CREATING MULTIFUNCTIONAL COATINGS USING Recycled Raw Material Streams

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> *"One such option for these materials is their rebirth and incorporation into high performance protective coatings for wood and metal. Through some careful synthetic breakdown and reassembly, their life as a durable, functional, tough, attractive coating can be realized."*

Environmental, health, and safety concerns continue to drive rapid growth for environmentally friendly, low volatile organic compound (VOC) coatings. This growth, further compounded by increased social awareness of mega trends—namely depleting finite resources, the growing world population, and constrained food resources—has companies seeking highly sustainable feedstock solutions. Although biobased materials have provided feedstock options, which are more sustainable than fossil petroleum alternatives, use of recycled content has remained relatively unexplored. With the U.S. production of plastic bottle containers at an amazing 9.4 billion pounds in 2013, and the total plastic bottle recycling collection rate at only 30.9%, there is a wide gap in unclaimed, uncollected, and discarded plastic bottles.[1] The math is staggering when one considers that the remaining 69.1% amounts to 6.5 billion pounds. Where is the unclaimed material going? Landfills and incinerators take in much of the excess.

We have markets and supply chains for recycled poly(ethylene terephthalate), or PET bottles, to take another life as a bottle, but this option is operating inadequately, and competing with virgin material for demand. There is a real need for other options to become available for the used PET material, to essentially reincarnate; to live a second life as high performance polyester, which its pedigree supports. One such option for these materials is their rebirth and incorporation into high performance protective coatings for wood and metal. Through some careful synthetic breakdown and reassembly, their life as a durable, functional, tough, attractive coating can be realized. This new use for a previously harvested material will help to create increased demand and reduce the overall amount that finds its way into a landfill or incinerator.

PET, which has been processed into bottles, has a significant energy history and environmental footprint accounted for in its production, and therefore the complete environmental, health, and energy impact of its life cycle has already been paid to that point. It is within the material, waiting to be reharvested, reused, or converted into a new application, reducing the future need for petroleum-based feedstocks. When this material is discarded, all of the energy spent during the creation of these molecules is wasted and must be exerted again to create new virgin material. We discuss here the means to reclaim used

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PET as a raw material in high performance coatings, and the surprising results that accompany high incorporation of previously "spent" materials. It is noteworthy that we were able to extend the life cycle of valuable, finite resources, all environmental expenses previously paid, and employ them to create high value coatings solutions.

PET PROPERTIES

Processed or spent industrial materials might have alternate uses, especially those with inherent recycling advantages. Thermoplastic polymeric materials have this advantage; by simply remelting them, they can be converted into many diverse and useful products. The basic material changes very little, if any, during a cleaning and reprocessing phase. The constituent polymer molecules that provide the bulk properties of the plastic remain essentially unchanged, rendering them as useful as the virgin material. PET is such a material. As a thermoplastic polyester, it provides performance for many applications, from water bottles, to fibers, to packaging. Much of the commercially spent PET material continues to end up in landfills, regardless of future recycle value. We have tapped into this resource, increasing demand and diverting its destiny. Our focus was on using this source as feedstock as a basis for high performance coating resins, and at the same time avoiding use of petroleum-based ingredients whenever possible. As an added advantage in the interest of global sustainability, we anticipate building market share based on customers who enjoy high performance coating materials with high recycle/renewable content.

PET is now a global commodity material, having excellent performance in numerous applications. It has chemical resistance to acids, alcohols, grease, oil, aromatic hydrocarbons, and ketone solvents. According to published data,[2,3] PET has excellent impact strength (13–35 J/m), tensile strength (80 MPa unoriented, 190–260 MPa oriented), and modulus (2–4 GPa). The limiting oxygen index value for PET is 21%; it has low water uptake (0.1% 24 hr) and good ultraviolet (UV) resistance. Finally, PET films have exceptional elongation at break (60–165%) and outstanding barrier properties to CO_2, N_2, and O_2. Not surprisingly, it is an excellent choice for many packaging applications. Many of these qualities are also valued in various coating applications.

Since one of the main purposes for coatings is to protect the substrates to which they are applied, it is likely that PET properties would serve well in such an application. However, the standard industrial bottle and fiber grades of PET have much too high viscosity and molecular weight for average use in most coatings. Additionally, its solubility is poor in common solvents, and the melting temperature is very high (260°C) for practical use without an ultrahigh temperature bake cycle. Therefore, in order to convert recycled PET (rPET) into a more useful form for liquid coatings, further processing is required. Modification of properties with comonomers is required to redistribute the molecular weight around a new, lower average. The process incorporates select building blocks to balance the final bulk properties for coating films, and fine-tunes the equivalent weight, architecture, and functionality specific to coating applications.

In the standard process for manufacture of PET, after initial reaction of ethylene glycol with either terephthalic acid or dimethyl terephthalate to form bis(hydroxyethyl) terephthalate (BHET), a high vacuum at high temperature must be applied to remove ethylene glycol and drive the molecular weight upward. Molecular weight is correlated with the intrinsic viscosity measurement from solvent, and can exceed 35,000 Da for film and fiber grades,[4] although injection-molding grades are usually twice that. This process is energy and resource intense, and is very successful in producing the desired bulk properties. If a much lower molecular weight is desired, it would still require the removal of ethylene glycol to build the molecular weight from BHET. Alternatively, starting with the discarded high molecular weight material, a much shorter and more direct path to coating materials is available by a proprietary digestion/functionalization process. This is crucial in producing practical polyols with high recycle content, while imparting desirable properties for the coatings industry.

LIFE CYCLE ASSESSMENT

The advantage of utilizing rPET is demonstrated by findings of the Life Cycle Assessment (LCA) review commissioned by Resinate Materials Group in 2014.[5] LCAs for virgin PET, biobased PET, and rPET were compared for cumulative energy demand, as shown in *Figure* 1. As previously

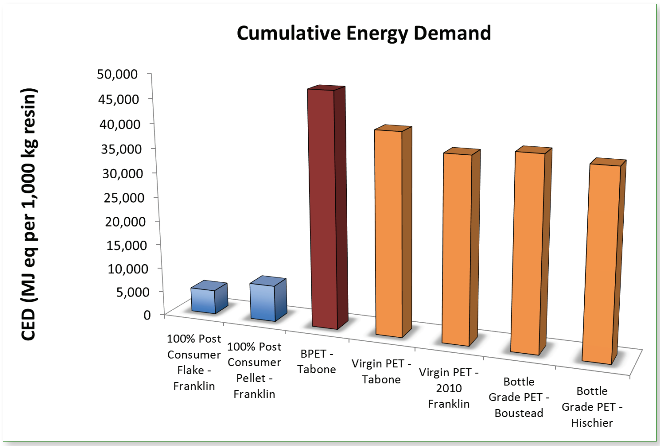


Figure 1—Comparison of the cumulative energy demand of PET types.

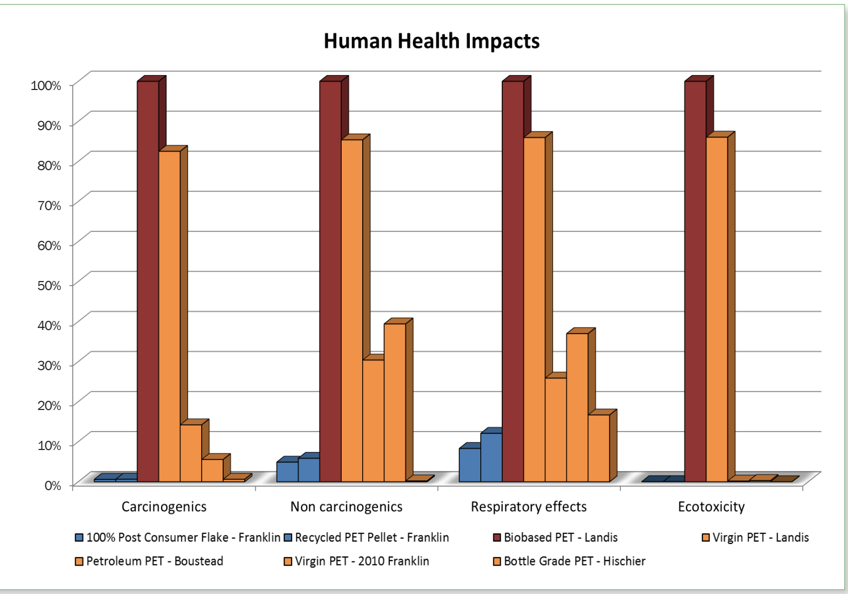


Figure 2—Comparison of health impact for PET.

discussed, because rPET does not require the energy necessary to produce the raw materials and then the polymer itself, it has a distinct advantage over both virgin and biobased PET. The advantages were calculated from the complete synthetic pathway. This includes contributions from the initial conversion of ethylene glycol and terephthalic acid to virgin PET, fabrication into bottles, and finally to various pellet or flake recycle streams. The starting materials are derived from either petroleum or biobased feedstocks. The calculated benefits carry through all the way to the final coating materials described in this work.

The same trend was observed when looking at human health impacts detailed in *Figure* 2. Life Cycle Impact (LCI) was modeled in SimaPro v7.3 and analyzed using the Tools for the Reduction and Assessment of Chemical and other environmental Impacts (TRACI). TRACI 2.1 was developed by the U.S. EPA to assist in impact analysis for LCAs. Human health carcinogen and noncarcinogen data are in Comparative Toxic Units (CTUh) for more than 200 chemicals, and represent the estimated increase in morbidity in the total human population per kg of chemical emitted. Respiratory effects are measured in kg particulates that are 2.5 microns or less in diameter (PM2.5 eq) and represent a different metric. Finally, the ecotoxicity was measured using a third metric, Comparative Toxic Units (CTUe), an estimate of the potentially affected fraction of species integrated over time and volume per unit mass of chemical emitted. The health impact scale was therefore normalized to 100% by taking the data from four separate categories, and adjusting their individual scale as a percentage of the highest overall calculated effect. This allows for the visualization of all the subcategories simultaneously within the same scale. More recent advancements in PET production technology may change the advantage differences between petroleum and bio-derived materials; however, even by these metrics, one can never match the environmental advantages from starting with recycled materials.

DESIGNING POLYOLS FROM rPET

Starting with the rPET stream, there are several parameters to consider in converting the bulk material into a useful form for coatings. The molecular weight issue was previously discussed. Furthermore, the functionality and equivalent weight required for most coatings polyols is not inherent in the rPET as supplied, so hydroxyl end groups need to be generated. This provides a handle for both secondary processing (PUDs) and for final curing for thermoset coatings (melamine, isocyanurate, etc.). Since PET is inherently semicrystalline, it is also necessary to determine whether to preserve or eliminate this property.

According to Schiraldi et al.,[6] "modifying substances" can be used to affect the properties and degree of crystallization, tune tensile and modulus properties, adjust the T_g and T_m, and modify barrier properties. Carefully selected comonomers can accomplish this while simultaneously contributing to other performance attributes. From the aromatic side, isophthalic acid (PIA) has become the most widely accepted modifier for packaging applications due to its relatively minor effect on the T_g, reduction in the crystallization rate but not in the ultimate level of crystallinity (at <5 mol%), and improved barrier properties.[6] Additionally, hydroquinone and 4,4'-bisphenol are known to accelerate crystallization rates over neat PET.

The introduction of long chain diols can impart desirable characteristics such as flexibility. Polyols such as hexanediol, butanediol, and dodecanediol are good examples. Polyethers such as polyethylene glycol (PEG) or poly(tetramethylene ether) glycol (PTMEG) are also good diol modifiers for flexibility. This increase in flexibility may come along with substantial changes in T_m, T_g, and crystallinity. As is true with many properties, opposing ends of the property spectrum must be balanced to maintain good overall performance in coating applications.

Additionally, in starting from mixed recycle streams of PET, there may be some unwanted color associated with prior use in packaging, and this may need to be eliminated for some coating applications. Clearcoat layers designed as final topcoats for wood and metal substrates are normally colorless, and decolorizing rPET streams has become necessary for consistency. A novel process has been established for reducing or eliminating color associated with recycle-grade bottle flakes, but will not be discussed here.

A final note on the design of polyols relates to natural and biobased modifications. These ingredients are also of high interest, and may include many different acids or anhydrides, such as adipic and succinic, and diols such as propanediol, ethylene glycol, and others. Multifunctional intermediates such as pentaerythritol (Voxtar™)[7] are now being made through a renewable and sustainable biobased process, and can provide needed hydroxyl functionality for coating applications. Of course, most fatty acids are naturally derived and can provide some level of hydrophobicity in the polyol when needed. Polyether polyols have found some level of "green" as well, with alkoxylated hydroxyl-functional natural oils[8] and epoxidized methyl oleate polyether polyols.[9] Our corporate philosophy with respect to green chemistry is to use recycle content raw materials first and biorenewable content second. If the performance requirements set by our customers cannot be met with these first two options, only then do we use petroleum content raw materials or ingredients. This approach leads to the highest "green" content possible in the final polyol.

EXPERIMENTAL

Waterborne Coatings from rPET Polyols

We have successfully made high-PET-content PUD compositions. A variety of polyols can be used alone or in blends to achieve the desired balance of properties, as well as the target particle size and long-term stability of the dispersion. The dispersions were made using a modified acetone process, without additional co-solvent, where the final dispersion has been vacuum-stripped to remove residual acetone.

Table 1—Ambient PUD Liquid Properties

Property	Typical Value	Test Method
Appearance	Milky white to hazy, slightly green hue	Visual
Nonvolatile Content	35–40%	ASTM D2369
pH	~ 9	
Viscosity (25°C)	150-850 cP	Brookfield DV-III
T_g (dry film)	50–55°C	ASTM D7426
Density	8.8–8.9 lb/gal	ASTM D1475
VOC	< 30 g/L	Calculated

Table 2—Ambient PUD General Film Properties

Property	Typical Value	Test Method
Pencil hardness	HB–2H	ASTM D3363
Mandrel bend	< 1/8 in	ASTM D522
Film clarity	Clear	Visual
Time to tack free	< 30 min	ASTM D1640
Gloss 60°	89 GU	ASTM D523
MEK resistance	~ 100 MEK double rubs	ASTM D5402
Adhesion to maple and aluminum (crosshatch)	5B	ASTM D3359

One early prototype was designed for ambient, or low temperature oven-dried applications. This PUD utilizes a novel proprietary polyol that is made up of 99% combined recycled and renewable raw materials. As stated previously, the dispersion was finished co-solvent free, and still had excellent film-forming properties at room temperature. The physical properties and performance of the PUD are a direct outcome of the polyol design, beginning with rPET and selected modifying substances. The liquid properties of the resulting PUD can be found in *Table* 1.

To test the performance of the resulting film, the unformulated dispersion was drawn down over aluminum substrate after isopropanol wipe using a #50 wire-wound rod and then kept under ambient conditions for seven days. The films were evaluated for typical coating properties including hardness, flexibility, clarity, etc. The performance results and associated test methods are shown in *Table* 2. Gloss and adhesion to maple were done over maple lumber as described in the next section.

We based total green content of our polyols on the combined weight percentage of recycled and renewable content, and for the polyol used in this dispersion it was >99%. The resulting overall green content of the final PUD was at 50% on a solids basis. This dispersion exhibited a great balance of flexibility and hardness, low VOC and co-solvent free formulation capability, high gloss

Table 3—UV-Curable Liquid Properties

Property	Typical Value	Test Method
Appearance	Milky white	Visual
Nonvolatile content	~ 40 wt%	Calculated
pH	7–8	
Viscosity (25°C)	6–10 cP	Brookfield DV-III
Density	8.8 lb/gal	ASTM D1475
VOC	< 100 g/L	Calculated

Table 4—UV-Curable Film Properties

Property	Typical Value	Test Method
Film clarity	Clear	Visual
Pencil hardness	3H–4H	ASTM D3363
Time to tack free	<30 min	ASTM D1640
Gloss 60°	90 GU	ASTM D523
MEK resistance	>200 Double rubs	ASTM D5402
Adhesion to maple	5B	ASTM D3359
Taber loading 500 g	16–19 mg	ASTM D4060
Taber loading 1000 g	39–48 mg	ASTM D4060
Block testing	No damage	ASTM D2793
Print testing	No damage	ASTM D2064

Table 5—Energy Dosage Used for UV Cure

Property	Typical Value	Test Method
UV-A	313.4	1556.2
UV-B	268.3	1341.4
UV-C	57.2	294.2
UV-V	404.6	1929.3

and excellent clarity, while bringing a distinct sustainability advantage. The dispersion has demonstrated shelf-life stability at 50°C for more than eight months. Average particle size was around 150 nm. While this material was designed for wood coating applications in particular, it will likely find utility in other markets and substrates as well.

A similar synthetic process was used to make a UV-curable PUD, which provided thermoset properties through UV-radiation exposure. This dispersion also incorporated a specially designed polyol comprised of 99% recycled and renewable raw materials. The UV-curable nature of the polymer eliminated the need for acetone in the synthesis of this dispersion. This distinction reduced the amount of waste associated with production and eliminated the energy necessary to strip solvent. The liquid properties of this UV-curable PUD can be found in *Table* 3. Again, oven-aging stability at 50°C has been superior at 90 days so far without separation or settling.

Table 6—Household Chemical Stain Results

Stain	Rating	Stain	Rating
Vinegar	5	100 Proof vodka	5
Lemon juice	5	Windex	5
Orange juice	5	Water	5
Grape juice	5	Hot coffee	5
Ketchup	5	Artificial perspiration	5
Olive oil	5	1% Detergent solution	5

Evaluation of PUDs over wood was performed on select maple lumber acquired from a local lumber yard. The surface was prepared by sanding with 220 grit sandpaper and then thoroughly cleaned using tack cloth. After adding approximately 2 wt% of Irgacure 1173, the UV-curable dispersion was drawn down at a thickness of 5 mils (wet). The sample was allowed to dry at ambient temperature until tack- free, then placed in an oven at 50 °C for 5 min. At this point, the film had a hardness of 42 Konig seconds and demonstrated low solvent resistance of <10 methyl ethyl ketone (MEK) double rubs. This open time allows the applicator to inspect coated materials prior to UV curing and potentially fix any defects by means of solvent or abrasion. The film was then cured using a Heraeus Noblelight Fusion UV LC6B benchtop conveyer with a 558432 H+ bulb with a belt speed of 25 ft/min. A second coat was applied by repeating the same steps as the first. The final cured film was tested for general properties. The results and associated methods are shown in *Table* 4.

Measurements were made for energy dosage by using an EIT Power Puck II Radiometer. The data are shown in *Table* 5.

By leveraging unique polyol technology based on rPET, this dispersion contained 16% overall recycled/renewable content on a solids basis. The result was an environmentally friendly material based on high strength ingredients, which produced an extremely hard coating with high resistance to many physical and chemical assaults. The UV-curable component of this polymer helped provide excellent block and print resistance, high gloss and exceptional film clarity, as well as excellent chemical and stain resistance. In 24-hr stain resistance testing, this UV-curable PUD formulation was exposed to several household liquids. The coating was assessed on a scale of one to five after the stains had been in contact with the coating for 24 hr. A rating of five indicates no change in the coating, while a rating of one indicates complete staining or coating failure. This novel UV-curable PUD composition received a grade of five (excellent) for all 24-hr stain testing, making it a strong candidate for wood flooring, kitchen cabinet, and wood furniture applications (*Table* 6).

Like its ambient counterpart, this UV-curable PU dispersion was also low VOC (calc'd. 18.1 g/l), provided co-solvent free formulation capability, and brings the same sustainability advantage when compared to other UV-curable PUDs on the market. This PUD can be used for protecting furniture, kitchen cabinets, prefinished/OEM wood flooring, as well as onsite wood refinish markets.

Solventborne 1K Melamine and 2K Urethane Baked Thermosets

We also have prepared novel polyester polyols that are delivered in various solvents for industrial coating applications. *Figure* 3 displays a variety of physical properties for the various polyols, and the resulting film performance characteristics produced from them. Depending on the polyol composition, the crosslink system used, the functionality, and the equivalent weight, a wide spectrum of combined properties are available. Incorporation of high levels of rPET was possible, and provided a variety of excellent performance qualities. Mixed recycle streams and renewable streams were used to tailor the properties while maximizing the overall green content. Green content was a combination of the recycle and renewable raw material content for the polyol portion. The materials listed here were of varying composition, viscosity, molecular weight, and architecture. These polyols all generally demonstrated good hardness, toughness, flexibility, and chemical resistance, while incorporating a high level of recycle feedstock. Not shown in the graph are the crosshatch adhesion results over aluminum substrate (4B–5B), the mandrel bend results (all passed 1/8 in.), and pencil hardness (F–4H). Development of superior performance compositions with high recycle and renewable content is under way. Additionally, incorporation of blended recycle streams is under investigation for synergies in a variety of coating applications.

Solventborne 2K Urethanes from Various Commercial 1000 MW Polyols

To gauge the relative value of rPET derived polyols for coatings, a representative sampling of various commercial polyols were obtained and formulated into two-component clear coatings using hexamethylene diisocyanate (HDI) trimer (isocyanurate). The polyols were all commercially available as 1000 mol wt, linear, diol functional materials. We blended with HDI trimer at an index of 1.05:1.00 isocyanate:hydroxyl and diluted in a 2:1 blend of MEK and propylene glycol monomethyl ether acetate (PM Acetate) solvents. The solventborne 2K coatings were drawn with a wire wound bar to a resulting dry film thickness of 1.0–1.2 mils over 4 x 6 aluminum panels, first wiped clean with isopropanol. After a 60-minute ambient flash, they were baked briefly at 130°C and then tested according to ASTM guidelines. We compared the cured systems with each other across chemistry groups, and within chemistry groups.

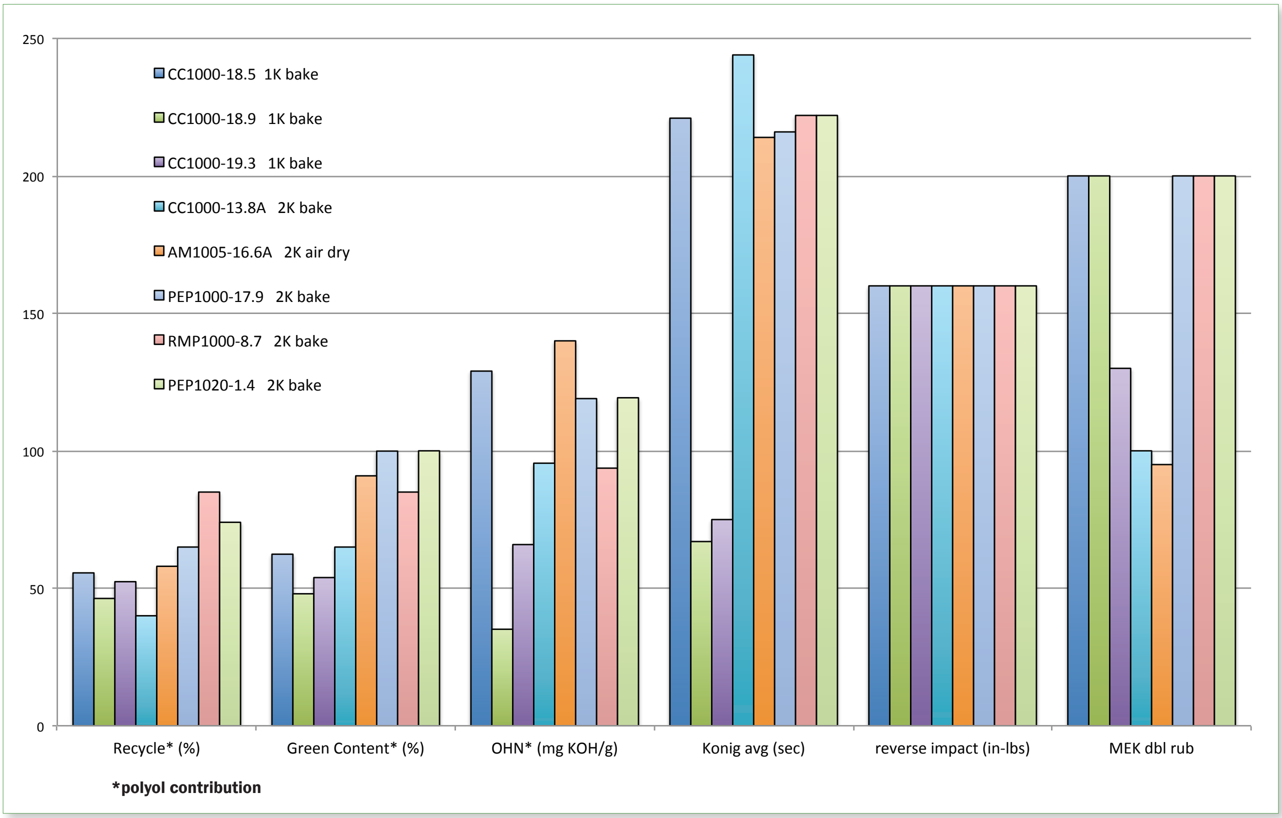
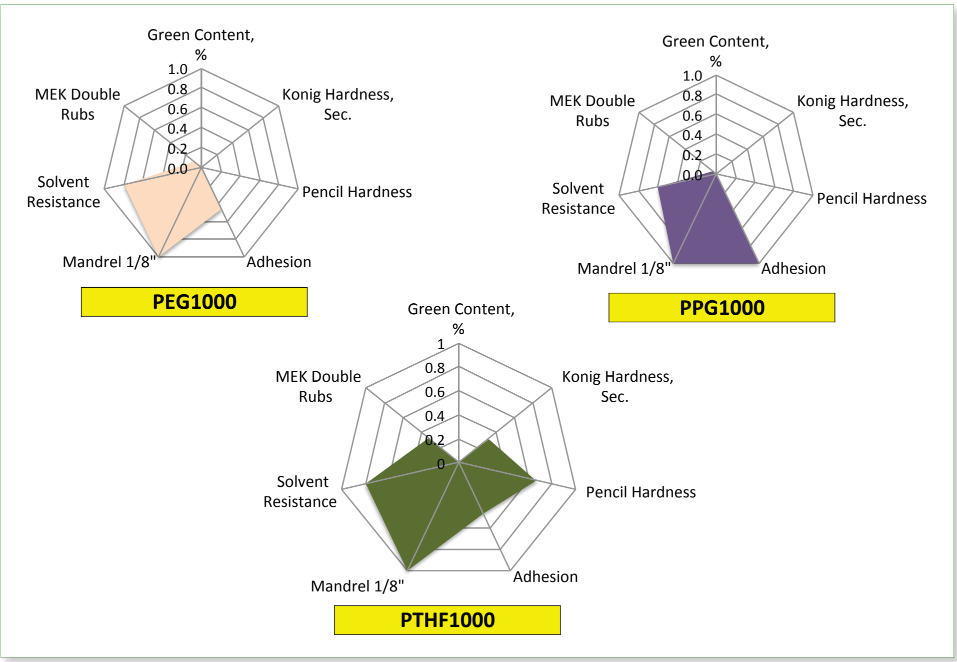


Figure 3—Some typical coating properties for thermosets.

Figure 4—Polyether polyol 2K properties.

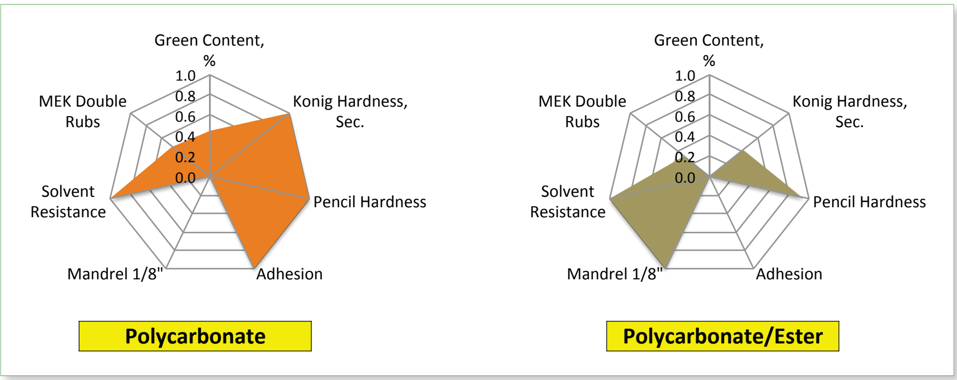


PEG1000

PPG1000

PTHF1000

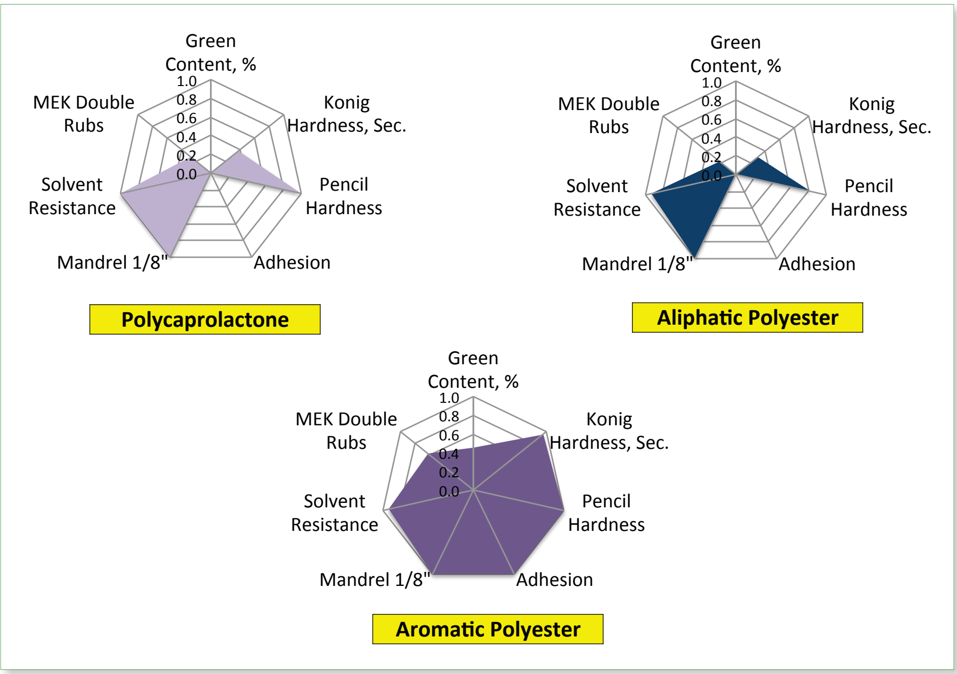
Figure 5—Polycarbonate polyol 2K properties.



Polycarbonate

Polycarbonate/Ester

Figure 6—Polyester polyol 2K properties.



Polycaprolactone

Aliphatic Polyester

Aromatic Polyester

The best observed performance from all the different coatings for each individual test was normalized to 100% on the radar plots (*Figures* 4–7), so that the best overall performing materials ultimately have the most shading. All test results were scaled relative to the top performer and are shown in the plots grouped by backbone structure (i.e., polyether, polycarbonate, polyester). The first three groups (*Figures* 4–6) were commercial polyols from other sources; the last group (*Figure* 7) was experimental polyols developed in our labs. There were not many surprises; the observed properties were due to a blend of effects from both the HDI trimer and the polyol. However, the equivalent weights were the same and therefore the relative amount of trimer was essentially the same. One significant note about this test is related to the size of the repeat unit (unknown in some cases), which varied with the chemistry type. With a 500 Dalton equivalent weight used for this evaluation, a varying number of repeats can be assumed and therefore we are testing this uncontrolled variable as well, as defined by the polymer itself. Tighter constraints on this variable would be a natural expectation for comparisons within each chemistry group, but not across groups.

Polyether Polyols

The first group of polyols tested were polyethers, consisting of poly(ethylene glcol), PEG; poly(propylene glycol), PPG; and poly(tetramethylene glycol), PTHF. As mentioned, all were sold as 1000 mol wt diols, and used as received. *Figure* 4 shows the relative performance in the areas of focus: Konig hardness, pencil hardness, crosshatch adhesion, mandrel bend 1/8 in., combined solvent exposure testing for water (24 hr), MEK (1 hr) and isopropanol (1 hr) by covered spot, MEK double rubs, and the calculated "green content" based on the total of percent recycle content and the percent renewable content.

The polyethers were, as a general class, a bit soft by Konig and pencil hardness standards, and the MEK double rub performance was very poor. However, they did have excellent flexibility and a high degree of solvent resistance, and showed moderate adhesion to the aluminum substrate. The materials we tested did not have any green content, from either a recycle or a renewable source.

Polycarbonate Polyols

The second group of polyols that was studied in this evaluation were the polycarbonates. The distribution of properties for each of these is shown in *Figure* 5. Conducting the same battery of tests, the radar plots displayed slightly better coverage of properties with this group than the polyethers. The MEK double rub resistance for this group showed signs more typical for a crosslinked network. The solvent resistance was significantly improved, and very good pencil hardness (H) was common to both polyols in this group. Surprisingly, significant Konig hardness was only present for the polycarbonate and much lower for the copolycarbonate-ester, indicating that the two types of hardness measurements were actually measuring different sets of properties. Having good pencil hardness does not necessarily predict good pendulum hardness. The pendulum was measuring the damping effect

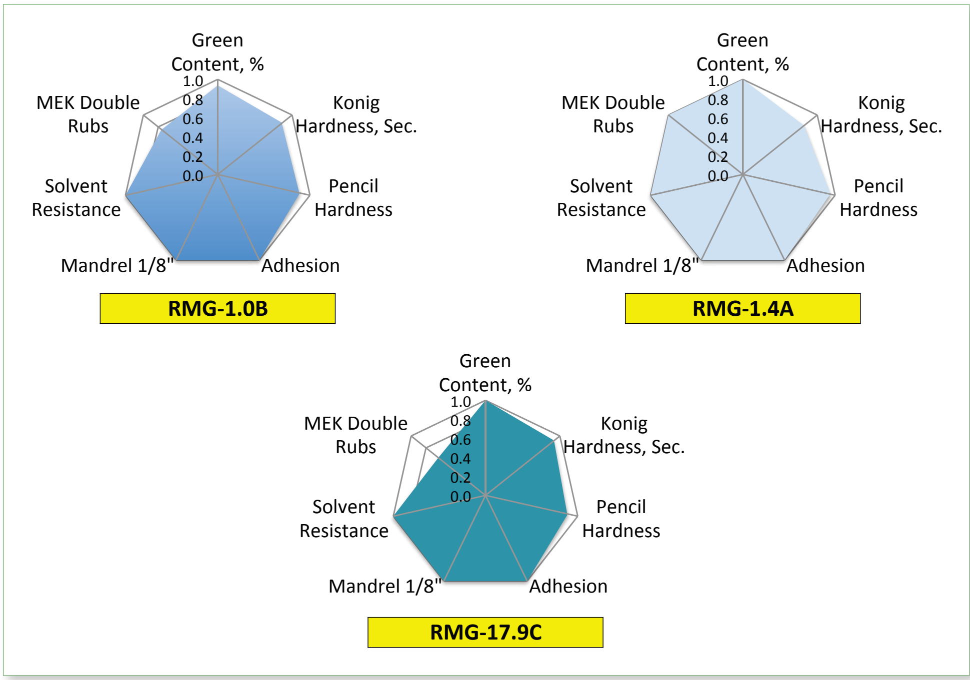


Figure 7—High recycle content polyols.

Table 7—Viscosity and Potlife for 2K Polyol Study

Polyol	T_0 Viscosity, cPs	T2x Potlife, min	Starting Torque (%)	Final Torque (%)	Thermosel Temperature
PEG 1000	28.6	10	20.3	42	25°C
PPG 1000	18.6	54	15.6	31.6	25°C
PTHF 1000[a]	28	10	24	47.5	40°C
Polycarbonate	66.5	31	50	99.5	25°C
Polycarbonate/ester[a]	89	13	47	78	40°C
Polycaprolactone	60.1	10	50	85.1	25°C
Aliphatic polyester[a]	400	11	47	96	40°C
Aromatic polyester	119.1	46	50.1	94.5	25°C
RMG-1.1C[b]	1635	7	50	91.1	25°C
RMG-1.4A	166	24	50	90.4	25°C
RMG-17.9C	354	25	48	93	25°C

(a) These materials could not be processed at 70% solids in the selected solvents at 25°C without crystallization occurring and had to be evaluated at higher temperature.

(b) RMG-1.1C was evaluated in this test as a direct replacement for 1.0B.

related to the non-elastic response of the paint film, while the pencil scratch hardness is positively correlated with indentation hardness, elasticity modulus, film thickness (constant here), and fracture toughness of the film.[10] Good flexibility by mandrel bend was only observed for the copolycarbonate–ester material, not for the straight polycarbonate. It was also interesting that the polycarbonate–ester polyol demonstrated very low adhesion to aluminum substrate. The polycarbonate material tested was given credit for green content, based on the manufacturer's information available at the time.

Aromatic and Aliphatic Polyester Polyols

The third group of polyols studied was the aromatic and aliphatic polyester polyols. This group was a combination of aliphatic and aromatic polyols, and the differences are apparent (*Figure* 6). The two aliphatic polyester materials demonstrated performance very similar to the copolycarbonate–ester material in the previous group. In fact, the properties measured in this study indicate almost identical profiles for the polycaprolactone, aliphatic polyester, and polycarbonate–ester. What was striking in this group was the effect from building aromatic character into the backbone of the polyol, where we saw very good property development overall. For the aromatic polyester, there was a significant improvement in all categories. While bringing adhesion, pencil hardness, and flexibility, this material was able to nearly fill out the chart with excellent Konig hardness, good solvent resistance, and moderate MEK double rub resistance. For the aromatic polyester, some green content was noted by the manufacturer, and overall composition was around 60%. This was the first group to deliver strong properties in most of the categories.

High Recycle and Renewable Content-Based Polyols

One of the main goals of this research was to investigate the impact on coating properties from the incorporation of high levels of recycled materials of various types. While this work mainly focused on the use of rPET, there were other significant streams from both post-consumer and post-industrial waste that were leveraged for a desired effect on coatings. Full optimization of mixed chemistries is still being investigated. Here, we group the high-rPET materials, which are expected to perform in a similar way to the commercial aromatic polyester previously discussed. The results are shown in *Figure* 7, and it is evident from the overall shading that they performed as anticipated. In fact, while maintaining the performance seen previously for aromatic polyester, these materials have additional improvement on the MEK double rub resistance while maximizing the total green content. Slight differences in the overall balance of properties for these materials are attributed to their individual compositions, and this information can be used in the future to tailor properties for specific coating functions. An additional comment worth mentioning is the inherent need for photostabilizers with aromatic polyesters intended for exterior exposure. The aromatic monomers are known for increased susceptibility to photooxidation, leading to yellowing of the film over time.[11]

VISCOSITY AND POTLIFE

For each of the above tested polyols, solutions were made in a 2:1 blend of MEK and PM Acetate, as described previously. With such a wide variety of polyol chemistry to evaluate, this presented some difficulties in trying to find a universal system. To enable a side-by-side comparison under identical conditions, we chose to hold the solids level constant. A useful viscosity range for this study was found with solutions at 70% solids. At constant solids, the polyol initial viscosities varied substantially (spanning two orders of magnitude) depending on the basic polyol chemistry. Future work will focus on holding initial viscosity constant. A substantial level of dibutyltin dilaurate was used as catalyst, at 0.05% on solids. A Brookfield Rheometer Model LVDV-III U (Spindle 31) with a Brookfield Thermosel Model HT-60 was used for this work, along with a Brookfield Temperature Controller (Model Brookfield 106).

The data for the initial viscosity and the corresponding potlife measurement based on doubling of viscosity are shown in *Table* 7. It is interesting to note that the semicrystalline polyols could not be evaluated at ambient temperature due to their tendency to crystallize in this solvent mixture. The three materials which demonstrated this property had to be evaluated at 40°C rather than the standard 25°C test conditions. The polyether group generally had very low solution viscosity in this study. A significant difference was observed in potlife between the PEG and the PPG sample. The aliphatic polyester, the PTHF, and the polycarbonate/ester had similar potlife activity, however, they were all evaluated at elevated temperature, accelerating the reactivity and shortening potlife in a similar manner. The aromatic polyester and the PPG polyol had the longest potlife under the conditions of this study at 25°C. RMG polyols covered a range of viscosities, as their compositions are very different. Sample 1.1C had high viscosity, but also had higher functionality and was a bit more reactive as noted by its very low potlife. The other RMG polyols had very useful viscosity properties at this solids level under ambient conditions.

SUMMARY AND CONCLUSIONS

With the motivation to take advantage of any and all performance properties of PET, we have developed new polyols from recycled PET that have demonstrated usefulness for various coating applications. The polyols evaluated in this work clearly show desirable performance in the most popular wood and metal coating test categories. Hardness, flexibility, toughness, strength, and chemical resistance are all benefits our company has taken from materials that have already been spent for their designed purpose. The polyester material is still useful but has to be slightly re-engineered for a new life as a coating. The data presented here are consistent with, and reinforce, the valuable contribution that spent recycle streams can bring to high performance areas in the coatings arena. We are acquiring the performance data and design feedback from its process and composition variables to meet and exceed the needs in the coating resin sector, with a high metric for sustainability. CT

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