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CHEMURGY

Chemurgy is defined as that branch of applied chemistry devoted to industrial utilization of organic raw materials, especially from farm products. A more modern and general definition for chemurgy is the use of renewable resources particularly biomass, usually plant or microbial material, for materials and energy (see Fuels from biomass; Fuels from waste).

Chemurgy was really a social movement during the 1920s and 1930s when there were large surpluses of agricultural materials and severe economic problems in the farm areas. Using farm commodities as chemical or industrial raw materials was seen as a means of solving the economic problems (1-5). One significant outgrowth of this movement was the founding of the regional laboratories of the U.S. Department of Agriculture (USDA) which were considered to be chemurgic laboratories (6). Another success involved the making of strong paper from the fast-growing southern pine, leading to the foundation of the southern pulp (qv) and paper (qv) industry. This industry had been largely centered in the north and northeast and based on slower growing species.

Some of the early work on the manipulation of proteins (qv) arose out of the chemurgy movement. This technology has found application in synthetic meat production. Other technologies that may be classified as chemurgic include that of the cellulosic fibers, eg, rayon and cellulose acetate (see Cellulose esters; Fibers, cellulose esters); the recovery of turpentine and rosin from paper and pulping processes (see Tall oil; Terpenoids); and the oils and fatty acids business (see Carboxylic acids; Fats and fatty oils). Oils and fatty acids are used in a wide variety of chemical products including soap (qv) and detergents (see Detergency; Surfactants), cosmetics (qv), and coatings (qv).

1. Industrial Materials from Renewable Resources

One distinction that can be made in the area of chemurgy is between the use of natural products that are grown solely for industrial purposes as compared to those that are grown primarily for food. In the latter class, industrial materials may be either by-products from food production or substitutes for food uses when the commodity is in surplus.

1.1. Industrial Crops as Raw Materials

Trees are by far the largest commodity grown solely for industrial use. About two-thirds of the trees harvested are used for construction or structural uses (see BUILDING MATERIALS, SURVEY; Wood) and about one-third are used for pulp and papermaking. About one-third of the pulp is made from residues of other timber conversion, such as trimmings and sawdust. Small quantities are used for fuel. Plywood is manufactured by peeling large strips from logs and then laminating with adhesives (qv). Still more complicated wood products are made from chips or slivers pressed together using binders such as phenolic resins (qv) (see Wood-based composites and laminates) (7).

Other purely industrial crops include cotton (qv) and flax. Cotton is grown primarily for its fiber, although the protein and oil contained in the seed also contribute to farm income. Manufacture is simply a mechanical separation of the fiber from the seeds, followed by cleaning and mechanical processing into thread and ultimately textiles (qv). Flax is grown primarily for its seed which yields linseed oil, a drying oil used in coatings (qv) (see Drying oils). The fiber from flax grown in the United States is short and is used in certain types of fine papers such as cigarette papers. Longer flax fibers grown in other parts of the world are manufactured into the fine textile, linen.

There are relatively few other crops that are grown solely for industrial purposes. Among these are tung, a tree nut from which tung oil is produced, and castor from which is obtained castor oil (qv). Tung oil can be used as a drying oil in coatings. Wild crops being investigated for cultivation include jojoba, crambe, guayule, kenaf, and lesquerella.

Jojoba is a desert crop that gives a small bean containing about 50% of a wax, a fatty acid ester with a fatty alcohol. The only other large source of such a wax is sperm whale oil, traditionally used in fine lubricants (see Lubrication and lubricants). Because the sperm whale is an endangered species, relatively little sperm whale oil is available and there is a large market for a substitute. Jojoba oil has been found to be usable for most of these applications. The jojoba oil is obtained by simply pressing the nut followed by conventional refining. Some jojoba oil is used in cosmetics (qv).

The problems with jojoba as a commercial crop are the usual ones of domestication and cultivation. It is a slow-growing plant, available only in the wild and therefore has very wide genetic variability. Efforts are underway to select the most promising variants and cultivate these as a crop in the southwestern United States deserts (7). A possible alternative for producing jojoba oil is to culture plant embryos in bioreactors (see Cell culture technology).

Guayule, potentially a source of natural rubber, is an unusual crop in that it has been an article of commerce in the past. Guayule grows wild in northern Mexico and the southwestern United States. When the leaves are milled in water, a latex is released that coagulates into natural rubber worms. These can easily be collected and relatively easily refined to give a product that is almost identical to the natural rubber from southeast Asia. During World War II there were several thousand acres of guayule planted in California and a small plant established to extract the rubber for military use. After the war, however, this effort was shut down. Because it is believed that natural rubber trees may have reached their genetic potential and because these trees are in politically vulnerable areas, guayule may once more become an important crop (8). Guayule may also have use as a pesticide or coating (9, 10).

Crambe was introduced to the United States in the 1970s. It is an oilseed, the oil of which is very high in erucic acid [112-86-7] (13-docosanoic acid), $C_{22}H_{42}O_2$. This oil can be used to provide industrial lubricants, especially those needed for the basic oxygen furnace process for making steel (qv). Crambe is grown in relatively small volume in the midwestern United States.

Kenaf is a grass fiber crop that has been proposed as a papermaking source. It is an annual crop and can be grown in many parts of the country. There have been agronomic problems with kenaf, primarily its vulnerability to nematode pests. Another problem is that kenaf is harvested once a year and must then be stored, usually as silage. There is some loss of the fiber in storage.

Lesquerella is an oilseed crop that has been mentioned as a potential industrial crop but is not yet grown in significant quantities. Castor beans have been a crop in the United States and provide a well-known lubricating product, castor oil [8001-79-4]. However, castor is grown only in small quantities and most of the castor used in the United States is imported from Brazil (11).

The economic disposal of oilseed meal presents a problem for almost every new oilseed. The large-volume oilseeds such as soy and peanut are particularly valuable because their oilseed meals contain large amounts of protein, which can be used almost directly for animal or even human food (see Feeds and feed additives). Many oilseeds contain toxic or other undesirable materials complicating use. For example, the crambe seed contains toxic and allergenic substances that must be removed before being fed to monogastric animals. A large part

of the research on crops and oilseeds is devoted to upgrading the seed meal once the oil has been obtained by conventional pressing and solvent extraction methods. The seeds may be cooked first to release the oil. Cooking is often by direct contact with steam.

1.2. Food Crops as Raw Materials

Crops that are grown primarily for food can be used for industrial purposes when the crops are in surplus or are found to be unfit for their intended purpose. Historically, when agricultural surpluses are high, the distinction between industrial and food use is not significant. Additionally, there are large industries based on food commodities; eg, the corn and wheat starch separation processes to give starch (qv) used in paper sizing and textiles (see Wheat and other cereal grains). The starch is separated primarily by gravity in water slurries. Other starchy cereals could as easily be used, eg, milo or grain sorghum, rice, oats, and barley, although rice, oats, and barley are usually too valuable as food crops to be diverted to starch production. Potatoes and sweet potatoes also can be fractionated to give a very good starch product. Usually only the culls from fresh or processed potato manufacture are used for this purpose, but this is an important economic use because of the consumption of materials that would otherwise be wasted. Potato starch manufacture has suffered in the 1990s because the effluent from the process is a severe pollutant. Research has been directed to the utilization of this material, which contains high quality protein (12).

Oilseeds grown primarily for use in salad dressings, margarines, and cooking oils also produce an important by-product in the form of high protein meal. Historically, this meal was fed to animals but increasingly it is refined and fractionated for human consumption. Soy, in particular, is used to produce a large variety of specialized protein concentrates and isolates by various refining processes usually involving caustic solution and precipitation. A large quantity of soybean oil is also epoxidized to make an important plasticizer. Some other oilseeds that are important in the United States include rapeseed, sunflower seed, and safflower.

By-products from meat animals, which may be viewed as a food crop, are also commercially significant (see Meat products). In addition to meat, each of these produces bones, trimmings, fat, hides, and in the case of sheep, wool (qv). The hides are turned into leather (qv) by tanning or are converted to commercial gelatin (qv) or other animal glue products. Inedible animal tallow may be exported and refined in foreign countries for edible use; some is added to animal feeds. A large amount is used in soaps and detergents or converted to fatty acids that may be refined or used as a mixture in soaps. Bristles from pork are used to a small extent in brushes. Animal hair is a waste product that creates disposal problems. Uses are still being sought for this material.

1.3. Wastes or By-products as Raw Materials

By far the largest volume of natural products for industrial use, aside from the forest products, are wastes or by-products of food processing (qv). The largest use of these wastes is as animal feeds. Because they are used rather than becoming a disposal problem, they are considered to be chemurgic products.

Wastes from the pulp and paper industry are finding increasing applications. These include lignin (qv) and tall oil (qv), as well as sugars in the form of sulfite waste liquor. Although the largest use of lignin [9005-53-2] is as a fuel in the pulping process, a wide variety of products can be made from it. Lignin, a polymer containing aromatic and phenolic functionalities, can be converted to a mixture of aromatic and phenolic compounds by hydrogenation. These compounds occur in large variety and only relatively small quantities of any one substance can be isolated. In this respect, converted lignin is more like coal tars from coking than fractions from crude oil. Lignin is used in dispersants (qv), adhesives (qv), additives to drilling muds (see Petroleum, drilling fluids and other oil recovery chemicals), and fillers (qv).

Tall oil [8002-26-4] has been referred to as the largest and fastest growing source of extractives such as turpentine and resin. It can be refined to give tall oil fatty acids (see Carboxylic acids) and tall oil pitch as well

as resins. These fatty acids compete with fatty acids from vegetable sources for many of the same industrial markets.

Sulfite waste liquor from the pulping industry contains a large amount of hydrolyzed sugars. Some sulfite waste liquors are now being fermented to give both ethanol (qv) and single-cell protein in the form of yeast (see Foods, non-conventional; Yeasts).

The trimmings and slash from forest operations historically have been left in the forest but increasingly these materials are being used in much the same way as higher quality timber, primarily for pulping or chipping. Agriculture produces large amounts of wastes in the form of animal manures, branches, stems, stalks, and straws. These wastes also have historically been returned to the land. Changing patterns of production have made it less convenient to use these for their fertilizer value, however, and have aggravated the pollution problem they create. It is possible to recycle animal wastes as animal feed because a large amount of nutritive value is retained in the waste. The enormous volume of animal waste is also being considered as a potential energy source, probably by anaerobic fermentation to produce methane [74-82-8] (13). Other agricultural wastes, such as straw, have been considered for pulp and papermaking and for digestion to produce fuel. They are also candidates for hydrolysis of the cellulose content to produce glucose (see Carbohydrates). Hydrolysis can be acidic or enzymatic, and products are primarily ethanol but can include other chemicals (14–16).

The processing of agricultural products to make foods and feeds yields wastes that have traditionally been considered valueless. Constraints on dumping make it necessary to consider these wastes as raw materials. A good bit of research into uses for wastes has been motivated primarily by environmental considerations. For example, kraft black liquor, sulfite waste liquor, and other dilute streams from pulp and papermaking are potential fermentation substrates because of the dissolved sugar, which is also the material that is the most serious pollutant. Cereal grain milling produces a variety of fractions that have found historic uses in animal feeds. Research has been conducted on isolation of protein concentrates from these materials for upgrading as human foods.

Cellulosic materials, such as farm wastes, can be upgraded for animal feed by simply bringing them into contact with ammonia(qv). The cellulose swells and is made more digestible, and at the same time some ammonia nitrogen, which is a nutrient for ruminants, is left behind. Supercritical ammonia improves susceptibility to enzymatic hydrolysis (17).

Some of the forest wastes and pulp and paper processing wastes contain dilute concentrations of important acids such as acetic acid (see Acetic acid and derivatives) and solvents such as methanol (qv), formed from the wood in the high temperature digestion of pulping. Although these waste streams have been discarded as too dilute for commercial recovery, it is increasingly attractive to use materials from such processes as liquid–liquid extraction (see Extraction, liquid–liquid), or adsorption (qv) on charcoal followed by steam stripping can be employed to remove and concentrate the organic chemicals.

2. Processing Methods

The largest class of processes applied to farm commodities are separations, which are usually based on some physical property such as density, particle size, or solubility. For example, the milling process for cereal grains involves size reduction (qv) followed by screening to yield products that have varied concentrations of starch, fiber, and protein. Milling of water slurries is practiced to obtain finer separation of starch, fiber, protein, and oil.

Solubility is used in the refining of oilseed meals to give protein isolates and concentrates (18). Proteins are highly soluble in basic solutions and the processes of isolation and concentration involve repeated dissolution, physical separation by centrifuging or filtration, followed by precipitation of the protein. Solubility properties are used to extract oil from oilseed meals. The usual solvents are light hydrocarbons such as hexane. There is also some separation on the basis of volatility such as the distillation of essential oils, the recovery of solvent from edible oils, and the distillation of esters of fatty acids.

In general, the separation processes used in chemurgy are relatively simple. In contrast, the chemical reactions that are possible with chemurgic materials can be very sophisticated. The chemistry involved is often reductive because so many of the chemurgic materials, such as cellulose and starch, are carbohydrates. Also important are the carboxylic acids and the amine groups of protein. Reactions such as the esterification of sugar (qv) and the epoxidation of soy oil are of commercial significance. Also important are the acetylation of cellulose to make it soluble for subsequent spinning into fibers, and the solubilization of starch and cellulose as xanthates (qv).

Increasingly, biochemical transformations are used to modify renewable resources into useful materials (see Microbial transformations). Fermentation (qv) to ethanol is the oldest of such conversions. Another example is the cell-free enzyme catalyzed isomerization of glucose to fructose for use as sweeteners (qv). The enzymatic hydrolysis of cellulose is a biochemical competitor for the acid catalyzed reaction.

3. Uses

A rather impressive list of materials and products are made from renewable resources. For example, per capita consumption of wood is twice that of all metals combined. The cellulosic fibers, rayon and cellulose acetate, are among the oldest and still relatively popular textile fibers and plastics. Soy and other oilseeds, including the cereals, are refined into important commodities such as starch, protein, oil, and their derivatives. The naval stores, turpentine, pine oil, and resin, are still important although their sources are changing from the traditional gum and pine stumps to tall oil recovered from pulping.

Textile fibers are made from chemurgic materials such as cotton, rayon, linen, and wool (qv).

The transportation industry utilizes chemurgic materials such as fuels, lubricants, and coatings. Ethanol is added to gasoline to form gasohol and sold as a motor fuel (see Alcohol fuels). Additionally, vegetable oils can be used as fuels for diesel engines. Lubricants of the 1990s are subject to higher stress than previously because of antipollution devices and smaller engines. Whereas historically castor oil (qv) has been an important automobile lubricant, its application in that use is now small. Biomass materials can also be pyrolyzed to produce oils and fuel gas (19–22).

The housing industry represents a large market for chemurgic materials. Wood is the cheapest material of construction generally available and has the advantage of being a good thermal insulation material. In addition to use as foods, large numbers of food additives can be made from agricultural products; for example, modified starches, cellulose gums, flavors and aromas from turpentine, and other less obvious applications (see FLAVORS; Gums; Perfumes; Terpenoids). Also, rosin can be used to produce resins that compete directly with resins from petroleum (see Hydrocarbon resins; Resins, natural).

The fermentation industry is based almost exclusively on renewable materials in the form of molasses, starch, etc. Most products are of very high value and relatively low volume such as antibiotics (qv) (23).

Industrial alcohol (ethanol) may be made by fermentation or by hydration of ethylene. Beverage alcohol must be made by fermentation (see Beverage spirits, distilled; Beer; Wine). Since 1973, increases in the price of ethylene have made industrial alcohol by fermentation again competitive at ca \$0.25/L (\$1/gal). Fermentation plants that had been shut down for many years have been reopened and are likely to gain an increasing portion of this business. However, the efficiency of conversion of glucose to ethanol is less than 50% because so much carbon dioxide is also made. The state governments in the grain growing mid-western United States have revived the idea of ethanol as a fuel and have passed legislation providing economic incentives for the use of fermentation-based ethanol as a gasoline additive to form gasohol as a motor fuel (see Gasoline and other motor fuels) (24). An extensive ethanol fuel program based on sugar cane has been developed in Brazil.

Grain that is usable as food or feed is an expensive substrate for this fermentation process. A cheaper substrate might be some source of cellulose such as wood or agricultural waste. This, however, requires hydrolysis of cellulose to yield glucose. Such a process was used in Germany during World War II to produce yeast as a protein substitute. Another process for the hydrolysis of wood, developed by the U.S. Forest Products Laboratory, Madison, Wisconsin, uses mineral acid as a catalyst. This hydrolysis industry is very large in the former Soviet Union but it is not commercial elsewhere.

More recently, interest has developed in the use of enzymes to catalyze the hydrolysis of cellulose to glucose (25–27). Domestic or forest product wastes can be used to produce the fermentation substrate. Whereas there has been much research on alcohol fermentation, whether from cereal grains, molasses, or wood hydrolysis, the commercial practice of this technology is primarily for the industrial alcohol and beverage alcohol industries. About 100 plants have been built for fuel ethanol from corn, but only a few continue to operate (28).

Generating energy from biomass in some form is a use of chemurgic materials. The direct combustion of plant materials or fermentation to yield methane for combustion have both been topics for research. Wood has been burned as a fuel for many years and generates more energy in the United States than do nuclear sources. The bulk of this use was in forest products industries where wood wastes were burned on-site for energy generation. Small-scale anaerobic fermentations of agricultural wastes to yield methane are practiced but large-scale processes have not yet been built, except in sewage treatment plants to dispose of sludge (see Wastes; Water, sewage).

4. Potential for Renewable Resources

A 1976 study (29) on renewable resources for industrial materials emphasized forest products because these represent the largest volume of renewable resources; however, this study also considered such things as special crops, animal by-products, and marine by-products. The conclusion was that competition between chemurgic material and one from a nonrenewable resource would be resolved by economics and that there were significant energy economies in the use of renewable resources for a great many materials. One exception was the cellulosic fibers, acetate and rayon, which consume relatively large amounts of energy. This study was summarized (30).

The potential for converting wood to plastics has been considered (31). Because plastics and polymers are among the largest volume industrial materials, the ability to convert chemurgic or renewable resources to these substances would clearly be significant. With few exceptions, industrial polymers and plastics are based on petrochemicals (see Feedstocks, petrochemicals and feedstocks). Each of these important building blocks, primarily ethylene (qv), propylene (qv), and benzene (qv), could in theory be obtained from trees or agricultural wastes by an integrated series of processes involving the hydrolysis of cellulose using either mineral acids or enzymes. Wood or agricultural waste converted to glucose can then be converted to ethanol, which in turn can be dehydrated to ethylene or used to make butadiene(qv). These olefins are necessary for a large class of polymers, including the polyolefins and styrene(qv) (see Olefin polymers; Styrene plastics).

Hydrogenolysis of glucose, sucrose, starch, or cellulose can lead to glycerol(qv) and propylene glycol (see Glycols) and lignin can be hydrogenated or hydrolyzed to yield large fractions of aromatics and phenolics which in principle could be used as feedstocks. Thus the entire petrochemical base of polymers could be replaced by lignocellulosic materials (32). Phenols from lignin can be made competitively if the price of petrochemical phenol (qv) is high enough. The by-products of lignocellulosic chemical conversions would presumably include extractives and the products of acid catalyzed degradation of glucose. These latter products, such as hydroxymethylfurfural, levulinic acid, and, from the degradation of pentoses, furfural, could be recovered for use in the chemical industry (see Furan derivatives).

Many problems need to be solved before chemurgic materials can be economically used as feedstocks. Among these problems are the recovery, purification, and fractionation of the diverse materials. However, none of these problems are insurmountable. Serious concerns are the supply of the raw material, the relative costs

of competitive materials, and competition with other uses for the raw materials. Competition is particularly significant because materials, such as wood, could easily be used in many cases for pulping or even higher value products, such as structural timber. Municipal solid waste offers a substitute raw material with few other uses (33).

The future of chemurgy is interconnected with the economic future of energy, environment, and food. Several scenarios exist for future sources of energy and materials. Some involve the use of nuclear or solar energy (qv) leaving coal and oil available for chemicals and transportation fuels. Some involve the use of coal (qv) and oil (see Petroleum) almost exclusively for energy. Materials would then come from other sources and this would place great demands on chemurgic processes involving wood, foliage (34), or other by-products. However, it is most likely that the competition of chemurgic materials with synthetic ones will be settled in the marketplace.

Another substitution taking place is the use of vegetable protein for meat protein. Whereas this substitution does not directly affect any nonrenewable resource, there are consequences for the increased availability of by-products of vegetable protein processing. Additionally, a rather complex competition exists among the various vegetable oils, animal tallow, and hydrocarbons. Most of the vegetable oils are essentially interchangeable although the processes needed to make them useful vary depending on the type of contamination they may contain. An example of this is the removal of color from palm nut oil. Tallow can be fractionated to give materials equivalent to vegetable oils as well as higher melting materials. Triglycerides in fats, oils, and tallows can be used for many of the same purposes as hydrocarbons such as lubrication, as a fuel, or, as in the past, for illumination. However, as meat production declines or stabilizes, the relative amount of tallow available also declines.

The economic balance must be considered between recovery, reuse, and modification of a waste material or by-product and its disposal. The future is expected to bring increases in the practice of recycle, recovery, modification, and upgrading of wastes of all sorts, and a reduction in disposal by incineration (qv), biochemical oxidation, or discharge to the environment (see Recycling).

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Cellulose esters; Fibers, cellulose esters; Fats and fatty oils; Vegetable oils; Building materials, survey; Wood; Wheat and other cereal grains; Yeasts; Beverage spirit, distilled; Alcohol fuels; Water, sewage