#### SUSTAINABILITY FOR PRESSURE SENSITIVE ADHESIVES USED IN LABELS

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#### Introduction:

Sustainability is a major driver in many packaging applications and is becoming increasingly important to consumers and brand owners. Sustainability for pressure sensitive adhesives (PSAs) for labels is currently not well defined. This paper will begin with a discussion on the sustainability of labels in general and the differences and challenges of each type of label adhesive. It will then transition into detailing several routes to a more sustainable pressure sensitive label adhesive. These will include biodegradable, compostable, repulpable, wash-off, removable, dissolvable, and recycling enabling. We will explore each route in detail, including the technical and consumer behavior challenges, the certifications (if any) that are applicable in each case and testing that may be required. The focus will be on aqueous acrylic PSA's.

Sustainability in packaging is becoming a focus for brand owners, consumers, and legislators. 230+ bills were introduced in 40 states in 2019 focusing on packaging waste as well as a comprehensive federal bill (Udall bill).<sup>1</sup> Brands as diverse as Hellman's Mayonnaise (Unilever), Molson Coors and Nestle are making changes to their packaging to reflect a renewed commitment to sustainability.<sup>2-4</sup>Consumer interest can be gauged by exploring Google Trends where a search for Sustainable Packaging shows a threefold frequency increase from 2015 to 2023. (FIGURE 1).

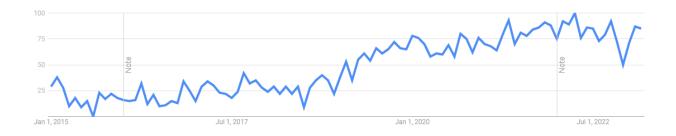


Figure 1- Google Trends searches for Sustainable Packaging

Labels are an integral part of packaging whether they are basic bar codes, address labels on inexpensive paper facestocks, highly decorated film labels for luxury goods like fine wine or high end cosmetics. In the past labels have not been an area of emphasis for those focused on sustainability in packaging, but this is beginning to change due to recycling issues and more focus by consumers on all areas of sustainably packaging. There have been labels and tapes introduced in the last 3 years claiming various sustainable characteristics including down gauging (using less film or paper), renewable forest certification for paper facestocks,<sup>5</sup> and post-consumer recycled content (PCR) facestocks. In addition, most attention has been paid to silicone release liner and matrix recycling which are beyond the scope of this paper.

In addition to the film and paper focus on sustainability, much attention is beginning to accumulate on both the adhesive and the overall label functionality itself. Many companies are focusing development resources on repulpability, compostability, ability to wash off and others. Some of these claims are backed up with various certifications and others are not. We will explore several of these routes to sustainability below as well as present some case studies.

### Challenges of Creating a Sustainable Adhesive Product for Labels and Tapes

Many sustainable solutions require newer or differentiating technologies which may increase the cost of components. Some of these sustainability claims require certifications while others have no current certifications available. The issue is further complicated by the fact that the label or tape makes up a small percentage of the package to be recycled so often times the focus is in other areas.

There are three main routes to developing a sustainable pressure sensitive adhesive:

- 1. Enable a change in consumer behavior
- 2. Design for recyclability
- 3. Reduce end of life impact

## **REMOVABLE ADHESIVES (Enable a change in consumer behavior)**

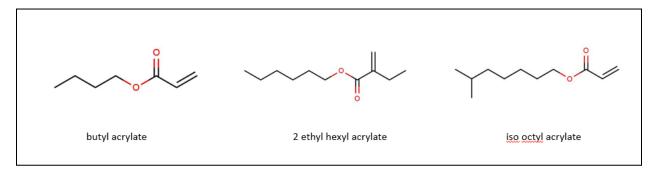
One route to achieving label or tape sustainability is through the use of more removable (low peel adhesion) adhesives. This is well known technology that exists today via many technical approaches. A removable adhesive (and hence a removable label or tape) would guarantee that no part of the structure would interfere with any downstream activities – whether they are container re-use, biodegradation or substrate recycling. The big drawback, however, is that this approach necessitates either consumer behavior changes or some other method of mechanically removing the structure. In addition, there might be product security concerns with the structure being removed prior to the product itself being used (i.e., in the supply chain, on a store shelf, or at home).

## **REPULPABLE ADHESIVES (Design for recyclability)**

Repulping is a way to reclaim paper fibers for reuse. Common repulping methods use highshear mechanical stresses with steam injection and/or chemical additives. Screens are used to remove contaminants. These screens can quickly become plugged – a process that can be exacerbated by sticky residue. A PSA coated on a label that is adhered to a paper or paperboard substrate can interfere with the reclamation of the fibers in this process. The PSA can cause issues with both the equipment used in the repulping process and can contaminate the finished paper product. A PSA that does not significantly fragment and is therefore easy to remove via filtration during the repulping process is desired in these applications. In the 1990s the United States Postal Service instituted a program for the design of a repulpable pressure sensitive adhesive for use in PSA postage stamps.<sup>6</sup> Careful design of the PSA can enable this behavior. For aqueous acrylic adhesives the distribution of acid in the backbone as well as the degree and type of neutralizer has been found to have a significant effect on the repulping behavior. More surface acid can lead to an increase in redispersability which potentially can result in the reduction of the "stickies" that contaminate the filter screens. This is well described in the literature.<sup>7a-d</sup>

# **COMPOSTABLE ADHESIVES (Reduce end of life impact)**

Most PSAs are purposefully constructed for excellent aging characteristics. This is especially true of aqueous acrylic adhesives. While some of these adhesives contain co-monomers that contain double or triple bonds and functional groups (styrene, acrylonitrile, vinyl acetate etc.) the acrylics themselves are fully saturated carbon-carbon bonds. FIGURE 2 shows the acrylic monomers that are typically used in PSAs: butyl acrylate (BA), 2ethylhexyl acrylate (2EHA) and iso-octyl acrylate. (iOA).



### Figure 2: Typical Acrylate Monomers used in aqueous PSAs

All three acrylates polymerize through the double bond via free radical polymerization. These also tend to be long chain molecules with very high molecular weights. In most cases the Mw and Mn cannot be fully measured via gel permeation chromatography and, therefore, other methods such as gel fraction must be used to estimate and compare molecular weights of different polymers.<sup>8</sup>

All of this leads to a very stable product that is not likely to be broken down into smaller pieces in a landfill or compost pile and therefore not viable for biodegradable or compostable certification. What is needed is a "weak point" in the backbone that enables breakdown of the chain into smaller molecules. This can be achieved in several ways.

Sugar, cellulose or some other highly susceptible moiety can be polymerized into the backbone to create this weak point. Polymers with high content of vinyl acetate (FIGURE 3) in the backbone (and subsequent side-chain hydrolysis) can introduce sufficient 1,3-diol content to promote subsequent enzymatic oxidations and eventual backbone cleavage.<sup>9-10</sup>

Figure 3: Vinyl Acetate

The use of high amounts of vinyl acetate can result in a major issue however: the PSA performance and aging characteristics may be severely impacted using high levels. The other drawback in creating an adhesive that breaks down is engineering it to break down at the right time – after the useful product lifetime. If the adhesive breaks down before this – i.e., in storage or in use, this can lead to product failure. FIGURE 4 below shows the typical product life for a PSA used in label constructions. Many adhesives are stored for long periods of time after coating and before use. It is not uncommon for the slit rolls to sit for multiple months in a distribution warehouse before being printed and applied. The product will need to be finely tuned to not break down during this storage time but still break down after the product life cycle is completed.

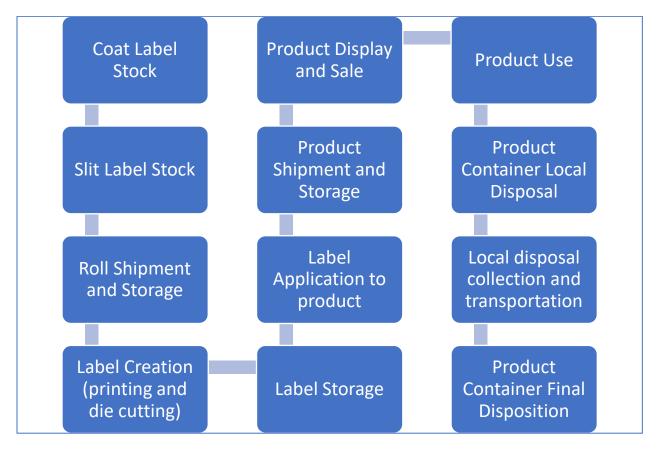


Figure 4: Label Life Cycle

ASTM method D6400 describes compostability testing - Standard Specification for Labeling of Plastics Designed to be Aerobically Composted in Municipal or Industrial Facilities.<sup>11</sup>

In order to compost satisfactorily the product must: disintegrate during composting, exhibit inherent biodegradation and have no adverse impacts on the ability of compost to support plant growth. Previous testing of a typical tackified aqueous acrylic in this test has shown that the material does not break apart even if applied to a compostable facestock like rice paper. Per the ASTM test method, less than 10% of the sample material can be greater than 2 mm in size after 12 weeks of composting. Table 1 below shows disintegration results. In practice, for a compostable adhesive to function, it is necessary

for a consumer to have access to composting through an industrial composting operation that accepts packaging which is not widely available in the United States today.<sup>12</sup>

	Average % wt. leftover – 2 mm screen after 12 weeks of composting
Plain Rice Paper Facestock	2.4%
Rice Paper Facestock coated with tackified acrylic	18.0 %
Semi-Gloss Facestock coated with tackified acrylic	24.3 %

Table 1: Disintegration Test Results

# WASH OFF ADHESIVES (Design for recyclability)

To enhance container reuse or substrate recycling, sometimes the label is removed via a washing step, utilizing high pH washing solution. This is especially prevalent in the recycling of polyester (PET) used in typical soda and water bottles. Many times, this wash solution is hot (50 - 85°C) and can be 1 to 2% caustic. The wash off efficiency can be greatly affected by the facestock. The labels need to be cleanly removed from the substrate (no sticky residue) and mechanically removed. This is generally achieved via float/sink separation and filtration. This seems to be the area of most focus in the development of PSAs for sustainable labels currently.

The composition of the polymer backbone, processing conditions, and type of additives used in the formulation are critical to obtaining a wash off adhesive solution. The composition includes the types of monomers, including special functional monomers, as well as the polymerization processing conditions. The formulation piece includes the type of emulsifier, thickeners, crosslinkers and neutralizers. Two example acrylic PSAs were tested for wash off ability where things like monomer, formulation or processing were modified. Table 2 below details initial work for the PET recycling solution. The American Plastics Recyclers (APR) have proposed regulations for this market.<sup>13</sup>

	Initial Screening Testing	Solution temp (°C)	Time tested (min)	# strips washed off*	Notes/Observations
Adhesive 1	~17gsm transfer to bOPP, closed to PET bottle	86	10	20	first wash off ~45s; PET bottle pieces curled during test - 100% wash off, adhesive stayed with the label
Adhesive 2	~17gsm transfer to bOPP, closed to PET bottle	85	10	3	3 washed off and adhesive remained on bottle pieces; PET bottle pieces curled during test – 15% wash off

\*Total number of strips used = 20

Table 2: APR Recycling Wash Off Testing Summary

Based on the results above a full development program was initiated with the purpose of developing an adhesive which passes the APR protocol for wash off labels from PET surfaces. The aim of the testing for certification is that the rPET (recycled PET) not be negatively impacted by the label – with a focus on haze, discoloration, visible contamination and clumping (which could hinder the processing). Figures 5, 6 and 7 below show the results of a 3<sup>rd</sup> party lab testing of a new adhesive designed for a wash off based on the above initial screening. The adhesive tested was applied to a standard bOPP film via a commercial coating process at ~ 17 gsm then applied to standard PET bottles covering approximately 40% of the surface of the bottles. The bottles with and without labels applied were then washed and ground and the resultant flakes and wash water were evaluated as per below.

	Control	Innovation	CG Guidelines	
Wash water evaluation	Clear	Slightly cloudy		
Elutriation loss	0.45%	0.42%		
Clumping Test With Load	0%	0%	<1 wt% retention on screen	✓
Clumping Test Without Load	0%	0%	<1 wt% retention on screen	✓

Figure 5 - 3rd party wash off evaluation testing

	Control	50/50 Control/Innovation	CG Guidelines	
Initial SIV (dL/g)	0.8	0.8		
Extruded SIV (dL/g)	0.73	0.73		
Extrusion SIV drop (dL/g)	0.07	0.07		
Δ IV Drop from Control (dL/g)		0	<0.025	✓
% Increase in Pressure		2%	<25%	$\checkmark$
Observation for fuming or odor at feed throat and die exit	None	None	No unusual fuming or odors observed	~
Observation for material sticking in drier or feed throat	None	None	No material sticks in drier of feed throat	~

Figure 6 - 3rd party wash off evaluation testing – continued

	Control	50/50 Control/In novation	∆ to Control	CG Guidelines	
L*	91.1	89.9		>82	<b>√</b>
a*	-0.77	-0.63	0.14	Δ<1.5	$\checkmark$
b*	4.74	5.45	0.71	Δ<1.5	$\checkmark$
Haze%	4.9	13	8.1	Control not exceed 9%, ∆<10%	$\checkmark$
Inclusions and specks	0	0		If control=0, Innovation<2	~
Pre-mold IV (dL/g)	0.73	0.73			
Post-Molding IV (dL/g)	0.7	0.7			
IV Drop (dL/g)	0.03	0.03	0	Δ<0.025	$\checkmark$

Figure 7: 3rd party wash off evaluation testing – continued

Another common wash off application is for wine labels where two circumstances lead to the need for the label to be washed off. If a wine isn't selling well, it may be relabeled to avoid waste. Also, some people collect wine labels, especially high end wines, so they want to remove the label intact. To enable both of these eventualities a wash off label adhesive is needed. A typical wine label adhesive needs high adhesion to the typical low energy glass coatings used on wine bottles and is therefore generally tackified. It also needs good adhesion at lower temperatures as wine bottles are often labeled outdoors or in unheated warehouses. A typical route to wash off behavior in this instance is a bit easier than the PET example above since careful hand removal and a very strong paper facestock is usually the norm. A formulation additive is the easiest approach to make the adhesive slightly hydrophilic without compromising the basic PSA properties. A quick screen study is detailed below using this formulation approach by post adding several different hydrophilic ingredients to a typical wine label adhesive. Details can be seen in table 3 below where subjective removal in warm water from 4 types of wine bottles was evaluated.

Additive Approach	Wash Off of Wine Bottles	Comments
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Control (no additive)	Poor	Good adhesion, coatability
Surfactant 1	Fair (3/4)	Very foamy
Surfactant 2	Fair (2/4)	Very foamy
Superabsorbent	Good (3/4)	Poor adhesion
Acid Containing Dispersion	Good (3/4)	Acceptable adhesion

Table 3: Subjective Wine Bottle Wash Off

### SUMMARY AND CONCLUSIONS

There are many ways to achieve a sustainable solution utilizing an aqueous acrylic PSA. Some of these solutions are closer to technical realization and definition than others. A summary table is presented below. There are not clear certifications in many of these applications.

Approach	Technical Approach	Complexity	Risks	Certification
Compostable Adhesives	Insert "weak" point to allow for break down in compost pile	High	"weak" point could jeopardize aging properties; requires access to composting	ASTM methods including D6400
Wash Off Adhesives	Insert appropriate hydrophilic functionality (lots	Moderate	Product line could proliferate; wash off could occur prematurely	Some specific to certain applications. APR

	to choose from here)			
Removable Adhesives	High MW and/or cross-linking – some existing products	Low	Would require consumer behavior changes; primers may be required	None required
Repulpable Adhesives	Insert "weak" point to allow for breakdown in pulper	High	"weak" point could jeopardize aging properties	USPS-P-1238E for "benign" PSAs
Biodegradable Adhesive	Insert "weak" point to allow for break down in process	High	"weak" point could jeopardize aging properties	EN13432 and ASTM D6400

The focus on sustainability for PSA labels is rightly focused on matrix, trim and liner recycling. Careful construction of the PSA label adhesive can enable sustainable solutions but there is still much work to be done, and some thought should be given to the complexity and potential risks. Initial work is promising.

### NEXT STEPS

Further work to develop a full line of adhesives designed for recyclability when paired with a specific substrate and application.

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