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THE TERMINOLOGY OF SEAWEED COLLOIDS[1]

By Dr. C. K. TSENG

SCRIPPS INSTITUTION OF OCEANOGRAPHY

SINCE the outbreak of the war, there has been a considerable interest among some of the United Nations in investigating and developing their seaweed resources. The principal stimulus, in most instances, has been an agar shortage. In 1940, America had a single agar factory, which made 24,000 pounds of agar. There are now four factories[2] actively engaged in this industry with a combined production capacity of about 200,000 pounds per annum. The principal agarophytes are *Gelidium cartilagineum* from California and Baja California, Mexico, and *Gracilaria confervoides* from Beaufort, N. C.[3]

The interest shown in agar does not stop there; it extends to related seaweed colloids capable of serving as substitutes for agar. One of these is carrageenin, generally known in commerce as Irish moss extract. Its principal source is carrageen, better known as Irish moss (*Chondrus crispus*), which is now obtained in large quantities from Massachusetts and Maine in the United States and from the Maritime Provinces in Canada. The name carrageen is also applied to *Gigartina stellata* (*G. mamillosa*), commonly harvested together with the *Chondrus* and used similarly. Previously the production of Irish moss extract was rather small, since most consumers preferred to buy the seaweed and make their own extract. In recent years, there has been a great demand by various industries for a highly purified, standardized product

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[1] Contributions from the Scripps Institution of Oceanography, New Series No. 259.

[2] There are three more factories, still in the pilot plant stage, but expected to produce agar soon.

[3] C. K. Tseng, *Food Industries,* 17: 140, 1945.

and hence the production of the extract is greatly increased. At present there are in the United States at least three companies producing the extract and many others processing carrageen. It has been estimated that the United States made about half a million pounds of the extract in 1944.

The other seaweed colloid which has received considerable attention lately is algin, an alkali-extracted polysaccharide from various kinds of kelps and fucoids. In the United States the kelps utilized are *Laminaria digitata* (horsetail kelp) and *L. saccharina* (broadleaf kelp) from Maine and Nova Scotia, and *Macrocystis pyrifera* (giant kelp) from California. Although relatively young, the algin industry has been growing steadily and fast, especially in America and recently in Great Britain. This is shown by the fact that in 1941 the United States produced about one and one-half million dollars worth of algin.[4] War has greatly stimulated the growth of this industry, and undoubtedly much more algin is now being produced than in the pre-war period. There are four concerns in the United States making algin and its derivatives.

While seaweeds have served rather extensively as food in the Orient, they have never been as popularly utilized for culinary purposes by the Occidentals. In the past, soda, potash and iodine had been extracted, and organic solvents such as acetone derived from seaweeds on a large scale in America and Europe. In countries enjoying relatively high standards of living, these materials can no longer be produced profitably from marine plants, since they can be obtained more cheaply elsewhere. The important seaweed products in recent years have been the polysaccharides agar, algin and carrageenin. Because of their unique colloidal properties, these substances are valuable for special purposes and, in spite of their relatively high cost, have been extensively used in food, pharmaceutical, cosmetic and other industries. Agar, for instance, is still the undisputed material for preparing solid bacteriological culture media, and is extensively employed in bakery products, health food and dental impression materials. Algin is most valuable as an ice-cream stabilizer, and is regarded by dairy experts to be as good as, if not better than, gelatin, which was the standard stabilizer until recently. Carrageenin is now the standard suspending agent for cocoa particles in chocolate milk preparations. To the unique properties of these seaweed colloids the modern seaweed industry in the United States owes its existence and its present prosperity.

The increasing interest in seaweed polysaccharides will certainly lead to more research activities in the studies of these products and their sources from biological, chemical and technological points of view. The progress of such researches, however, will be greatly hindered by the loose and inconsistent, sometimes inaccurate, use of terms applying to these colloidal substances and their sources. It is the firm belief of the writer, therefore, that such confusion should be clarified, and the application of the different terms involved be standardized as far as our present knowledge of the sources and chemistry of these products permits. Whenever necessary, new terms should be introduced.

Phycocolloids. In the early days, when agar was the only commercial seaweed colloid, it was variously called "vegetable isinglass" or "vegetable gelatin." These terms have not been used in recent years, since it is well known that agar and the other commercial seaweed colloids are all polysaccharides and are not nitrogenous compounds as they were first thought to be. At present, they are commonly called "seaweed gums" and "seaweed mucilages" by various authors.

Gums occur chiefly as exudations on the barks of trees or on fruits, and mucilages are commonly extracted from seeds. Seaweed colloids, however, are chiefly cell wall constituents, comparable to the pectins of the land plants. They should therefore not be grouped with the gums and mucilages, in spite of certain resemblances, since they differ from the land plant products not only in origin, but also in chemical nature.

Phylogenetically, the primitive marine plants known as the brown and the red algae, from which we derive our agar, algin and carrageenin, are not directly related to the modern highly evolved land plants. There are consequently numerous differences between the plants of the sea and those on the land in their chemical constitution. For instance, it has been well established in recent years that chlorophyll *b* is not present in the brown and the red algae, which have, instead, chlorophyll *c* and *d*, respectively.[5] The kinds of reserve food of these seaweeds are also different from those of the land plants. The seaweed polysaccharides are consequently expected to be different chemically from their counterparts in the land plants. This is quite true, so far as our limited knowledge of their chemistry is concerned. The gums and mucilages are polyuronides consisting of several types of much branched monosaccharide units. The seaweed colloids, on the other hand, consist of generally one or two types of simple monosaccharide units, and with the exception of alginic acid, are not polyuronide. Although alginic acid resembles the gums in being a polyuronide, it is quite unique in being composed exclusively of mannuronic units.

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[4] V. B. Scheffer, *Fish. Mark. News,* 5(6): 1, 1943.

[5] H. H. Strain and W. M. Manning, *Jour. Biol. Chem.,* 144: 625, 1942; W. M. Manning and H. H. Strain, *ibid.,* 151: 1, 1943.

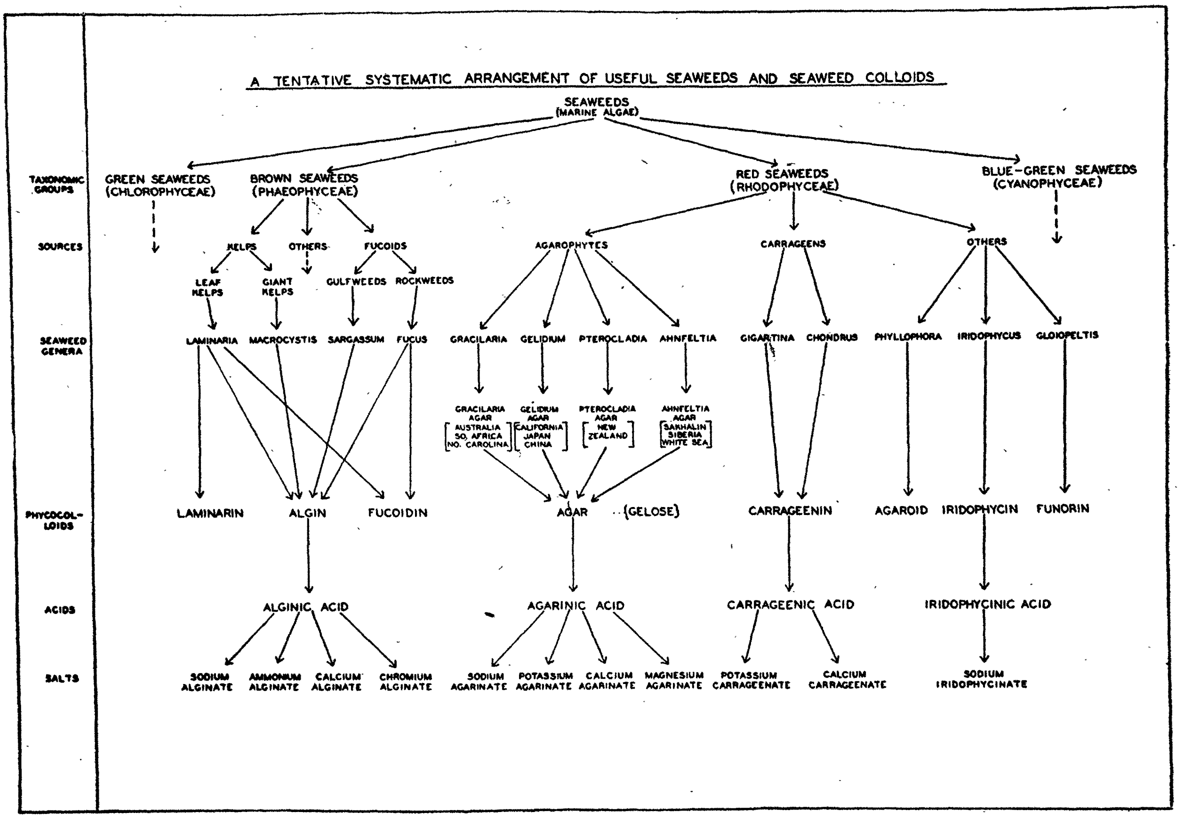

FIG. 1

That the seaweed polysaccharides should be divorced from the gums and mucilages has also been advocated by Norman,[6] who groups these marine products separately under "gel-forming substances." It should be noted, however, that, strictly speaking, not all the commercial seaweed colloids are gel-forming. Moreover, the use of such a complex term is rather inconvenient. The new term "phycocolloid"[7] has therefore been proposed to designate the polysaccharides which are derived from the brown and the red seaweeds and are able to form colloidal systems when dispersed in water.

Gelose. Payen[8] proposed the term "gelose" to designate the refined extract from "mousse de Chine" or "Chinese moss," under which name agar was known when first introduced into France from China in 1856. Later, various authors used the same term to refer to carrageenin. "Gelose" is a well-selected term, and is worthy of being kept to designate collectively the red algal extracts, such as agar and carrageenin, aqueous solutions of which are able to form a more or less firm jelly. It can not be used to include all the seaweed polysaccharides, however, since some of them, the alginates, for instance, are not strictly gel-forming substances.

Agar. The writer[9] has previously pointed out the existing confusion in the usage of the word "agar" or "agar-agar." To avoid using the same word in two or three different meanings, the writer has proposed the word *agarophyte* to refer to the agar-bearing seaweeds and to restrict the use of the word agar exclusively to the dried extract.

Chemically, agar is the sulfuric ester of a linear galactan. It consists of a long chain of *d*-galactopyranose residues, attached by 1:3-glycosidic linkages. This chain is terminated at the reducing end by one residue of *l*-galactopyranose. Attached to the rest of the chain through carbon atom 4, this *l*-galactose residue is esterified at carbon atom 6 with sulfuric acid.[10] There are probably as many as 53 galactose units to each SO_4H group, and at least 140 such units to each nonreducing end group.[11] Our present knowledge of the chemistry of agar is still incomplete. Moreover, with one exception,[11] the previous history

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[6] A. G. Norman, "The Biochemistry of Cellulose, Polyuronides, Lignin," etc., 1937, Oxford.

[7] From Greek φῦκος (seaweed) and κόλλα (glue) + -oid. C. K. Tseng, "Phycocolloids: Useful Seaweed Polysaccharides" in Jerome Alexander, "Colloid Chemistry," vol. VI (in press), Reinhold Publishing Company.

[8] M. Payen, *Compt. rend.*, Paris, 49: 521, 1859.

[9] C. K. Tseng, *Sci. Monthly*, 58: 24, 1944.

[10] W. G. M. Jones and S. Peat, *Jour. Chem. Soc.*, 1942: 225.

[11] V. C. Barry and Thomas Dillon, *Chem. and Ind.*, 1944: 167.

of the agar employed in the chemical studies is not known. While these investigators undoubtedly used Japanese agar, it should be noted that the Japanese product is not a pure agar from *Gelidium Amansii* alone. It probably consists of a mixture of seaweed polysaccharides, since the Japanese use several other red algae, besides the *Gelidium,* in their agar manufacture.

The agar which scientists have been using is principally prepared from species of *Gelidium* in Japan or California. Those which are made in North Carolina, South Africa, Australia, New Zealand, Russia and Sakhalin Island come from other sources. It is, as yet, not definitely known whether the so-called agar from sources other than species of *Gelidium* are the same chemically as the "standard" agar. They do resemble the agar made from *Gelidium* in their physical properties. For instance, they are soluble in hot but not in cold water; they all yield firm gels in dilute aqueous solutions of one or two per cent.; they have low ash contents, generally less than 5 per cent.; and the setting temperatures of their sols, as well as the melting temperature of their gels, are reasonably close to each other. It is proposed, therefore, to classify all of them tentatively as agar, and to designate them as Gelidium-agar (California, Japan and China), Pterocladia-agar (New Zealand), Gracilaria-agar (Australia, South Africa and North Carolina) and Ahnfeltia-agar (Sakhalin Island and White Sea, Russia).[12]

Any attempt at a critical definition of agar is, of course, premature. Nevertheless, a tentative definition will help in the clarification of the conception of agar. The writer has previously proposed such a definition.[9] It is slightly modified and presented as follows: The dried amorphous, gelatin-like, non-nitrogenous extract from *Gelidium* and other agarophytes, being the sulfuric acid ester of a linear galactan, insoluble in cold but soluble in hot water, a one per cent. neutral solution of which sets at 35° to 50° C to a firm gel, melting at 80° to 100° C.

It has been observed by various investigators that when agar is hydrolyzed with an acid, a free "agar-acid" is formed which fails to gel until neutralized by the addition of a base. It is also quite generally accepted that in the natural condition agar probably occurs as the calcium salt, or a mixture of calcium and magnesium salts of this free "agar-acid." Fairbrother and Mastin[13] and Hoffman and Gortner[14] have prepared potassium and other salts of the acid which gel as readily as does the natural agar. For the sake of uniformity of nomenclature, it is here suggested that this "agar-acid" be called agarinic acid, since the term agaric acid has already been used to apply to another totally different compound. Accordingly, the agar as occurring in nature may be regarded as calcium agarinate, and the corresponding magnesium, potassium and sodium salts as magnesium agarinate, potassium agarinate and sodium agarinate, respectively. While these salts are not prepared commercially at present, it is not impossible that they may eventually be made for special uses.

Carrageenin. The name carrageenin was given to the extract from *Chondrus crispus* by Stanford.[15] It was, however, first isolated by Schmidt[16] in 1844. Although carrageenin is a very appropriate term, it has not been well received. Scientists working on this polysaccharide have been calling it under such compound names as "carrageen extract," "*Chondrus* polysaccharides," "Irish moss mucilage," etc. For the sake of simplicity and uniformity in nomenclature, the writer is in favor of using the term carrageenin for this gelose.

Carrageenin resembles agar in a general way, but has a much higher ash content (about 20 per cent.), and requires higher concentrations (3 or more per cent.) to form firm gels. Its gel melts at much lower temperatures (27° to 30° C for a 3 per cent. gel and 40° to 41° C for a 5 per cent. gel).[17] Chemically, carrageenin is similar to agar in being a carbohydrate ethereal sulfate and in containing galactose residues[18] joined by 1:3-linkages. The sulfuric ester group, however, is attached to carbon atom 4, not 6. The galactosidal linkages combining the residues seem to be of the α-type. Like agar, carrageenin is considered to be a cell wall constituent, and is presumed to occur *in situ* as a mixture of sodium, potassium and calcium salts.[19] While the acid is still imperfectly known, it may be conveniently called carrageenic acid. The carrageenin in the natural condition is accordingly a mixture of sodium, potassium and calcium carrageenates.

Funorin. This term has recently been used to designate the polysaccharide of *Gloiopeltis* (funori in Japanese),[20] which gives the gluey extract used for hun-

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[12] This is better known as Sakhalin agar, since it was first extracted from *Ahnfeltia plicata* from Sakhalin Island (or Karafuto) by a Japanese about forty years ago. The seaweed was subsequently found in large quantity in the White Sea region.

[13] F. Fairbrother and H. Mastin, *Jour. Chem. Soc.,* 123: 1412, 1923.

[14] W. F. Hoffman and R. A. Gortner, *Jour. Biol. Chem.,* 65: 371, 1925.

[15] E. C. C. Stanford, *Jour. Soc. Arts,* 10: 185, 1862.

[16] Carl Schmidt, *Annal. Chem. und Pharm.,* 51: 29, 1844.

[17] Paul Haas and T. G. Hill, *Ann. Appl. Biol.,* 7: 352, 1921.

[18] E. G. Young and F. A. H. Rice have recently reported in *Jour. Biol. Chem.,* 156 (2): 781, 1945, the isolation in considerable quantity of 2-ketogluconic acid as a constituent of carrageenin, in addition to the galactose residues.

[19] J. Buchanan, E. E. Percival and E. G. V. Percival, *Jour. Chem. Soc.,* 1943: 51.

[20] C. K. Tseng, *Sci. Monthly,* 59: 37, 1944.

dreds of years in the Orient as a textile-sizing material. Its exact chemical nature is still unknown.

Iridophycin. From a California red alga, *Iridophycus flaccidum*,[21] Hassid has isolated a galactan ethereal sulfate, which upon hydrolysis gives only galactose.[22] The name *iridophycin* is tentatively given to this polysaccharide for the sake of convenience and uniformity. It contains over 25 per cent. of ash which consists chiefly of sodium sulfate. It is presumed to be of the 1:4 normal pyranose type of constitution, with the sixth carbon atom bearing the ethereal sulfate group. The free acid has been prepared by electrodialysis, and may be termed iridophycinic acid. The colloid occurs *in situ* principally as its sodium salt.

Agaroid. From Odessa in the Black Sea region, the Russians have extracted from the red seaweed *Phyllophora*,[23] a substance which was first regarded as a kind of agar. Later, it was named agaroid[24] because of certain differences between it and agar. Its exact chemical nature is still not known.

Algin. Stanford[25] reported in 1883 that by digesting kelps with sodium carbonate solution, a glutinous mass was obtained which, when evaporated to dryness, presented "an appearance which is not unlike gum tragacanth." To this new substance Stanford gave the name "algin," derived from the word alga. This term has since been variously used by its discoverer and others to apply to: (1) sodium salt of the substance; (2) natural state of this substance, which according to some, may be the calcium salt; (3) alginic acid; (4) soluble alginates, and (5) alginic acid and its salts in general. As it is used in commerce to-day, "algin" refers to the soluble alginates, especially sodium alginate and, to a lesser extent, ammonium alginate. To avoid confusion, the term algin is better restricted to sodium alginate, which is the primary product in all the processes of manufacture and from which other alginates are derived. Since "agar" and "carrageenin" have been accepted to designate respectively the water-extracted colloids from agarophytes and carrageens, it is logical to limit the use of "algin" to the soda-extracted substance, *i.e.*, sodium alginate, from kelps. The other alginous products may be designated as ammonium algin, calcium algin, chromium algin, etc. From the limited information we have at present, it seems that algin from different sources belongs to the same chemical entity.

Alginic acid is a polyuronic acid with the empirical formula $(C_6H_8O_6)_n$. It is composed entirely of *d*-mannuronic anhydride residues,[26] in which the hydroxyl groups are attached to carbon atoms 2 and 3, while bridge and ring are attached to carbon atoms 4 and 5. Although evidence is not yet sufficient to decide between pyranose and furanose structures, the former is more probable, in view of the large negative rotation of alginic acid and its resistance towards hydrolysis. Thus, the mannuronic acid units are linked in such a way that the carboxyl group on each unit is free to react, while the aldehyde group is shielded by linkage.[27] Like agar and carrageenin, algin is presumed to occur in nature as a cell wall constituent, probably existing as a mixture of the acid itself and its calcium, magnesium and alkali metal salts.

Laminarin. Although laminarin was discovered only two years later than algin, being isolated by Schmiedeberg in 1885, it has not yet found commercial applications. Laminarin is the reserve carbohydrate of the kelps, analogous to, but different in structure and properties from, starch of the land plants. It consists exclusively of glucose residues, with the empirical formula of $(C_6H_{10}O_5)_n$. There are probably 16 glucose units, which are combined by 1:3-glycosidic linkages. The linkages in laminarin are regarded to be of the β-type.[28]

Fucoidin. In 1913 Kylin[29] reported that he had extracted from *Laminaria* and *Fucus* a new polysaccharide which he called "fucoidin." A similar water-soluble carbohydrate was isolated from the giant kelp, *Macrocystis pyrifera*, by Hoagland and Lieb[30] in 1915. Fucoidin is presumably a carbohydrate ethereal sulfate, occurring in nature as a calcium salt. On hydrolysis, it gives rise to pentoses and methyl-pentoses, particularly fucose. Like laminarin, fucoidin has not yet found commercial uses.

Classification of phycocolloids. In view of our incomplete knowledge of their chemistry, it is still too early to propose a critical classification of seaweed colloids. It may be said, however, that there seem to be three groups of the phycocolloids. First of all, we

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[21] Erroneously called *Iridaea* (*Iridea*) *laminarioides* by various authors; *cf.* G. M. Smith, "Marine Algae of the Monterey Peninsula," p. 288, 1944, Stanford.

[22] W. Z. Hassid, *Jour. Am. Chem. Soc.*, 55: 4163, 1933; *ibid.*, 57: 2046, 1935; *Plant Physiol.*, 11: 461, 1936.

[23] V. L. Elin, R. Grayerova and G. Fisher, *Bull. Nauch. Issledovatel Khim-Farm. Inst.*, 1931: 140; A. Korentzvit, *Khim-Farm. Prom.*, 1934(4): 36.

[24] V. Gryuner and N. Tauson, *Colloid Jour.* (*U.S.S.R.*), 2: 783, 1936; V. Gryuner and L. Veronyan, *ibid.*, 5: 851, 1939.

[25] E. C. C. Stanford, *Chem. News*, 47: 254, 1883.

[26] V. C. Barry and T. Dillon, in *Sci. Proc. Roy. Dublin Soc.*, 21: 285, 1936, expressed their conviction that the polymerizing unit in alginic acid is the complete uronic acid and not the anhydride, and hence the empirical formula for alginic acid should be $(C_6H_8O_6 \cdot H_2O)_n$.

[27] E. L. Hirst, J. K. N. Jones and W. O. Jones, *Jour. Chem. Soc.*, 1939: 1880.

[28] V. C. Barry, *Sci. Proc. Roy. Dublin Soc.*, 21: 615, 1938.

[29] Harold Kylin, *Z. Physiol. Chem.*, 83: 171, 1913.

[30] D. R. Hoagland and L. L. Lieb, *Jour. Biol. Chem.*, 23(1): 287, 1915.

have the water-soluble ethereal sulfates as represented by agar, carrageenin and fucoidin; they are similar to the mucilages in some of their properties. Secondly, there are the water-soluble reserve carbohydrates consisting exclusively of glucose units; they are represented by laminarin and occupy a position similar to that of starch in the land plants. In the third group we have the alkali-soluble polyuronides, represented by algin, which are analogous to pectin. The accompanying diagram summarizes the writer's view of the sources of these interesting seaweed colloids, or phycocolloids, and their interrelationship. It is to be noted that there are as yet no commercial colloids extracted from the other two seaweed groups, the green algae (Chlorophyceae) and the blue-greens (Cyanophyceae).

OBITUARY

MILO HERRICK SPAULDING

HERRICK SPAULDING was born on June 10, 1879, at Rutland, Vermont. He died on Easter Sunday, April 1, 1945, after a long illness. At the time of his death he was professor of zoology at Montana State College, where he had taught for thirty-seven years. He was a member of Sigma Xi, Phi Kappa Phi and Phi Sigma as well as many of the technical societies in his two main fields of mammalian anatomy and embryology. He was graduated from Stanford University in 1903 and received the M.A. degree in 1906 from the same institution.

Herrick Spaulding was one of the real old-time zoologists—painstaking in detail, patient and a never-ending source of help and encouragement. Not only his colleagues but two generations of students will remember him, as a teacher, for longer than we remember most.

CLARENCE ARCHER TRYON, JR.
MONTANA STATE COLLEGE

RECENT DEATHS

DR. WILLIAM WORTHINGTON HERRICK, president of the New York Academy of Medicine, died on June 1 at the age of sixty-six years.

DR. MERTON BENWAY WAITE, principal plant pathologist at the station at Beltsville, Md., of the U. S. Department of Agriculture, with which he had been associated for more than fifty years, died on June 6 at the age of eighty years.

CURTIS J. HESSE, curator of the museum of the Agricultural and Mechanical College of Texas, died on May 8. Mr. Hesse was a vertebrate paleontologist who had specialized in the vertebrate faunas of the later Tertiary beds of the Gulf Coast Plain.

Nature reports the death on May 7 of Dr. K. J. W. Craik, director of the unit of applied psychology of the Medical Research Council, at the age of thirty-one years, and of Thomas J. Nolan, since 1932 professor of chemistry at University College, Dublin, on March 12 at the age of fifty-six years.

SCIENTIFIC EVENTS

THE INTERNATIONAL COMMISSION ON ZOOLOGICAL NOMENCLATURE

THE International Commission on Zoological Nomenclature announces its intention of publishing at an early date a revised and up-to-date edition (1) of the International Code of Zoological Nomenclature and (2) of the Official List of Generic Names in Zoology. The following is a statement of the plans:

THE INTERNATIONAL CODE OF ZOOLOGICAL NOMENCLATURE

The last edition of the English text was published some years before the war and is now out of date for various reasons, including the adoption by the International Congress of Zoology of changes in some of the articles in the International Code. The International Commission has, therefore, arranged for the early publication of a revised and up-to-date edition consisting of the substantive French text (on left-hand pages) and the English translation (on right-hand pages). The volume, which will be fully indexed, will also contain a detailed analysis of all the "Opinions" so far rendered by the International Commission in regard to the interpretation of the provision of the code.

THE OFFICIAL LIST OF GENERIC NAMES IN ZOOLOGY

The "Official List" was established by the International Congress of Zoology at its meeting at Monaco in 1913 for the purpose of recording full particulars relating to the names of the 5,000 to 10,000 best-known and most important genera in the Animal Kingdom with their type species. So far, however, only about 700 names have been placed on the "Official List." This is largely due to the fact that the decisions taken by the International Commission have never hitherto been brought together in a single volume and with a full index.