BIOBASED COATINGS

Collaboration at the Intersection of Innovation, Ecology, and Economics

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In the past several years, many in-depth articles have been written across the industry heralding a wide variety of advances suggesting that biobased materials were on the verge of achieving explosive growth.

Some of these coatings manufactured from "natural" or ecofriendly sources are commercial and manufacturing success stories. Yet, many other developments and advancements are still waiting for the right resin, application, or manufacturing opportunity to manifest itself.

In 2019, an in-depth look at the role that academia plays in successfully making coatings "benign by design" hinted that success in mass utilization of biobased coatings must include a several pronged industry-wide approach.[1]

Following up on those observations, this article further examines today's biobased coatings world from the perspective of sustainability, standardization and industry/government/university collaboration as a basis for progress moving forward.

THE COATINGS INDUSTRY SUSTAINABILITY CHALLENGE

Since the turn of the century, every industry has made efforts to some extent to build sustainability into their product portfolios with varying degrees of success. Recycling was one of the initial movements to gain traction.

Simply put, if things could be reused or reinvented as a new use, less would end up in the burgeoning landfills—sort of like hand-me-downs of previous generations, but on a much wider scale.

Over the past 20 years, sustainability has evolved into a much more complex paradigm with four main pillars encompassing human, social, economic, and environmental interactions.

Together, these four pillars intertwine to personify sustainability defined by the *Oxford English Dictionary* as "*the avoidance of the depletion of natural resources in order to maintain an ecological balance*." Merriam-Webster further describes sustainability as "*a method of harvesting or using a resource so that the resource is not depleted or permanently damaged.*"

The coatings industry first approached sustainability through reductions in manufacturing emissions and commercially successful product developments such as water-based technologies that lowered VOCs.

Lately, the industry has continued sustainable market-wide initiatives by developing a portfolio of biobased coatings. These products attempt to substitute raw materials derived from fossil fuels with products made from plant-based biomass raw materials such as vegetable oils or sugars.

Key components of these initiatives are rooted in the use of renewable raw materials for producing biobased solvents, resins, additives, pigments, colorants, and crosslinkers. Lately, these bio-raw materials have tapped into the Renewable Carbon Initiative. Carbon dioxide that is released from a host of industrial and commercial processes is captured and used as feedstock for producing polymeric building blocks.


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Some of these coatings have been technical successes but are presented and even marketed as niche products. Even today, 100% biobased coatings command only 1-3% of the global market volume. Market share rises to a generously estimated 10% if formulations that contain *any* amount of biobased raw ingredients are included, no matter how minuscule.

In order for biobased coatings to leap into a more mainstream view and command a larger market share, a number of hurdles still need to be overcome. The availability and price of biomaterials, as well as high technical requirements have had a limiting effect despite regulatory efforts to force the issue.

The fact is that the market for these coatings is still rather small, and production is more complex and more expensive than conventional products. Availability of renewable biomass materials would seem to be an easy challenge to overcome, but the fact remains that even renewable harvests take time to "grow" and can be thwarted by something as mutable as bad weather.

The industry is working on solutions that would be derived from biowaste, recycled content, and cellulose from carbon capture. But these solutions will take time to become practical realities.

The biobased coating market is still in its infancy primarily because the industry (which consists of individual companies trying to maximize market share and profitability) either cannot or is unwilling to invest heavily in biomass development for raw materials and in new manufacturing processes for those materials.

Research staffs across the industry have been slashed to the bare bones over the years. This leaves little time for the innovative, time-consuming, thoughtful development needed to ensure that biobased coatings and raw materials will ultimately achieve the same level of field performance and across-the-board acceptance as traditional products.

In short, the main challenges to large-scale acceptance of bio-coatings are consistent raw-material availability, affordable cost of manufacturing, comparable (or better) performance characteristics, and the time for scientists to innovate in this new dimension.

However, there are forces challenging the status quo within the coatings industry from other directions that have sparked a push to find new ways to address these dilemmas. Consumer awareness around "green" technologies is increasing, and customers are seemingly willing to reward companies that address those concerns by favoring their products.

Health problems emanating from well-publicized scandals such as excess formaldehyde in imported manufactured flooring and improperly constituted drywall that caused unexpected and enormous VOCs in the residential building industry further exacerbated the public's growing demand for cleaner products.

Regulations are becoming more stringent around the entire paradigm of sustainability, driven in part by consumer demand and government initiatives. However, not all these challenges have resulted in punitive demoralizing actions or results.

Rather, paint companies have begun to approach the challenge differently by taking important smaller steps to develop crucial new molecules that act as "drop-ins" for segments of the paint recipe. This is especially important for balancing performance criteria and pricing competition with well-established petrochemicals.

Using drop-in building blocks provides a faster way to integrate biomass feedstock without the need to make large investments or drastically change high-capital investment production processes.

In addition, many of these monomers require UV light, oxygen, and renewable raw materials, providing more sustainable manufacturing and broader flexibility into a wide variety of markets. Examples of some of the more prominent biobased building blocks for resin synthesis are listed in *Table 1*.

Polyesters are formed from the reaction product of polyols and a di- or multifunctional acid or carboxylic acid and anhydride for ester linkages in the polymer chain.

Solventborne alkyds have been used in coatings for decades. However, stricter pollution regulations have nudged the development of waterborne alkyds or polyurethane dispersions (PUDs).

PUDs offer improved performance over alkyds due to the robust urethane linkages. They are linear or lightly branched and of relatively high molecular weights dispersed in water. Lower film-forming temperatures at higher glass transition temperatures can be achieved because urethanes bond strongly to water.

Latex particles swell, causing a plasticizing effect. This allows PUDs to have lower VOCs and lower film-formation temperatures together with improved mechanical, chemical-, and corrosion-resistance properties than waterborne alkyds or conventional latexes.

Some of the more recent biobased resin technologies are low-odor, low or no VOCs, and free from phenolethoxylates (APE). Performance characteristics seem to approach solventborne counterparts, although long-term durability testing is still ongoing. In any event, this is a positive step in manufacturing biobased coatings that does not compromise performance standards.

TABLE 1—Examples of Biobased Building Blocks for Resin Synthesis[2]

NATURAL OIL	SOURCE	ACID
Castor	Castor oil seeds	Bosuccinic acid
Coconut	Coconut kernel	Biooctadecanoic acid
Linseed	Linseed seeds	Bio 2,5-furandicarboxylic acid
Safflower	Sunflower seeds	Bioazelaic acids
Sunflower	Sunflower seeds	Bio 3-hydroxypropionic acid
Tung	Tung tree	Bioacrylic acid
Tall	Wood pulp	

INDUSTRY SOLUTIONS: STANDARDIZED METRICS FOR BIOBASED MATERIALS

One thing that has stymied the industry is the definition itself of biobased coatings. To some, it meant using any amount of non-traditional material not derived from fossil fuel. Others defined bio-coatings (or biomaterials) as having greater than 90% biomass.

It was not practical to dictate an across-the-board percentage for all products because performance and cost were also necessary criteria—but manufacturers did want credit for their endeavors. Some even thought that if their processes emitted less pollution, then that would somehow qualify in the biomass calculation.

The USDA BioPreferred® program, which was created in 2002, was one of the first to try to standardize and quantify exactly what amount of incorporated material defined a biomaterial or biobased coating. More importantly, the percentage of biomaterial in the coating would vary depending upon the resin system to ensure that performance standards remained intact.

The original goal of this federal program was to encourage the use of agricultural products in a wide variety of industries. The idea was to boost farm income, economic development, and job growth.

But simply relying on a "nice to do" policy was not sufficient. Teeth were added to the program by requiring federal agencies to give purchasing preference to biobased products. Further, companies that met the criteria could have their products USDA Certified Biobased and listed on the database as choices for these purchases.

The 2014 farm bill expanded the original mandate to include forestry products. Now, the program defines biobased products as those derived from plants and other renewable agricultural, marine, and forestry materials, excluding food, feed, biofuels, livestock, and pharmaceuticals.

Starting with just six product categories, the program now includes 139 federal purchasing categories. As of 2021, 17,000 products have been tested and approved. Categories important to the coatings industry include adhesive removers, paint removers, wood and concrete stains, exterior and interior paints and coatings, binders, enamel coatings, wood and concrete membrane and penetrating sealers, epoxy systems, inks, printer toner, chemical, oil, fat, and wax intermediates, roof coatings, water tank coatings, powder coatings, and lubricant components.[3] *Table 2* lists the BioPreferred® categories specifically affiliated with the coatings industry.

More importantly, each category now has a defined percentage of the minimum renewable carbon content (MRCC) required for certification and inclusion. The biobased content is calculated using the ratio of "new" organic carbon (plant or agricultural base) to the total organic carbon ("new" organic carbon + "old" or fossil-fuel-based organic carbon).[4]

ASTM D 6866-21 *Standard Test Methods for Determining the Biobased Content of Solid, Liquid, and Gaseous Samples Using Radiocarbon Analysis* is the standard test method developed by ASTM International to determine the biobased carbon/biogenic carbon content. This standard is the basis for independently certifying and classifying each product. The equivalent standard used in Europe to determine the carbon content of products is ISO-16620-2:2019 *Plastics-Biobased Content–Part 2: Determination of Biobased Carbon Content.*

One measure of success is whether or not there is a positive economic impact on these biobased material initiatives. The latest report published in 2017 indicated that out of $450 billion in federal purchases, $450 million went toward biobased products. While this represents only 0.1% of the total, the bioproducts industry generated $400 billion in added value and supported 4.2 million jobs in 2014 alone.[5]

From the sustainability perspective, these biobased products have a noticeable environmental impact by displacing about 9.4 million barrels of oil a year. There is also a calculated potential to reduce greenhouse gas emissions by an estimated 127 million metric tons of CO_2 equivalents per year.[6]

In 2021, the federal government purchased $500 billion in goods and services. That translates to $500 million in biobased material purchases, even while holding conservatively to the 0.1% conversion ratio.

TABLE 2—BioPreferred® Coatings Categories[3]

PREFERRED® CATEGORY	MINIMUM BIOBASED CONTENT (%)
Interior Paints and Coatings	
Latex and Waterborne Alkyd	20
Oil-based and Solventborne Alkyd	67
Neither Oils nor Solvents Included	25
Exterior Paints and Coatings	83
Industrial Enamel Coatings	25
Polyurethane Coatings	25
Epoxy Systems	23
Roof Coatings	20
Water Tank Coatings	59
Wood and Concrete Sealers	
Penetrating Liquids	79
Membrane Concrete Sealers	11
Wood and Concrete Stains	39
Intermediates	
Binders	22
Chemicals	22
Paint and Coatings Components	22
Lubricant Components	44
Oils, Fats, and Waxes	65
Paint Removers	41
Inks	
Specialty	66
Printer Toner (> 25 pages/minute)	20
Printer Toner (< 25 pages/minute)	34
Sheetfed (Black)	49
Sheetfed (Color)	67

In fact, the last six months of 2021 experienced the largest growth in the intermediate categories of rubber materials (>300%), textile processing materials (38%), fibers and fabrics (36%), and chemicals (31%). This indicates that companies are more successfully incorporating sustainability through biobased materials and coatings to the point of certification.

It also suggests that in some instances, there has been progress in meeting performance attributes at least equal to those derived from using higher levels of petroleum-based materials. Finally, it also appears that the USDA BioPreferred® certification is worth a company's time and money for development of biobased products. These are positive steps for the biobased market, even though, at a 0.1% conversion ratio, it seems to be taking a long time to materialize.

The biobased material market is clearly gaining traction and visibility when a consumer giant such as Proctor and Gamble successfully integrates renewable materials into their flagship classic Tide detergent. Their Tide purclean™ detergent is USDA Certified Biobased, free of dyes, chlorine, phosphates, or optical brighteners.

As for performance, this plant-based laundry detergent is suitable for both high efficiency (HE) and regular washing machines and reviewers are satisfied with cleaning and stain removal. This development, along with acceptance from the consumer, bodes well for the ultimate future of biobased coatings.[7]

INDUSTRY SOLUTIONS—COLLABORATION

As mentioned earlier, the coatings industry is not overwhelmingly staffed with research and development personnel to delve into basic research in the biobased arena. Those companies that have developed somewhat viable portfolios have often collaborated with each other, academia, and supporting technology companies.

Sustainable development is a major challenge for the chemical and coatings industry. Reconciliation of science, technology, economic viability, performance, and consumer approval are some of the keys to a truly viable market share.

Here is one example where basic research done outside the typical paint company confines can be translated into viable commercial product. Laboratoire de Chimie des Polyméres Organics (LCPO) in France has been on the cutting edge of polymer synthesis for biobased building blocks.

The core expertise of LCPO in biomass chemistry allows a deeper dive into the methodologies and concepts of green chemistry. While their researchers are academic experts, their practical objective is to design high-value-added synthons and polymers to design functional biomass polymeric material with features useful for sustainable technologies for various markets.

Some of the basic research used to bring bio-coatings to the forefront include biomass deconstruction, controlled depolymerization of biopolymers and polymer synthesis directly from biobased building blocks.

The aim is to combine the cutting edge of polymer chemistry with industrial implementation of the latest advances of bio-sourced products, including coatings. LCPO says that this is one of the first converging solutions to minimize the environmental impact of traditional polymer-based materials and technologies.

This type of expertise can create the basis of pathways for designing new tailor-made building blocks and original macromolecular architectures. The diversity inherent in hetero-functional building blocks from biomass are a huge opportunity to develop new polymers with properties that are not available even in petroleum-based materials.

Some contributions to date from LCPO are novel. The company's deep expertise in the use of derivatives resulting from vegetable oil (triglycerides) and lignin (phenolic substrates like vanillin) is developing synthons that can be used for new pathways in the synthesis of polyurethanes, polycarbonates and epoxy thermosets.

One of LPCO's chief contributions to date was in 2015 with the use of triglyceride derivatives for synthesizing polyurethane precursors, particularly aliphatic polyamines and poly(cyclic carbonates). Polyaddition of poly(cyclic carbonates) through the amniolysis of cyclic carbonates provides a route to an isocyanate-free polyurethane.

In 2016, new routes for polyamine and poly(cyclic carbonates) starting from aliphatic backbones derived from fatty acids were developed. The carbonate functions were derived from glycerol, which, with fatty acids, is the other constituent of a triglyceride.

The new biobased precursors addressed several limitations of polyurethane synthesis based on polyadditions of amine and cyclic carbonate. The carbonate-amine reaction was accelerated and *matched the kinetics of conventional isocyanate-alcohol chemistry* by activating the cyclic carbonate with hetero-functions in the ß-position of the cycle.

This allows conventional, already built, high capital-intensive equipment to be used with only minor modifications to the process. This game-changing concept makes biomass-based coatings suddenly become more practical, cost effective, and by definition, more viable (assuming, of course, that field performance is not compromised).[8] *Figure 1* presents some of the chemical structures developed from this research.

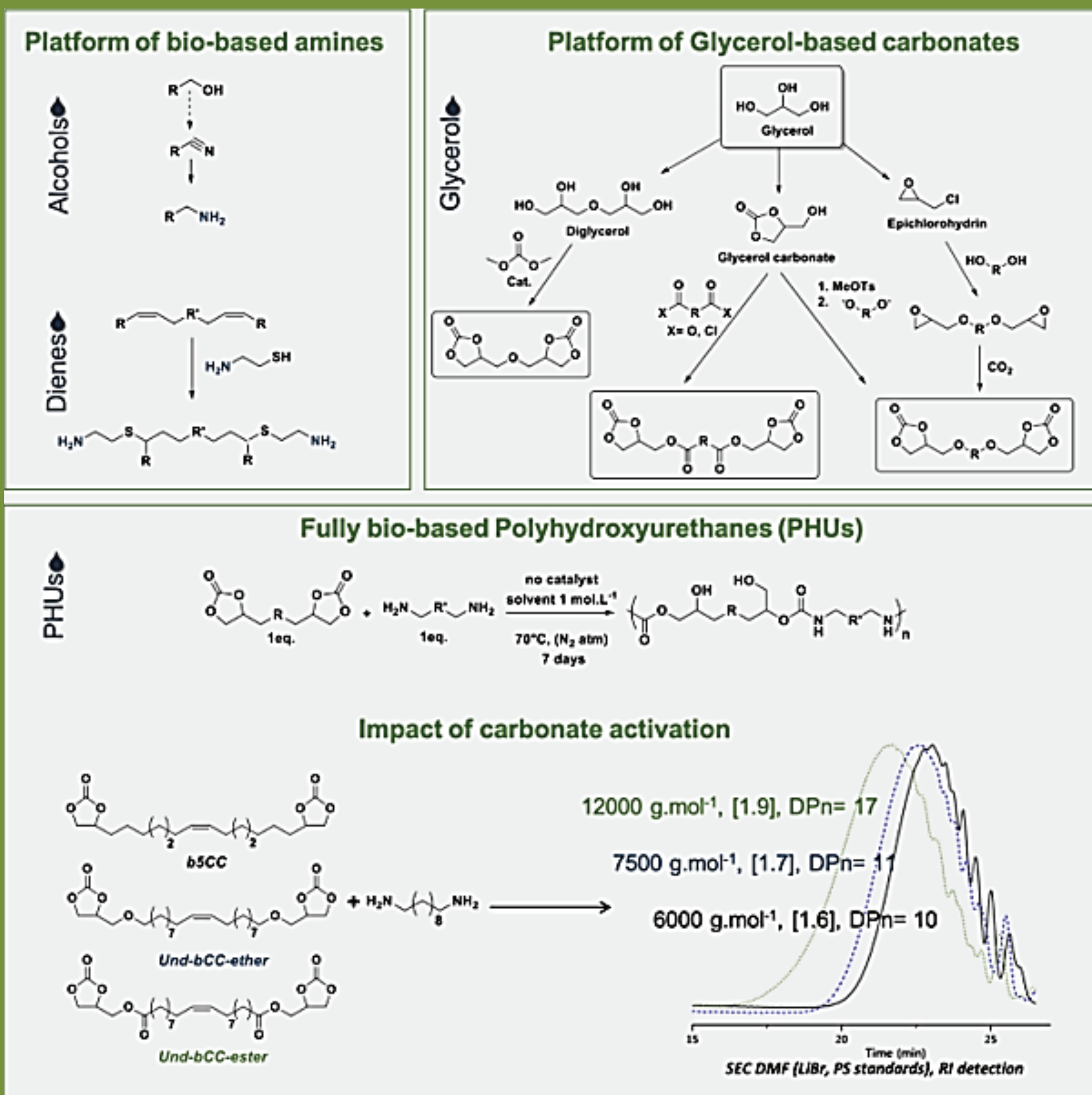
Using the results of the deep-dive fundamental research work done at LCPO is one way that the coatings industry can leverage the development of bio-coatings despite a shortage of industrial research scientists.

Another way to place the needed emphasis on biobased research that leads to actual market implementation is to participate in a consortium consisting of companies, research institutes, and universities formed to develop specific aspects of biobased portfolios, rather than take on the whole biobased coatings world.

One of the more interesting and focused consortium experiments, the SUSBINCO project in Finland, is targeting sustainable biobased binders that can be used in fiber-based packaging, gaskets, wood products, paints, adhesives, and sealants. Eleven companies and seven research organizations and universities officially joined co-innovation forces in September 2021 to research and develop **SUS**tainable **BI**obased bi**N**ders and **CO**atings.

The project is well funded through Business Finland with a *"Veturiohjelma"* (Venturi) budget of €10.1 million.

FIGURE 1—Chemical structures developed for biobased amines, glycerol-based carbonates and fully biobased polyhydroxyurethanes.[8]



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Venturi programs are designed to accelerate developmental initiatives within the strategic circular bioeconomy paradigm in Finland. Professor Chunlin Xu from the Laboratory of Natural Materials Technology at Åbo Akademi University is leading this ambitious project, which will run through November 30, 2023.

SUSBINCO has targeted sustainable biobased binders and coatings made with 80-100% biobased content that can be used in fiber-based packaging, gaskets, wood products, paints, adhesives, and sealants with a subgoal to make Finland a global leader in biobased packaging. The initiative is also part of the Strategic Research and *Innovation* Agenda (SRIA) for the *Forest*-Based Circular Bioeconomy sponsored by CLIC Innovation.

The consortium is divided into five work groups. Within these groups, research organization partners will work closely with industrial partners to identify and target all the demands of various biobased binders for different product applications. The point here is that a well-funded academic/industry coalition targeting binders would be able to develop highly competitive products in a very short time.

Figure 2 depicts the consortium work groups and associated organizations. Note that each group plays a role in the value chain and is paired with a research partner: Raw material suppliers, formulators, technology providers coatings and end-users are integral to the development cycle.[9]

The initiative is not only ecologically motivated, but more importantly, economically focused. The stated aim of SUSBINCO is to develop sustainable biobased binders and coatings as durable and safe alternatives for fossil fuel materials. The targeted products have a total market value of €1000 billion.

Because this is a very ambitious two-year program, non-food-competing, wood-based alternatives needing limited chemical modifications will be targeted first. Product performance, raw material availability, technical feasibility, economics, applicable regulations, and sustainability will be woven into the developmental matrix.

Cradle-to-grave aspects will also be foremost in the development. Further, the project will focus on using offal and residual material such as bark, sawdust, and pulp. Materials development will be the basis for addressing changes in processing.

If this project is successful, it could be a model for in-depth yet practical investigations into other paint and coating building blocks that will provide basic technology throughout the industry.

RESEARCH AND DEVELOPMENT ADVANCES

In the past couple of years, several novel solutions have come into commercial view on the molecular-building-block level. One involves the development and usability of carbodiimide crosslinkers as environmentally friendly enhancers for coating characteristics.

Crosslinking works by connecting the polymers through reactive chemistry, thereby strengthening the molecular network of a specific coating. Even so, a high degree of crosslinking is not advantageous for flexibility, elongation, and strength.

Waterborne systems require a high level of flexibility to maintain desired characteristics. Polycarbodiimide crosslinkers have a number of advantages over traditional aziridine, isocyanate, and melamine crosslinking agents. They are much less sensitive to the presence of water or moisture and can be formulated to have long pot life. These materials are highly reactive, enabling

FIGURE 2—Schematic of the SUSBINCO alliance.[9]

curing with carbonyldiimidazole (CDI) crosslinking at room temperature and/or under typical oven conditions used for drying applied coatings.

More importantly, these CDI crosslinking chemistries can also be suitable for 2K coating systems through 2-stage reactivity, as shown in *Figure 3*.[10] In this case, a second reactive group is attached to the polycarbodiimide so that upon curing, the crosslinker reacts to the -COOH groups in the polymer chain.

This allows the development of a much denser network structure leading to improved adhesion, abrasion resistance, strength, hardness, and chemical and stain resistance. In addition, these crosslinkers are non-toxic unclassified products, which allows for utilization within a wide range of regulatory environments.

Film-forming carbodiimide homo- and copolymers are already used in amino encapsulation techniques for pressure-sensitive adhesives. A variety of carbodiimide crosslinkers are used for dying wool or hair, as they enhance color fastness.

Polycarbodiimides have practical coatings applications for primer adhesion and as an economical way to enhance topcoat surface characteristics such as hardness and mar resistance. Leather properties are enhanced with polycarbodiimide crosslinking agents.[11]

Some work remains to determine the optimum amount of crosslinker for various resins to improve properties without crossing into brittle behavior. Once these formulation issues are solved, commercial applications are considered relatively easy to implement.

THE NEXT FRONTIER

While basic and commercial research into biomaterials and bio-coatings continues to evolve by providing building-block alternatives to current fossil-fuel-based products, another environmental movement is gaining momentum.

The Plastic Waste Challenge was founded in 2010 by researchers from the Hebrew University of Jerusalem in conjunction with industrial composite experts from the clean-tech sector. The basic goal was to keep used products out of landfills and waterways and at the same time enhance recycling methods through biodegradation and composting.

The coatings that resulted from this initiative are the best definition of sustainability—truly from cradle to grave—and beyond.

In January 2022, two coatings were commercialized for use by paper and packaging producers whereby cellulose-based barrier coatings were applied to packaging paper to protect foods. These two-coat systems act as a barrier against oxygen, oil, and grease transmission, while at the same time protecting from water-vapor transmission.

Coatings are applied as a primer and topcoat onto paper substrates. The resulting system essentially replaces plastic products. The coating that inhibits grease, oil, and oxygen transmission can even be applied singularly to recyclable plastic to replace non-recyclable plastic packaging that currently ends up in landfills.

To begin, cellulose nanocrystals were extracted from wood pulp that came from trees grown in an industrial forest. This material was selected because cellulose is one of the most abundant biopolymers on the planet.

It is the core that gives plants their strength yet is lightweight—the perfect composite. More importantly, it is a material that is easily renewable, particularly because care has been taken to harvest from industrial forests planted specifically for this type of use.

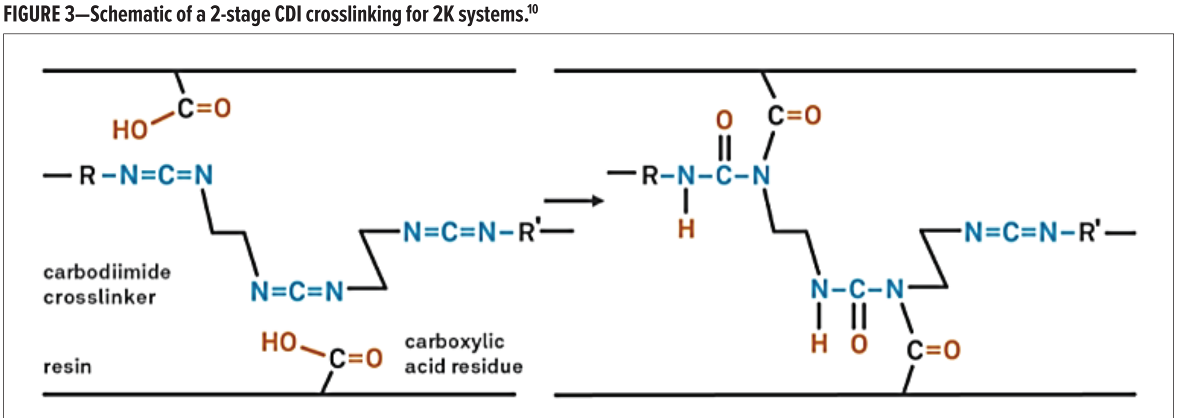
Coatings can be applied to paper at film thicknesses tailored for the application. Films ranging from ultrathin to thick are applied onto paper used for pouches, lids, and cartons using conventional paper-coil-coating methods such as slot dies and roll coaters with smooth or gravure rolls. This means that there is no need for capital investment for new or retrofitted equipment—thereby making the coatings cost effective.

Finally, the coatings are recyclable, biodegradable, and compostable, providing many options for responsible and more sustainable alternatives to plastics ending up in the oceans or littering landfills.

Medium-size Tier 1 food and consumer goods companies across the packaging value chain have conducted shelf-life and other specification testing and found these coatings to perform very well in pilot testing—comparable to plastic products currently on the market.

These coatings are an example of a confluence of sustainability parameters: renewable, ecofriendly resources, the ability to run coating production on conventional equipment without increasing cost, demonstrating the required performance characteristics and ultimately dying a clean death by composting or biodegradation (or recycling if preferred).[12]

FIGURE 3—Schematic of a 2-stage CDI crosslinking for 2K systems.[10]



One of the reasons that this coating conversion was successful is that the life-cycle environmental impact assessment dovetailed into the techno-economic analysis of green bio-coatings. Consumer awareness and regulation may be two big drivers, but so too are competitive performance and cost.

Successful bio-coatings have long moved past formulating for low VOCs. Today, the intersection of systemic energy and resource saving strategies, renewable resources with versatile functional capabilities, lower prices, manufacturing considerations, and waste minimization across the board are the new paradigms.

Performing a holistic sustainability assessment including cradle-to-grave and beyond determinations, coupled with realistic cost parameters and product targeting will enable the coatings industry to develop viable, long-lasting bio-coatings. ❊

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