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Obituary

Contributions of Roy L. Whistler to carbohydrate polymer science

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ABSTRACT

Contributions of Professor Roy L. Whistler (1912–2010) to the carbohydrate polymer field include the following: determination of structures of guaran, annual plant hemicelluloses, and other polysaccharides; fragmentation analysis to accomplish the structural determinations; electron microscopy of starch granules; identification of the products of oxidation of starch; preparation of new derivatives of starch and amylose; alkaline degradation of polysaccharides; preparation of synthetic carbohydrate polymers; polysaccharide nomenclature; compilations of the literature on polysaccharides in the form of books, book chapters, and reviews.

The aims of this journal are to cover "the study and exploitation of carbohydrate polymers which have current or potential industrial applications". Among the specific topics covered are studies of their structure, function, and chemical modification. With the addition of biological activities, these aims are congruent to the life, research, and philosophy of Roy Lester Whistler (31 March 1912 to 7 February 2010).[1]

Roy Whistler earned a B.S. degree in 1934 at Heidelberg College, where he majored in chemistry, physics, and mathematics. Heidelberg College is in Tiffin, OH (USA), the town in which he grew up. Immediately upon graduation, he entered the post-graduate program in chemistry at The Ohio State University (Columbus, OH) and earned a M.S. degree (1935) under the guidance of Professor M.L. Wolfrom. His Ph.D. work, which was done in the laboratory of Professor Ralph Hixon at Iowa State College (now Iowa State University, Ames, IA) (1935–1938), resulted in five papers, none of which involved polysaccharide chemistry.

However, Professor Whistler's association with polysaccharides (carbohydrate polymers) may have begun when he was a Ph.D. student in the laboratory of Professor Hixon, since Dr. Hixon was converting his research program from one studying the chemistry of simple sugars to one studying starch at that time. As a post-doctoral fellow in the laboratory of Dr. Milton Harris at the U.S. National Bureau of Standards (Washington, DC), Dr. Whistler developed a method to determine the uronic acid contents of polysaccharides (Whistler, Martin, & Conrad, 1940; Whistler, Martin, & Harris, 1940a, 1940b).

He plunged headlong into polysaccharide research when, in 1940, after only 2 years of post-doctoral research and only 28 years old, he was hired to head the Starch Structure Section of the newly established Northern Regional Laboratory of the U.S. Department of Agriculture (Peoria, IL). There, he perceived of and promoted the industrial utilization of amylose, beginning with investigations of compounds that would complex with amylose so that it could be separated from amylopectin by crystallization (Whistler & Hilbert, 1945). He converted the amylose into amylose triacetate, from which he made films and fibers (Whistler & Hilbert, 1944; Whistler & Richards, 1958a; Whistler & Schieltz, 1943) and determined that the fibers made from it had a tensile strength comparable to that of cellulose acetate (rayon) and greater elongation.

In 1946, at the age of 34, Dr. Whistler accepted a faculty appointment in the Department of Agricultural Chemistry (now the Department of Biochemistry) at Purdue University (West Lafayette, IN). Immediately, he began a program on the industrial utilization of agricultural crops (Whistler, 1976). He began promoting guar as

[1] A more complete biography of Professor Whistler can be found in Volume 64 of *Advances in Carbohydrate Chemistry and Biochemistry*.

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a crop from which to obtain a valuable polysaccharide (Whistler, 1948a, 1948b). At the same time, he initiated research to determine the structure (Ahmed & Whistler, 1950; Baker & Whistler, 1975; Whistler, Bachrach, & Bowman, 1948; Whistler & Durso, 1951, 1952; Whistler, Eoff, & Doty, 1950; Whistler & Smith, 1952; Whistler & Stein, 1951) and properties (Heyne & Whistler, 1948) of guaran and converted it into its triacetate for films and fibers (Smart & Whistler, 1949a). He was the first to claim conversion of guaran into a polysaccharide with properties more like those of locust bean gum by treating it with α-galactosidase (Whistler, 1982). His studies of the guar plant and guar gum culminated in a book (Whistler & Hymowitz, 1979).

Concurrently, he began studying and promoting annual plant hemicelluloses as potential industrial gums, an interest that stayed with him the rest of his career. This aspect of his research began with the preparation of holocellulose from a crop residue – corn cobs (Whistler, Li, & Dvonch, 1948). And, of course, early on he investigated films and fibers made from hemicelluloses (Smart & Whistler, 1949b; Whistler & Smart, 1950). Then, he began the work of determining their structures (Whistler, Bachrach, & Tu, 1952; Whistler & BeMiller, 1956; Whistler, Conrad, & Hough, 1954; Whistler & Corbett, 1955b, 1956a; Whistler & Hough, 1953; Whistler & Masak, 1955; Whistler & McGilvray, 1955a, 1955b; Whistler & Tu, 1951, 1952a, 1952b, 1953 (although additional data obtained about 4 decades later indicated that the proposed branch-on-branch structure for the highly branched hemicellulose B was incorrect); Feather & Whistler, 1962; Gramera & Whistler, 1963; Sannella & Whistler, 1962; Whistler & Gaillard, 1961; Whistler & Lauterbach, 1958a, 1958b) and their properties (Zhang & Whistler, 2004).

To determine the structures of guaran and the hemicelluloses, he extensively employed partial depolymerization using acids and enzymes (fragmentation analysis) (Whistler & Corbett, 1955b, 1956a; Whistler & Durso, 1951, 1952; Whistler & Hough, 1953; Whistler & Masak, 1955; Whistler & McGilvray, 1955a, 1955b; Whistler & Richards, 1958b; Whistler & Smith, 1952; Whistler & Stein, 1951; Whistler & Tu, 1951, 1952a, 1952b, 1953; Whistler et al., 1950, 1952; Whistler et al., 1954). His use of fragmentation analysis was greatly facilitated by his development of carbon (charcoal) column chromatography, whereby products of partial hydrolysis could be separated into oligosaccharide classes in sizeable yields (Whistler, 1954a; Whistler & Durso, 1950) and fabricated fraction collectors (Durso, Schall, & Whistler, 1951; Hickson & Whistler, 1953). The availability of pure oligosaccharides as a result of carbon column chromatography, a technique that was short-lived but was a citation classic during that time, allowed determination of their structures by methods available at that time. So he was able to isolate pure polysaccharides (by selective extraction and fractional precipitation) and to determine their structures with a high degree of certainty if they were mainly linear (xylans of the hemicellulose A fraction, guaran) and with limited certainty if they were highly branched.

He also investigated structures of other polysaccharides from sources that were available in large quantities, including processing waste products (Richards & Whistler, 1973; Whistler & Deszyck, 1953; Whistler, Linke, & Kazeniac, 1956; Whistler & Richards, 1958b; Whistler & Saarnio, 1957), okra polysaccharide (Whistler & Conrad, 1954a, 1954b), alginic acid (Whistler & Kirby, 1959), chia seed polysaccharide (Lin, Daniel, & Whistler, 1994), tamarind xyloglucan (Niemann, Carpita, & Whistler, 1997), and even soil polysaccharides (Whistler & Kirby, 1956).

Later, he became interested in the biological effects of polysaccharides on animals (Doane & Whistler, 1963; Nakahara, Tokuzen, Fukuoka, & Whistler, 1967; Singh, Whistler, Tokuzen, & Nakahara, 1974; Whistler, Bushway, Singh, Nakahara, & Tokuzen, 1976; Whistler & Kosik, 1971; Whistler & Towle, 1970; Whistler, 1970; Witczak & Whistler, 1987; Zhang, Zhang, & Whistler, 2003; Zhang et al., 2004).

Starch continued to hold his interest after moving to Purdue University. His first paper on starch from research done at Purdue University, published in 1948 (Whistler & Johnson, 1948), described the effect of acid-catalyzed hydrolysis on the retrogradation rate of amylose. This paper was followed by the first report of retrogradation rates of different whole starches (Whistler, 1954b). At that time, he had already begun research on developing high-amylose corn/maize, which he believed would be important commercially and for which he secured the collaboration of Professor H.H. Kramer (Davis, Kramer, & Whistler, 1955; Dunn, Kramer, & Whistler, 1953; Kramer & Whistler, 1949; Kramer, Whistler, & Anderson, 1956; Pfahler, Kramer, & Whistler, 1957) and examined polysaccharides from it (Dvonch, Kramer, & Whistler, 1951; Whistler & Doane, 1961). He examined the behavior of low-molecular-weight amylose with complexing agents (Dvonch, Yearian, & Whistler, 1950) and isolated maltooligosaccharides from the starch polymers using the same fragmentation and separation techniques used with the hemicelluloses (Whistler & Duffy, 1955; Whistler & Hickson, 1954; Whistler & Moy, 1955).

He conducted a thorough examination of the oxidation of starch with chlorine and alkaline solutions of chlorine (Hullinger & Whistler, 1951; Ingle & Whistler, 1964; Uchino & Whistler, 1962; Whistler, Masak, & Plunkett, 1956; Whistler, Mittag, & Ingle, 1965, 1966; Whistler & Pyler, 1968; Whistler & Schweiger, 1957) and other polysaccharides (Whistler, Mittag, Ingle, & Ruffini, 1966; Whistler & Schweiger, 1958) and the oxidation of amylopectin with hydrogen peroxide (Whistler & Schweiger, 1959), and hypobromite (Doane & Whistler, 1964), in each case determining the products of oxidation. Other contributions to starch science include development of a microcrystalline starch product (Whistler, 1992), surface derivatization of granules (Whistler, Madson, Zhao, & Daniel, 1998), and microporous starch (Niemann & Whistler, 1992; Whistler, 1989, 1991; Zhao, Madson, & Whistler, 1996); and aggregates of small granule starch (Whistler, 1995a, 1995b, 1998a; Zhao and Whistler, 1994a, 1994b), which was not novel, although he thought it was; but he recognized a potential use for the microporous starch and the aggregates of small granules. He also had an interest in small granule starch (Zhao and Whistler, 1994a, 1994b) because he correctly predicted their fat mimetic property (Malinski, Daniel, Zhang, & Whistler, 2003; Whistler, 1996) and in banana starch (Whistler, 1998a, 1998b; Zhang, Whistler, BeMiller, & Hamaker, 2005) because of its availability from an agricultural waste.

He made new derivatives, primarily of starch and its constituent polymers, but also of other polysaccharides (Arnott, Hukins, Whistler, & Baker, 1974; Sannella & Whistler, 1963; Whistler & Hirase, 1961; Whistler, King, Ruffini, & Lucas, 1967; Whistler & Medcalf, 1964a, 1964b; Whistler & Roberts, 1959; Whistler, Ruffini, & Pyler, 1968; Whistler & Shasha, 1964; Whistler & Spencer, 1961; Whistler & Towle, 1969; Whistler, 1967, 1968, 1969), including the addition of glycosyl units to unbranched polysaccharides (Whistler & Doner, 1970; Whistler & Roberts, 1961; Whistler, Unrau, & Ruffini, 1968).

A most important contribution he made to starch science was the introduction of the new technique of electron microscopy (having obtained one of the first electron microscopes from RCA) (Nikuni & Whistler, 1957; Whistler, Byrd, & Thornburg, 1955; Whistler, Goatley, & Spencer, 1959; Whistler & Spencer, 1960; Whistler, Spencer, Goatley, & Nikuni, 1958; Whistler & Thornburg, 1957; Whistler & Turner, 1955).

An early part of his research program was also the alkaline degradation of polysaccharides (Corbett, Richards, & Whistler, 1957; Whistler & BeMiller, 1958, 1960a, 1960b, 1961, 1962; Whistler, Chang, & Richards, 1959a, 1959b; Whistler & Corbett, 1955a, 1956b; Whistler & Richards, 1958b).

In addition to researching the chemistry and properties of native polysaccharides, he was the first to make synthetic carbohydrate polymers via polymerization of monomers (Das Gupta & Whistler, 1964; Dick & Whistler, 1964; Whistler & Goatley, 1961, 1962; Whistler & Hoffman, 1967; Whistler, Panzer, & Goatley, 1962; Whistler, Panzer, & Roberts, 1961; Whistler & Seib, 1964, 1966), although a 2004 review (Ladmiral, Melia, & Haddleton, 2004) states "The first glycopolymer synthesis was reported by Horejsi et al. in 1978" (Horejsi, Smolek, & Kocourek, 1978), even though that paper was published 16 years after Whistler's first paper.

(He had no journal papers on polysaccharides during the period 1976–1991 when he concentrated on small molecule carbohydrate chemistry.)

No summary of the contributions of Roy L. Whistler to the carbohydrate polymers field would be complete without mentioning his contributions to polysaccharide nomenclature, the compiled literature, and as a consultant. He joined the U.S. Carbohydrate Nomenclature Committee in 1951 and became chair of its Polysaccharide Nomenclature Subcommittee. Later, he also chaired the Polysaccharide Nomenclature Subcommittee of the (U.S.) National Research Council. His idea that names of polysaccharides should carry the suffix –an and other suggestions were first incorporated into his book *Polysaccharide Chemistry* (Whistler & Smart, 1953). Eventually a set of proposed nomenclature rules that came out of the two subcommittees were adopted by IUPAC (Anonymous, 1996, 1997). Because of his efforts, we now name carbohydrate polymers with names like guaran, carrageenan, arabinoxylan, and galactoglucomannan and speak of them collectively and generically as glycans.

Following his first book, *Polysaccharide Chemistry*, he was the organizer and co-editor of successful books on *Industrial Gums* (Whistler & BeMiller, 1959, 1973, 1993) and *Starch: Chemistry and Technology* (Whistler et al., 1965, 1967, 1984, 2009). He was the original organizer and co-editor of the *Methods in Carbohydrate Chemistry* series (Volumes 1–10, 1962–1994), which originated from a laboratory procedures manual he had put together for his students. He also contributed many review articles and book chapters to other works.

Industrial applications of his research were always in the front of his mind. He was a consultant to many companies that produced or utilized carbohydrate products. Among them were CPC International, Hercules, Kelco, and Westvaco. Although similar statements had been used for centuries, there is no doubt that Sir Isaac Newton wrote, probably sarcastically, in a letter to his rival Robert Hooke dated 5 February 1676 "If I have seen a little further it was by standing on the shoulders of Giants". Professor Whistler was certainly a giant who elevated us and gave us a broad vision of the landscape of polysaccharides and their potential applications.

Professor Whistler was an extremely hard-working person with a strong will to succeed, which he did. He touched the lives of many students, post-doctoral fellows, industrial scientists, and those involved in the business side of industry, in which he was equally comfortable. He was, and is, held in admiration, esteem, and respect by many in industry and universities, many of whom never met him in person. His legacy is knowledge, people/scientists he was involved in teaching and training, organizations, and literature, mostly in the field of carbohydrate polymers.

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