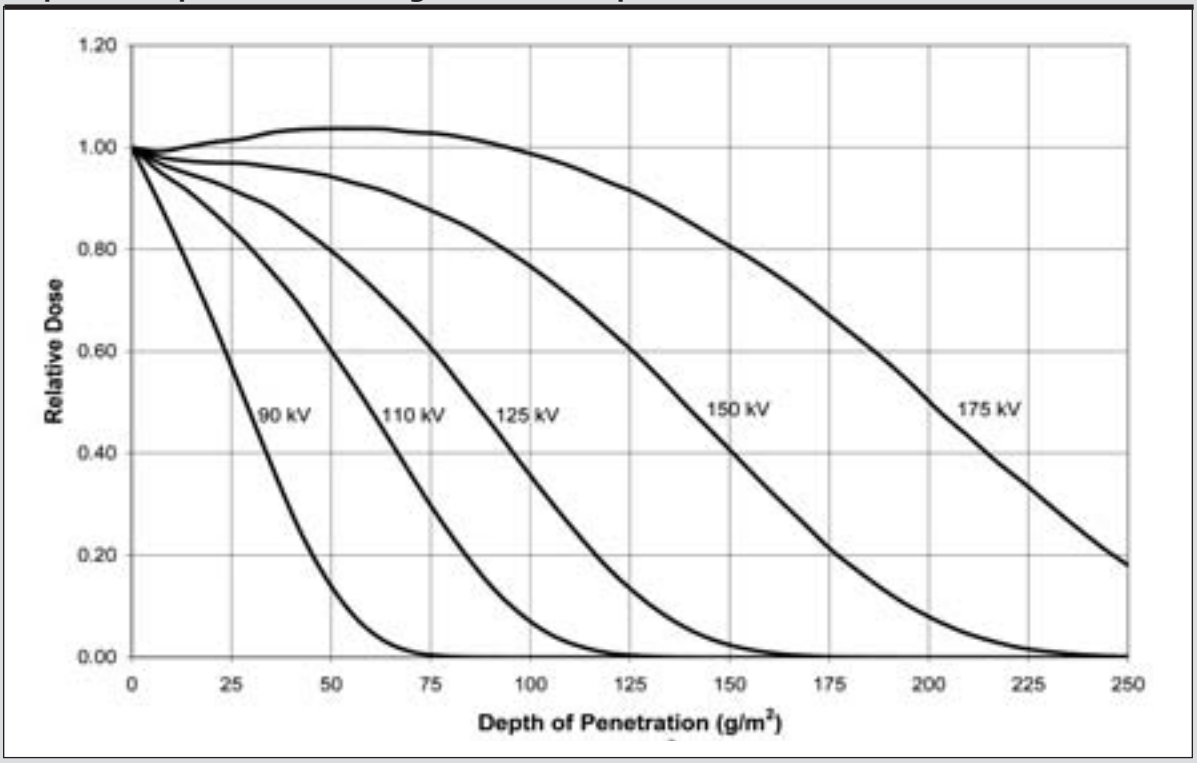


Figure 4

Industrial EB processor (150 to 300 kV) on left compared to new-generation lower (80 to 150 kV) EB unit

PSAs is from about 20 to 100 kGy (2 to 10 Mrads). Industrial EB processors are capable of very high throughput, delivering a 50 kGy dose at over 300 m/min.

The accelerating voltage of the EB determines the penetration into the material being processed. Industrial low-voltage energy electron beam processors historically operated in the range of about 150 to 300 kV. A 300 kV EB is capable of penetrating typical PSAs as thick as about 400 microns (16 mils)(thickness that results in 20% or less energy loss at the backside of the adhesive layer).

A new generation of lower energy EB processors were introduced about 10 years ago.[9] This equipment operated in the range of about 90 to 125 kV. This energy range is well-suited for EB curing of coatings and inks for printing and packaging applications. The 125 kV voltage is also useful for processing adhesive layers up to 50 microns (2 mils) thick (thickness that results in 20% or less energy loss at the backside of the adhesive layer). More recently, new low-voltage equipment has been developed that operates up to 150 kV.[10] According to the most recent report, this range has now been extended to 175 kV.[11] A depth/dose profile for this low-energy EB equipment is shown in Figure 3. The 175 kV curve shows energy losses of less than 20% for adhesive weights of 150 g/m^2. This corresponds to an adhesive thickness of 150 microns (6 mils) for an adhesive with a density of 1.0 g/cm^3. This penetration depth should be quite useful for a number of PSA applications. It also allows for the possibility of irradiation through the substrate or release liner being used in the converting process.

Photographs comparing an industrial EB processor with a new-generation, lower energy processor are shown in Figure 4. Other benefits of new-generation EB equipment have been described, including:

- Much lower capital costs compared to industrial EB processors
- Smaller size allowing integration into new and existing PSA converting lines
- Low operating and maintenance costs
- Outstanding uniformity (<8% variation across the web)
- High throughput (30 kGy at 400 m/min)
- Proven reliability
- Integrated chill rolls providing excellent web handling and temperature control[10]

Many of the benefits of the new EB equipment are attractive for use in PSA applications.

Conclusions

EB technology has been used for many years in PSA applications. A significant enhancement in hot-melt adhesive properties has been shown in numerous patent references. Many of these early patents have now expired which opens the possibility for development by merchant adhesive

suppliers. The availability of new-generation EB equipment operating up to 175 kV should make this technology attractive to a larger number of converters of pressure-sensitive materials. ◗

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