CHAPTER XIV

Disinfection

Disinfection, or germ-killing, first more or less incidentally and with vague or no ideas as to the how and why, then with intent and under scientific control, has been practiced for milleniums. Broadly, the agents employed have been heat, copper, silver, chlorine, ozone and ultraviolet rays. The greatest of these is chlorine.

Boiling to improve the quality of water was probably employed from the beginning of civilization. The earliest written mention of boiling that has been found is in Herodotus (484(?)-425 B.C.) (1) but the passage refers to Cyrus the Great, who lived a century before him:

The Great King, when he goes to the wars, is always supplied with provisions carefully prepared at home, and with cattle of his own. Water too from the river Choaspes, which flows by Susa, is taken with him for his drink, as that is the only water which the Kings of Persia taste. Wherever he travels, he is attended by a number of four-wheeled cars drawn by mules, in which Choaspes water, ready boiled for use, and stored in flagons of silver, is moved with him from place to place.

Hippocrates, the father of medicine (460-359(?) B.C.) (2) declared that boiling and straining rain waters was necessary to prevent them from having a "bad smell" and causing "hoarseness and thickness of voice to those who drink them." Aristotle (384-322 B.C.) is said to have advised Alexander the Great (356-323 B.C.):

Do not let your men drink out of stagnant pools. Athenians, city born, know no better. And when you carry water on the desert marches it should first be boiled to prevent its getting sour.⁺

Much currency has been given to alleged passages from the Sanskrit advising that water be treated by boiling and plunging hot metal into it. Place, writing from India in 1905 (3), cites two medical maxims

• This statement by Herodotus is echoed by various writers (Plutarch, de Exil., Vol. II, p. 601 D; Athenaeus, Deipnosoph., II: 23, p. 171; Solinus, Polyhist., XLI, p. 83; Eustath, ad Dionys. Perig., 1073, etc.). The water under consideration is said at the present day to be excellent, and the natives vaunt the superiority of these two rivers over all other streams or springs in the world (Jour. Geog. Soc., Vol. IX, Part I, p. 89).-From footnote in Rawlinson's translation of Herodotus (1).

 \dagger Search of Aristotle's works, of the lives of Alexander by Arrian and by Quintus Curtius Rufus, disclosed no such statement. Inquiries of specialists in Aristotle have brought assurances that his works contain no such passage.-M.N.B.



from the Sanskrit, of about 2,000 B.C., one advising that water be exposed to sunlight and filtered through charcoal; the other directing that "foul water" be treated by boiling and "by dipping seven times into it a piece of hot copper," and then filtering it. Sus'ruta Samhita, attributed to the fourth century A.D. (4), reiterates these directions and specifies that filtration be through sand and coarse gravel (see Chap. I).

Like Cyrus the Great of Persia, centuries earlier, the Roman Emperor Nero (reigned 54-68 A.D.) had a predilection for boiled water but as befitted a more luxurious age he had it cooled. Pliny (5) states, "It was the Emperor Nero's invention to boil water, and then enclose it in glass vessels and cool it in the snow. . . . Indeed, it is generally admitted all water is more wholesome boiled."

Pliny (5), expressing his own opinion about 77 A.D., wrote, "The best correction of unwholesome water is to boil it down one-half." Plutarch (6), a few years after Pliny, mentions boiling and cooling of water somewhat incidentally, with an interesting reference to heat transference. He says:

All water, when it hath been once hot, is afterwards more cold; as that which is prepared for Kings, when it hath boiled a good while on the fire, is afterwards put into a vessel set round with snow, and so made cooler; just as we find our bodies more cool, after we have bathed, because the body, after a short relaxation from the heat, is rarefied and made porous. . .

Corroborating the foregoing quotation is the following statement by Ellen C. Semple, in an article on ancient water works in Mediterranean lands: "The Roman plutocrats had their water first boiled, then chilled by mountain snow; if the snow was not clear, it was strained through fine linen cloths" (7).

All these references are to water boiled for kings or plutocrats. If a cat could look at a king so might the common people, even though poor, boil their drinking water—provided they thought it worth while to gather a few sticks for that purpose or to put a vessel of water over their charcoal braziers. No specific evidence that they did so has been found, but from the time that the Chinese began to drink no water except that boiled to infuse tea they, knowingly or not, had provided themselves with a large measure of protection from the water-borne diseases.

Paulus Aegineta, in his compendium of medical lore written late in the seventh century A.D. (8), says that "waters which contain organic

322

impurities, [and] have a fetid smell or any bad quality may be so improved by boiling [or by mixing with wine] as to be fit to be drunk." In Adams' commentary on Aegineta, it is noted that Rhazes [or Rasis, an important Mohammedan physician of the ninth century] directed that water drawn from a deep well be boiled before use. Avicenna, an early eleventh century Persian physician (9), discusses boiling and distillation with apparently conflicting opinion as to which was preferable. His ideas on why either should be used are somewhat vague.

Passing over a long blank period, we come to Boerhaave (10), the noted Dutch chemist (1668–1738). Writing of "putrid water," he says, "But when this Water has thus spontaneously grown putrid, it may easily be rendered wholesome again, and may be drank without being offensive; for if you give it only one boil on the Fire, the Animals that are in it will be destroyed, which, with the rest of the impurities, will subside to the bottom"-but, as a finishing process, he advises adding a small amount of acid. Experience under the equator, he states, "where the Waters putrify horribly, and breed such quantity of insects, and yet must be drank," had proved this treatment to be effective.

In what appears to be the first American book on public health (1835), Dunglinson (11) says, "Whenever water is unusually contaminated, it may be boiled, filtered and agitated. . . . There are many valetudinarians, and some whole nations—as the Chinese—who never drink water that has not been boiled."

With the knowledge and acceptance of the fact that drinking water is a vehicle for spreading typhoid and cholera, together with the widespread use of filtration, private means of treating water for domestic consumption became less necessary. There was still a considerable demand for the use of heat to disinfect water before the advent of chlorination. Boiling was too costly and complicated for public use but became more feasible for special cases when the principles and practice of heat transference were developed. According to Samuel Rideal (12), sterilization by heat was first practically applied about 1888 by Charles Herscher, while other means, such as the Vaillard-Desmoraux apparatus and, in the United States, the Waterhouse-Forbes apparatus were introduced later. At Brest in 1892, say Samuel and Erik Rideal (13), there was a test of heat sterilization apparatus, supplied by Rouart-Herscher & Co., in which water was not exposed to contamination and heat interchange was employed. They also mention the Forbes apparatus in which water boils only a few seconds and so "retains most of the original gas and taste (U.S. Patent, December 13, 1898)."

M. Bechmann (14), in 1904, while chief of the water service of Paris, stated that "a single city, Parthenay [France, with] 7,500 inhabitants, has sterilized its water by heat, treating 3 cu.m. (793 gal.) per hour with Rouart sterilizers." Imbeaux's *Annuaire* (15) states that this installation was abandoned on the introduction of water from springs in 1905, shortly after the date of Bechmann's paper. Imbeaux's data show that the capital and operating cost for the Rouart installation were very high. He does not say whether the sterilizing equipment was put in when the river supply was introduced in 1895. After 1905, he states, Parthenay had a double system of water supply, including pumps and distribution system. No other example of heat sterilization for a municipal water supply has yet been found but in the discussion of distillation (see Chap. XV) two small plants in Texas will be noted.

Copper and Its Compounds

Abundance of copper in some parts of the world and the ease with which the metal can be worked have led to its wide use for water containers and cooking utensils. This use has extended through thousands of years in India and other Eastern countries. The passages from Place already quoted here and at more length in Chapter I have been used by several later writers to indicate that copper was regarded as a disinfecting agent centuries ago in India. These have included Professor J. J. Hinman Jr. of the University of Iowa (16), Samuel and Erik Rideal of London (13), Lt.-Col. C. H. H. Harold of the Metropolitan Water Board, London (17), Rideal and Baines (18), and Henry Kraemer (19). Hinman wrote in 1918: "It seems that colloidal copper is given off into water that is kept in a bright copper vessel." Kraemer in 1905 gave 28 references to papers on copper as a germicide. From these papers and from his own experiments he concluded that copper in small quantities is harmless to human beings. Apparently he did not know of the fanatical campaign against the use of copper vessels as filter containers and for cooking utensils conducted by Amy at Paris in the 1750's (20) (see Chap. IV).

After weighing all the citations I have examined, I have concluded that, although under some conditions the use of copper vessels may

have had germicidal value, they were not used for that reason, but rather because copper was locally available, easily worked into desired shapes and could be easily cleaned and polished.

Concern over using copper in any form as a bactericide subsided soon after the epochal Moore-Kellerman studies on copper sulfate both as a germicide and as an algaecide (21, 22). Papers by D. D. Jackson (23) and a symposium (24), both given in 1905, dealing with copper sulfate and water supplies, describe experiments and opinions on copper sulfate as a bactericide. The experiments on "copper-iron sulfate" as a disinfectant which were made at Anderson, Ind., by C. Arthur Brown, were described by him in a paper presented in 1905 (25) (see also Chap. XVII).

Silver

Silver in minute quantities will destroy some macro- and microorganisms in water. Attempts to devise a practicable method of utilizing this agent to disinfect water have resulted in a few installations for special purposes where proprietors were concerned with avoiding the real or imagined objections to chlorination, or were enticed by the claims of a new magic process, in either case with little regard to cost. Many examples of the application of silver to swimming pools but few to city water supplies have been found.

Forerunner of many investigators of silver as a destroyer of waterborne organisms was Carl von Nageli, a Swiss botanist. In 1880 he observed the disappearance of algae, particularly *Spirogyrae*, from water containing minute quantities of copper or silver or their salts. The only paper on the subject recorded as published by him appeared in 1893 (26). Nageli named the action of silver "oligodynamic" or "forces of triffes." The field seems to have lain fallow until 1899 (27) and again until 1917 (28). After that many papers appeared up to 1935.

The first paper in English appeared in 1932, at London (29). All but one of the papers up to that time were printed in German; the exception was in Russian. In 1934–36 four or five papers on silver treatment were published in the United States. Two of these, although written in English, originated abroad: one was by S. V. Moiseev, of the Leningrad Branch of the Union of Scientific Research of the Institute of Water Supply and Sanitary Engineering (30), and one by Just and Szniolis, Assistant and Chief Engineers of the Department of Sanitary Engineering of the State School of Hygiene, Warsaw (31). Each of these papers reviewed the subject up to its time and listed authorities—about 25, excluding duplicates. Moiseev gave an account of laboratory experiments with silver-coated sand, conducted by him on water from the River Neva in 1930-31. Just and Szniolis described their own laboratory experiments, started at Warsaw in 1929, "with filters containing metallic silver, with solutions of silver salts and with electrocatadynization."

Between the dates of publication of the two papers just noted one appeared in New York. It dealt chiefly with silver treatment as developed by Krause in Germany. It named several small installations in that country, including one for drinking water, and also one in this country at a swimming pool in Washington, D.C. (32). In 1936, Shapiro and Hale published results obtained in both laboratory and swimming pool tests of the Katadyn Process (electrolytic method of treating water with silver) made in New York City with the cooperation of American representatives of Katadyn, Inc., in 1934–35. These authors reported (33) a number of reasons for their conclusions "that in the present state of development the Katadyn Process cannot be approved for use in swimming pools."

Several successful instances of the use of silver disinfection of swimming pools were cited and some test results reported in a paper by J. H. Dorroh, of the Department of Civil Engineering at the University of New Mexico (34), in 1936. Fifteen swimming pools in England had been equipped with silver treatment apparatus in 1934. One, for a swimming pool at the Congressional Country Club, Washington, was put into use on July 4, 1935.

Conclusions regarding disinfection by silver compounds, in the American Water Works Association's Manual of Water Quality and Treatment (35), are that "the silver process has not proved to be equal to other methods" and that "its use is not justified for public water supplies."

Chlorine

Nothing in the field of water purification came into use as rapidly and as widely, once it got a good start, as chlorination. Its impetus sprang from its adoption on a large scale in 1908 at the Boonton Reservoir of the Jersey City Water Works. It had been used before on small water supplies in the United States and abroad, in most cases

experimentally. Still earlier, it had been used on sewage, also in America and Europe, but tentatively.**

The earliest proposals to chlorinate water were made before there was knowledge of water-carried disease germs. First of these found on record is a statement by Dr. Robley Dunglinson in his *Human Health* (11), published in 1835 at Philadelphia.] To make "the water of marshes potable," he says, "it has been proposed to add a small quantity of chlorine, or one of the chlorides; but a quantity sufficient to destroy the foulness of the fluid can hardly fail to communicate a taste and smell, disagreeable to most individuals." When and by whom this proposal was made the Scotch-American physician did not say. His recognition of taste-and-odor difficulties arising from chlorination is significant in view of later experiences with the use of minute quantities of chlorine compared with the large amount he had in mind.

Patented Processes.—During the last 60 years of the nineteenth century there were granted fifteen British patents on water treatment by voltaic action, magnetic action, electric currents or the addition of a chemical oxidizing agent. Summaries of British patents on a wide range of methods of water treatment are given in a series of pamphlets (43). The earliest of these on chlorination and related processes are here outlined:

Voltaic action between unnamed filter media was claimed at least as early as 1839 for the Royal Patent Filters of George Robins. [Ure's Dictionary of Chemistry, 1839; Encyclopedia Britannica, 1842. The Britannica questioned the voltaic action but gave space to exterior views of household filters.]

Pocock took out a patent November 27, 1852, which called for precipitation by a "salt," followed by the addition of a "hypochloride," then filtration through charcoal.

Harrison patented November 16, 1863, a magnet adapted to filters of asbestos, talc, sponge or carbon, to polarize water.

Kühne, August 16, 1866, patented a method of disinfection and purification by such oxydizing substances as "chlorine-permanganates."

• Chronologically arranged references and other data on the discovery and early manufacture of chlorine, its adoption for bleaching in the late eighteenth and early nineteenth centuries, trials of the application of chloride of lime to London sewage in 1854 and 1884 and the promotion of electrolytic methods of sewage treatment by Webster in England, Hermite in England and France, and Woolf in the United States are found in Race (36), Hooker (37), and an A.P.H.A. Committee Report (38). A comprehensive exposition of the Webster process, by Webster himself, is given in Crimp (39) and abstracted at considerable length by Fuller (40). See also Metcalf and Eddy (41) and my summary of 1912 (42).-M.N.B.

Davis's patent, August 29, 1866, called for destruction of "infusoria and fungi together with animalculi and other insects" by the use of "caustic alkali"; then filtration through sand, gravel or vegetable charcoal to render the water "bright and limpid" (see also Chap. XVIII).

Pope and Sawyer, on May 7, 1873, patented the idea of passing electric current through metallic wire arranged in horizontal coils in water, thus precipitating any substance in solution.

Webster, January 27, 1887, patented "methods of purifying sewage and other impure liquids by electrolysis, applicable also in connection with filters, for purifying potable waters."

Destruction of dangerous germs by passing an electric current through an electrode immersed in water, it may be interjected here, was briefly described in 1874 by a French journal, *La Nature*, which credited its information to a statement by Dr. Dobell in the *Times* [London] and said that the same idea had been conceived by Dr. Stephen Emmons. The article characterized Dr. Dobell's process as a "new application of electricity" whereby cholera and typhoid germs would be destroyed by nascent oxygen. Whether the process had been covered by patents and, if so, when and where the patents had been granted the article does not say.

In the United States, from 1887 to 1898, a half dozen patents on water treatment by electrolysis were granted. Webster's British patent of January 27, 1887, may be considered the forerunner if not the model of later British and American patents on the use of electrodegenerated electric current for purifying sewage and water. Although water treatment was claimed in the Webster patent, no record of such use of the process has been found. Nor does it appear that any process for the direct application of electricity to water has ever been permanently adopted. As will be shown, none of the electrolytic processes, whether proposed for sewage or water, does more than produce a chlorinating or oxidizing agent that can be applied to water or sewage.

The first American patent on chlorination of water was granted to Albert R. Leeds, Professor of Chemistry at Stevens Institute of Technology, Hoboken, N.J., on May 22, 1888. His application, however, was filed in mid-September 1887 or nine months after Webster's patent.* Leeds applied for a process patent on September 15, 1887, and for an apparatus patent four days later. His process application

[•] Ahead of Leeds, but for treating sewage, was the American patent of J. J. Powers, granted May 10, 1887. His claims were for apparatus generating gaseous chlorine from manganese dioxide, sodium chloride and sulfuric acid. He built six sewage works, all in New York State, of which four were within the present limits of New York City (44).

seems to have been denied-perhaps because of anticipation by Webster. He was granted a British patent, apparently on apparatus only, on May 22, 1888, the date of his American patent. Three paragraphs from Leeds' American patent specifications, which demonstrate his conception of how water purification may be induced by an electrical current, are the following:

In the art of purifying water it has been found that many waters contain certain organic impurities, which it is highly important should be removed in order to render the water fit for drinking and many other uses, but which cannot be removed by ordinary mechanical filtering. In the treatment of waters containing impurities of this class many attempts have been made to purify the water by the use of chemicals, which acted either to precipitate the organic impurities or to reduce them to such condition that they could be readily removed by filtration.

I have discovered that the organic impurities which are contained in large quantities in many waters when in their natural condition, as well as in factory slop and sewage, can be readily and economically removed, so as to render the water pure and wholesome, by treating the water with the gases obtained by the decomposition of water containing an acid or salt in solution, the decomposition being effected by means of an electric current. The acid employed may be hydrochloric, nitric, phosphoric, chromic, or sulfuric; or the salts of these acids may be employed, or a mixture of these acids or salts or acids and salts may be employed. The best results are, however, obtained by the use of hydrochloric acid.

The present invention relates particularly to an apparatus for effecting the purification of water by means of gases generated as above stated. . . .

Leeds' apparatus was, in substance, a tank containing an acid solution; electrodes in the tank, through which an electric current passed; pipes leading from the solution tank and the raw-water tank or filter to a contact chamber; and a filter or a final tank to receive the electrically treated water. That is, the water could be filtered before or after it had been subjected to electric action, or it could be doubly filtered. The gases of electric decomposition "will have the effect," the specifications stated, of destroying "the organic impurities contained in the water, so that they will be precipitated or reduced . . . [so] that they can be readily removed by passing the water through an ordinary filter."

Electrodes combined with a mechanical filter were patented in Great Britain and the United States by Omar H. Jewell of Chicago in 1888 (British patent, February 7, 1888; American patent, July 10, 1888). Application for an American process patent was filed December 7,

329

1887, in the names of Omar H. and William M. Jewell * (father and son) but was not granted. Application for an apparatus patent was made ten days later by the father only. The applications were filed three months after the Leeds' applications and nearly a year after the date of the British patent to Webster.

The electrolytic portion of the Jewell apparatus consisted of electrodes set vertically in the dome of a mechanical filter. Current was supplied from a battery or other generator. It was assumed that carbonic acid gas and common salt would be used to form "an insoluble bicarbonate of soda, which is precipitated or caught by the filter material below, through which the water subsequently passes. Pure fresh water will be discharged" from the filter. For process claims the specification refers to the application for a process patent, which was not granted.

Laboratory experiments on passing low-voltage current through water were reported early in 1891 by R. Mead Bache. Although the results were not convincing, he suggested further study on the Philadelphia water supply (45).

Early Application of Electrolysis.—The first use of electrolysis in the field was in the Croton gathering ground of the New York City supply. There, on July 1, 1893, electrozone produced from salt brine by apparatus devised and installed by Albert E. Woolf was added to the sewage of the village of Brewster before it was discharged through perforated pipe into the East Branch of the Croton River. A pipe from the main electrozone conduit led to a second pipe discharging into Tonetta Brook about 500 ft. above its junction with the Croton River. The Brewster electrozone plant continued in operation until it was destroyed by fire in 1911. Subsequently bleaching powder was used.

To determine the value of electrozone in purifying Croton water and sewage, a laboratory was installed by Edward A. Martin, Chemist in the Department of Health, New York City. His investigations were begun in August 1893 and continued several months. In a re-

• The younger Jewell, who graduated from the University of Illinois School of Pharmacy in 1887, was chemical engineer of the Jewell Pure Water Co., to which the patent in question was assigned, and it may be assumed that the germ of the patent was his conception. Ten years later a different form of electrical apparatus, devised by William M. Jewell, was given a brief test at the Louisville Filtration Experiment Station, and a little afterward he applied chlorine to the effluent of a demonstration filter at Adrian, Mich.

port made public early in 1894, Martin stated that the addition of electrozone in suitable quantities rendered water sterile, induced sedimentation, removed taste and odor due to decomposition of organic matter in stored water and tended to decolorize water coming from peaty bogs. The report gave many and various data on the tests (46).*

Bleaching Powder.—Chlorination with bleaching powder (chlorine de chaux) was invented by Traube in 1894, states Dr. Edouard Imbeaux (48). In 1896, a typhoid epidemic at Pola on the Adriatic Sea (one-time chief naval station of Austria-Hungary) was stopped by the use of bleaching powder. The excess of chlorine was neutralized by sodium sulfite.

Professor Drown's Dicta.—"The so-called electrical purification of water by treating it with an electrolyzed solution of salt is thus seen to be simply a process of disinfection by sodium hypochlorite; electricity, as such, has nothing to do with it. There is nothing peculiar in the sodium hypochlorite produced by electrolysis. . . ." Thus declared Professor Thomas M. Drown in 1894, after having reviewed the subject (49).

So far, so good! Then came a question and answer strange enough in the light of subsequent events:

Finally, is it desirable in any case to treat a city's water supply with a powerful disinfectant like the hypochlorites? When the question is put in this bald way I cannot think it will receive the approval of engineers and sanitarians . . . in cases where a water supply has got into such a hopelessly bad condition that nothing will render it safe but disinfection by chloride of soda or chloride of lime, it is high time, I think, to abandon the supply, and in this opinion I feel sure most water works engineers will coincide.

In his advocacy of naturally pure water and his zeal against humbug exploitation of the magic powers of electricity, Drown did not foresee the vast legitimate field of water chlorination. He did point the way to the early mass production of sodium hypochlorite by the

• Immediately after the Brewster plant was put into use, the inventor escorted a delegation of New York City officials, including Hugh J. Grant, then Mayor, on a tour of inspection. I went with the party. We saw tanks of salt brine in which electrodes were immersed and the treated brine which was applied to sewage and to a stream, as stated in the accompanying text. The Mexican government sent an engineer to New York to investigate electrozone. He made a favorable report (47) but said that the "Woolf system" was nothing more than the one used by M. H. Hermite at Rouen in 1889.-M.N.B.

THE QUEST FOR PURE WATER

use of the new electric process then on trial, which was to make "bleach" available to water works at a low price. He could not have been expected to visualize that the powder would soon be superseded by the less costly and more convenient liquid chlorine and the ingenious apparatus for its accurate application to water at any desired rate. Like Dunglinson 60 years earlier, Drown did not foresee that minute quantities of chlorine would be sufficient.

Ciemens Herschel, in discussing Drown's paper, deprecated Drown's slighting remarks on chlorination as discouraging to new processes, but he did not advocate chlorination. John C. Chase agreed with Herschel's plea against putting a brake on the development of new processes but said that he understood from Drown's paper "that, as a practical matter, purification of water by electricity is a humbug." And so it was, but in fairness to the long line of inventors and promoters of electrolytic treatment of both water and sewage it should be pointed out that most of them did not claim the process involved direct electrocution of bacteria.

French and English Skeptics.—Drown was not the only eminent scientist to be skeptical of what have later become revolutionary changes in water treatment. In France, Arago condemned the addition of alum to water a half century before the official Massachusetts taboo was placed on coagulation. In England, Edward Frankland testified in the late 1860's that although Clark's water softening process was "beautiful" it was impracticable—although soon afterwards he endorsed it (50). J. A. Wanklyn, sticking to his theory that dead organic matter rather than living organisms caused the spread of cholera and typhoid, "waxed sarcastic" over the botanical gardens of the bacteriologists (51, 52).

Chlorination and Electrolysis at Louisville, Ky.-William M. Jewell applied chlorine gas in January 1896 to the effluent from the Jewell rapid filter at the Louisville, Ky., testing station. "I set up this chlorine equipment on my own initiative," wrote Jewell to me in 1933 (53), "as we were not getting 97 per cent bacterial reduction in spite of all the coagulant we could use, even overrunning the alkalinity at that time." After the equipment "had been tested for about a week or two," Jewell states, he received a letter from Charles A. Hermany, President of the Louisville Water Co., "requesting the discontinuance of chlorine, on the ground that it was detrimental to the whole pro-

ceedings, would never come into general use and was unfair to our competitors in the test. Of course I acceded but protested their views."

The accompanying sketch shows one of the twelve chlorine gas generators used at Louisville. It was sent to me by Jewell in 1933 and is here published for the first time. The 12 U-tubes, wrote Jewell,

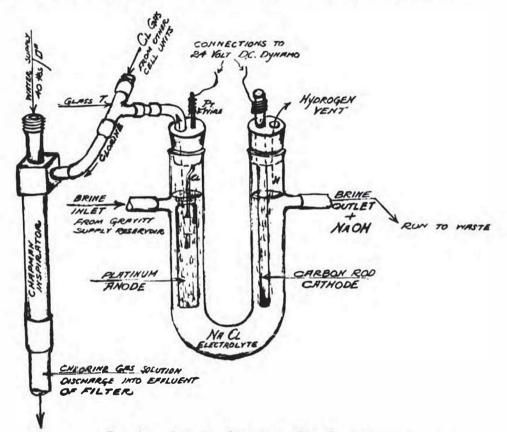


FIG. 64. JEWELL CHLORINE GAS GENERATOR

One of twelve generators installed by William M. Jewell for trial at Louisville Filter Testing Station in 1896; it was operated about ten days, applying chlorine at the rate of 0.25 ppm. to effluent from the Jewell filter (Reproduction of sketch made by William M. Jewell)

-----were connected to a gas header leading to the water inspirator which drew the gas from the cells and delivered it into the open gravity-type controller on the discharge side of the filter.

Mr. Fuller thought I used the cathode liquor along with the gas solution but I did not, as there was too much salt in it. It took about 10 lb. of salt to produce only 1 lb. of chlorine. This much salt would increase the chlorides and be objectionable, so I did not use [the cathode liquor]. Jewell's adventure in chlorination was given only these few lines in Fuller's Louisville report (54):

Jewell device for the application of chlorine . . . consisted of a set of small U-tubes, in which a common salt solution was decomposed by an electric current. A constant flow of water was maintained through the tubes. The water dissolved the hypochlorites and carried them with it to the water in the top of the filter. The apparatus was never used regularly, but was tried on January 21 and 22, and for very short periods at later dates. On January 22 available chlorine was applied in this way during the morning at the rate of 0.1 ppm. by weight of applied water.

The Jewell and Fuller statements differ as to whether chlorine gas or hypochlorite solution was applied to the water and as to the point of application but Jewell's sketch and statement are in agreement. The point of application is of minor importance. It seems safe to say that this was the first use of chlorine gas to reduce the bacterial content of the effluent from a working-scale water filter.

The Harris Magneto-Electric Water Purification System was tested at length in 1896–97 under the direction of George W. Fuller, at the Louisville testing station. Previously, small demonstration installations had been operated by Harris at a point on the badly polluted Passaic River in New Jersey and also in Brooklyn, N.Y. Fuller described at length Harris apparatus as first submitted and subsequently modified, and concluded that "the direct application of electricity and electro-magnets, as used in these devices, produced no substantial purification of the Ohio River."

The original Harris apparatus consisted of magnets charged with high voltage electric current discharged into water that subsequently passed through tanks, each of which contained electrodes. The fundamental principles of the system, Fuller says (54),

——were never explained to me. Electro-chemical action was considered to be an important factor in the destruction of the bacteria and organic matter in the water. It was intended that all suspended matter would be repelled by the action of the magnets situated at the top of the three tanks; and the magnets were to force the suspended matters, including the bacteria, to the bottoms of the tanks, where pipes leading to the sewer were provided.•

• Years later, Harris installed a sewage treatment plant at Santa Monica, Calif., which, after changes by associates or successors, remained in use for many years. A half dozen sewage treatment plants based on his system were built in Oklahoma. One of these, at Oklahoma City, was described at length by Hinckley, the description accompanying my review of attempts to treat sewage by electricity (42). A few years later all but one of the Oklahoma plants had been abandoned.-M.N.B.



Professors Palmer and Brownell of the Louisville Manual Training High School, stimulated by the trials of the Harris apparatus, set up experimental electrical devices in their laboratory. The Louisville Water Co. was induced to try their system to determine the relative merits of iron and aluminum electrodes to provide a coagulant for treating Ohio River water before filtration. Seventeen pages of Fuller's Louisville report (54) are devoted to these tests, made in the early part of 1897. It was found after work had been started that the devices made under plans provided by Brownell represented the Palmer and Brownell Water Purifier-patent applied for. The application was dated February 2, 1897; a patent granted February 15. Besides data pertaining to the Harris and to the Palmer and Brownell (or Mark and Brownell) electrical devices, Fuller's report contains an analysis of electrolytic action and comparative data on coagulants produced by the electrolysis of iron and aluminum electrodes and commercial sulfate of alumina, with conclusions favorable

to the latter for use with subsidence and mechanical filtration.

Chlorination at Adrian, Mich.-Chlorination was used in 1899 at Adrian, Mich., under the direction of William M. Jewell. In the spring, when unable to obtain bleaching powder, he improvised apparatus to produce chlorine gas in conjunction with a demonstration filter, using a horse sweep for power. In the autumn he used bleaching powder on the effluent from a permanent installation of filters, in order to bring the effluent from Jewell filters up to the Michigan standard of chemical purity. How long the bleaching powder was used is unknown, but in a report on analyses of the raw and filtered water made in April 1899 by Professor Victor C. Vaughan, of the Laboratory of Hygiene, University of Michigan (55), mention is made of a taste in the effluent due to the use of "sodium chloride." Chemically, Vaughan stated, the effluent did not come up to the standard shown on the blank form used in making the report, but the standard was an ideal one and the water was considered safe for use. Bacterial counts were 144 per ml. before filtration. The unfiltered water had been dosed with sodium hypochlorite. Recollections of these activities at Adrian by F. B. Smart, a company employee of that period (56), are in general agreement with the foregoing statements. Sodium chloride, he says, was never permanently adopted by the company and was soon discontinued. Since about 1915 the city has used chlorination continuously.

Permanent Chlorinating Plants.-The first permanent water chlorination plant anywhere in the world was put into use at Middelkerke, Belgium, in 1902. It was brought to the attention of water works men in America by George C. Whipple in 1906 (57) and again in 1931 by William Boby (58). From these sources and by later correspondence with Boby (59), the following notes have been taken: The installation was "invented" by Dr. Maurice Duyk, then chemist to the Belgian government. Chloride of lime and perchloride of iron (0.2 and 8 ppm.) were fed through drip cocks into badly polluted water before it passed through four gravity filters, each 5 ft. in diameter. This was continued until about 1921 when "pure spring water" was introduced. Mr. Boby states that in 1904, acting in collaboration with a consulting engineer, he delivered chlorinating apparatus to water works at Christchurch, England, but was informed by the local medical officer of the district that he would not allow any form of chemical treatment. "Thus," says Boby, "although a certain amount of contamination was known to be present in the water, I was debarred by the limited foresight and education of the medical officer in question from being the pioneer of chlorination in England."

The second permanent use of chlorination on a municipal water supply was begun early in 1905 at Lincoln, England, and continued until 1911. An alkaline solution of sodium hypochlorite, having the trade name "Chloros," was added to the water on its way to slow sand filters. It contained about 10 per cent available chlorine. The work was done under the direction of Dr. Alexander Houston, "with Dr. McGowan on the chemical side" (60). Considerably earlier (1897) Dr. Sims Woodhead used bleaching powder for water-main disinfection after the serious typhoid epidemic at Maidstone, England, attributed to pollution of the water by hop pickers.

Boonton and Bubbly Creek.-Impetus to chlorination came in 1908 from its introduction on a large scale at the Boonton Reservoir of the water works of Jersey City, N.J., and on a small scale at the Bubbly Creek filters of the Union Stockyards at Chicago. Chlorination at the Boonton Reservoir was not an outgrowth of chlorination at Bubbly Creek, as has been said repeatedly and as would be naturally inferred from George A. Johnson's statement (61):

The first demonstration in this country in a practical way of the usefulness of hypochlorites in connection with water purification was made in the fall of 1908 at the filter plant of the Chicago stockyards, on the recommendation

336

and under the direction of the writer. Following directly on the heels of the spectacular results obtained at Chicago, came the adoption of the process for the sterilization at Boonton, N.J., of the impounded and unfiltered water supply of Jersey City, with which work the writer was also connected.

Careful study of available data shows that although the Bubbly Creek plant was put in operation a few days before the one at the Boonton Reservoir, the decision to use chlorination at Boonton was made first, and preliminary tests were made there first as well. The Boonton plant was the conception of Dr. John L. Leal. It was the outcome of litigation brought by Jersey City for alleged non-fulfillment of a contract made in 1899 for a new water supply. Under this contract water of the Rockaway River was to be impounded by a dam and conveyed to Jersey City by a conduit. Owing to litigation instigated by rival water supply interests, delays and financial embarrassment postponed the beginning of construction until 1902. Meanwhile the contract was taken over by the rival interests and executed by the East Jersey Water Co. The supply works were completed in 1904.

The contract provided that the water should be "pure and wholesome and free from pollution deleterious for drinking and domestic purposes" (62). Rightly or wrongly, it may be interjected, the people of Jersey City had come to believe that the sewage of a number of small towns above the dam would be diverted from the river or at least treated to avoid infection of the water supply. However this may have been, the city sued for non-fulfillment of the quality clause of the contract and sought to prove that the contractor should filter the water.

On the basis of evidence submitted to it the court ruled, on May 1, 1908, that although "perhaps two or three times a year" the bacterial count was too high and coliform organisms were found in too small samples of water, and although filtration would prevent this, yet evidently the contract did not contemplate the construction of filters. The court suggested, on the basis of evidence submitted by the city, that diversion sewers and sewage treatment works be constructed by the contractor.

Dr. Leal, sanitary adviser to the contractor, a few days after the court ruling, "strongly advised" the water company to suggest its own method for the complete fulfillment of the contract. "On June 4, 1908," says Leal, "the court authorized the company to submit to it

within 90 days plans for meeting the terms of the contract." Accordingly, Leal recommended the use of sodium hypochlorite as a disinfectant, and the company agreed. "On June 16, 1908," says Leal, "I engaged the firm of Hering & Fuller to design the necessary works and Mr. George A. Johnson of said firm to operate the same" (62). Design of the plant was begun by Hering & Fuller three days later, testified George W. Fuller before the court (63).

As early as 1897 or 1898, wrote Leal in his report of the case (62), "I had made rather extensive experiments with electrolytical solutions of salt and also with solutions of bleach in connection with the proposed purification of another water supply. The results were most favorable from a bacterial standpoint, although the method was not used because it did not fulfill all the requirements of the water under consideration." Leal's first idea was to use an electrolytic solution at the Boonton Reservoir, but, being unable to find a suitable cell, chloride of lime was adopted. He knew by 1909 that chlorination had been tried as a disinfectant, generally on sewage, at a dozen places in England, France, Belgium, Germany, India, Mexico and the United States, including the Woolf process in the Croton watershed and at Jerome Park Reservoir, New York City, and the Duyk process at Middelkerke and Ostend, Belgium. "No special discovery," he said, "is claimed in connection with the process in operation at Boonton, N.J." He added: "I do claim, however, that this is the first time it has been used on any such scale, or as a continuous or permanent system of water purification. I also claim that, as a result of the investigations made by us, certain facts in connection . . . with the process have been obtained, which had not been heretofore recognized."

The claims for size and for useful information on chlorination were justified but apparently Leal had overlooked the fact that the installations he mentioned at Middelkerke, Belgium, and Lincoln, England, were also permanent.

The Boonton chlorination plant went into use September 26, 1908, said Leal in his court testimony. Johnson testified that he was at Boonton "getting things ready" for a week prior to September 26, and was in charge of the operation of the plant until the end of the year. Johnson also testified that for about a month before March 20, 1908, he operated an electrolytic cell obtained from the National Laundry Machine Co., Dayton, Ohio, and that from March 20 to 23, 1908, he applied electrolytically prepared hypochlorite of sodium to the water

at Boonton in order to compare the relative efficiency of that agency and of bleaching powder. Comparative tests extending through three weeks showed no difference in the efficiency of the two (63).

The Boonton plant was approved May 9, 1910, by a special master in chancery. He reported the first cost of the plant as \$20,546 and the operation and maintenance expense as \$2,100 a year. Sewage works and the necessary trunk sewer to divert sewage from the water supply, he said, would have cost several hundred thousand dollars and required considerable capital and operating charges. Late in 1910, the New Jersey Supreme Court accepted the master's finding as to satisfactory character of the chlorination plant. Almost a year afterward the New Jersey Court of Errors and Appeals sustained the finding of the lower court (64).

Thus ended the litigation but the litigation is not all the story. Some years later the city completed at its own cost an activated-sludge plant to treat the sewage of some of the towns above the dam and a trunk sewer leading to the plant.

Chlorination at Bubbly Creek was adopted to bring the effluent from mechanical filters up to the contract guarantee. Data to establish the nature and course of events that led to chlorination for a short time have been obtained for use here from Arthur E. Gorman, Engineer of Water Purification, Chicago (65), and from Charles A. Jennings, an associate of Johnson in the tests on disinfection at Bubbly Creek (66).

In 1907, the Norwood Engineering Co. completed a 5-mgd. mechanical filter plant for the Union Stock Yards Transit Co., at Chicago. The filters were located on the south bank of Bubbly Creek, near Halstead St. Sulfate of iron and lime were used as coagulants. In 1908, George A. Johnson, of Hering & Fuller, New York City, made a series of four tests to determine whether the filter effluent complied with the contract guarantees. The test periods, says Gorman, were April 7–20, June 1–14, July 27–August 2 and September 3–17. Jennings, who assisted Johnson, states that sulfate of iron and lime worked nicely as coagulants and that the effluent looked well and was low in bacteria. But the organic matter was high and after the water had stood in the clear well the bacterial count "would jump very rapidly into the thousands and higher." First, copper sulfate was tried as a germicide. In the test runs of July 27–August 2 and September 3–17, hypochlorite of lime was used. The last-named run, says Jennings, may be considered as an acceptance test of the filter plant by the Stock Yards Co.

In February 1909 the city council passed an ordinance prohibiting the taking of water from the Chicago River or any of its branches for watering livestock or for use in preparing meats, poultry or provisions for human consumption or for any other use which endangers the public health. Use of such waters for motive power was not prohibited. In June 1909, sulfate of alumina displaced lime and iron as a coagulant at the Bubbly Creek filters. Later that year the city of Chicago brought suit against the Stock Yards Co. for violating the ordinance. A thousand pages of testimony were taken in the municipal court. In April 1910, states Gorman, partly as a result of the city's opposition and partly to avoid prejudice that might affect its business, the company discontinued the use of filtered water for its livestock. About November 1, 1915, a softening process was adopted at the filtration plant.

Race states that the hypochlorite of lime was applied $7\frac{1}{2}$ hours before filtration, at the rate of 45 lb. per mil.gal. (36, 64).*

¹Other Hypochlorite Applications.—Among the first American cities to adopt chlorination on a permanent basis was Poughkeepsie, N.Y., which began application of chloride of lime on February 1, 1909, and installed permanent apparatus on March 17 of the same year. It was the failure of combination treatment by sedimentation, coagulation and slow sand filtration to render the Hudson River raw water supply potable following the drought summer of 1908 that prompted the city authorities to consult George C. Whipple, and, on his recommendation, to substitute chloride of lime for alum in the treatment process. Introduced first into the low lift pump suction line by means of the regular coagulant apparatus, the chloride of lime was regularly applied at the inlet to the sedimentation basin as soon as a satisfactory dosing appliance could be devised₁(67, 68) (see also Chap. VI).

Largest city to adopt disinfection at the time was Philadelphia. Sodium hypochlorite, produced from electric cells, was applied in September 1909 to water in 200-mgd. prefilters at Torresdale on the Delaware River. Hypochlorite was again used in December 1910

340

[•] Articles on the Bubbly Creek plant were published in Engineering Record, 58: 659-68 and 58: 703-05 (1908); and in Engineering News, 64: 245 and 64: 342 (1910); the last of these was a 5,000-word letter from George A. Johnson, on litigation with the city.

but as the bacterial efficiency declined in cold weather it was decided to apply chloride of lime to the water in the clear-water basin. The chloride of lime treatment was discontinued in April 1911 but then resumed in December of the same year and maintained continuously until February 1913 (69).

The Application of Liquid Chlorine.-Liquid chlorine was first produced as an article of commerce in the United States in 1909 (70) and was used experimentally in 1910 for water disinfection by Major C. R. Darnall, U.S.A. Medical Corps, at Fort Myer, Va. Seven years earlier Lieutenant Nesfield, of the Indian Army Medical Service, reported the result of numerous experiments on the destruction of pathogenic organisms by chlorine and proposed its application to military use. "This," says Race (36), "was the first suggestion of the possibilities of compressed chlorine gas in steel cylinders."

In June 1912, Dr. Georg Ornstein of the Electro Bleaching Gas Co. experimented with liquid chlorine and developed the first equipment to employ the solution-feed process in which chlorine gas is dissolved in a minor stream of water and the solution is introduced into the major flow of water. At Philadelphia, liquid chlorine was used experimentally in September 1912, under the direction of Seth M. Van Loan, then Assistant Chief, Bureau of Water, assisted by George E. Thomas. They applied the gas directly to water in the clear-water basin of the Belmont filters.

The first full-scale application of liquid chlorine for water disinfection was made with the Ornstein equipment in November 1912, at the Niagara Falls filter plant of the Western New York Water Co. by Dr. Ornstein, who acknowledged the valuable technical assistance of H. F. Huy, Principal Assistant Engineer of the plant (36).

In December 1912, John A. Kienle, engineer of the water works of Wilmington, Del., began experiments with liquid chlorine. J In a paper on the subject he states that the first results were rather discouraging, and then mentions profiting by experience at Philadelphia and acting on some of Dr. Ornstein's ideas (71).

Subsequently Dr. Ornstein further developed the solution-feed process and the Electro Bleaching Gas Co., which acquired the rights to Ornstein's U.S. Patent 1,142,361, marketed the equipment; Kienle became associated with the company to manage its sales. In 1917, Wallace & Tiernan Co. became sole licensee of the patent.

341

V

V

The first permanent liquid chlorine plant was installed at Philadelphia's Belmont filters in September 1913. During October and November of that year, additional installations were made at all the other Philadelphia plants. The largest of these, capable of treating 200 mgd. was put in use November 25, 1913, at the Torresdale plant on the Delaware (69).

Early in 1941 chlorination in the United States was being used by 4,590 of the 5,372 water works using any kind of water treatment (72). Hypochlorite of lime had largely given way to chlorine gas.

Chlorination Abroad

In Great Britain, the use of chlorination spread slowly. Aside from the instances already cited "perhaps the earliest example of its use was as supplementary routine treatment in connection with Paterson's rapid filtration plant installed in 1911 for the Cheltenham" water works on the River Severn (73). In 1916, the London Metropolitan Water Board began to apply bleaching powder to water from the Thames before it entered the Staines Aqueduct on its way to slow sand filters. This was a war-economy measure which, with subsequent adoptions of chlorination, saved many thousands of pounds by obviating pumping water to storage to get the benefit of bacterial reduction before filtration. In the summer of 1917, the board began to apply liquid chlorine to one of its supplies, using a British proprietary dosing apparatus then coming into use. Early in 1919 the board installed this apparatus to treat New River water. The plant had a capacity of 48 mgd. (U.S.) and "was for many years the largest gaseous chlorine installation in the United Kingdom" (73). It was not until 1921, Lieutenant-Colonel Harold tells in his report for 1936 (74), that chlorination of filtered water was begun, "with its attendant bogey of taste." Subsequently the use of ammonia to control taste was adopted. "The close of 1936 marks the advent of a new epoch, when all filtered water receives chloramine treatment before being passed into the supply." To that end chlorine was being applied at 28 points and ammonia at 45 points. Pre- and postchlorination combined, 300 mgd. (360 mgd. U.S.) were being treated.

Summaries of a questionnaire sent out by the British Waterworks Association in 1939 or 1940 show that in a group of over 600 local authorities about 40 per cent of the water supplies were not chlorinated, most of them being small. Of 134 supplies, each serving a

population of 50,000 or more, only 26 did not chlorinate and, of those, twelve were in Scotland and three in Wales (75).

In France, chlorination goes under two names: javellization, named after *eau de Javelle*, first made in 1792 at the Javelle works near Paris, and verdunization, so named by Philippe Bunau-Varilla after he chlorinated the water supplied to French troops in 1916 at the Battle of Verdun. In a book by Bunau-Varilla (76) he states that at Verdun he used hypochlorite of soda in doses considerably smaller than had formerly been used in France. "Our experiences proved in the autumn of 1916," says Bunau-Varilla, "that when a water is clear it is not necessary to render it nauseating to make it safe for use." In January 1917, Bunau-Varilla treated clear water with chlorine at the rate of 0.1 ppm. Commenting on these statements, Francis D. West wrote in 1931 (77): "It is quite evident that they were playing safe with the water supplies for the army, by soaking them heavily with chlorine until they became so strong of chlorine that they could not be used. B-V., to my mind, went a little too far the other way."

In 1934, wrote Dr. Edouard Imbeaux (48), many French cities chlorinated their water supplies. Gaseous chlorine was used by Montpellier, St. Nazaire and Fremay. Many other cities used hypochlorite of soda or *eau de Javelle*. "Bunau-Varilla uses the term 'verdunization,' saying he invented the process at Verdun, during the war; it is not true, and we cannot accept the word 'verdunization' as scientific."

In 1935, S. McConnel (78) stated that French engineers preferred hypochlorite of soda or javellization to any other form of chlorine. Since Bunau-Varilla introduced verdunization he has pushed its use elsewhere, claiming no royalties. The process was being used in Paris, Lyons, Avignon, Amiens, Rheims, Bordeaux, Monte Carlo, Brussels, Geneva, Lisbon and Seville. Bunau-Varilla claims, "and is supported [in his claims] by French scientists," says McConnel, "that instantaneous sterilization is due to the emission of ultraviolet rays."

In Germany, wrote Dr. Karl Imhoff early in 1941 (79), chlorination apparatus was in place at that time on at least 30 per cent of the city water works, but in most cases was used only temporarily.

Ozonation

Ozonation began about the same time as chlorination. For some years it made more rapid progress, then chlorination shot ahead and left ozonation far behind. In America and Great Britain, ozonation

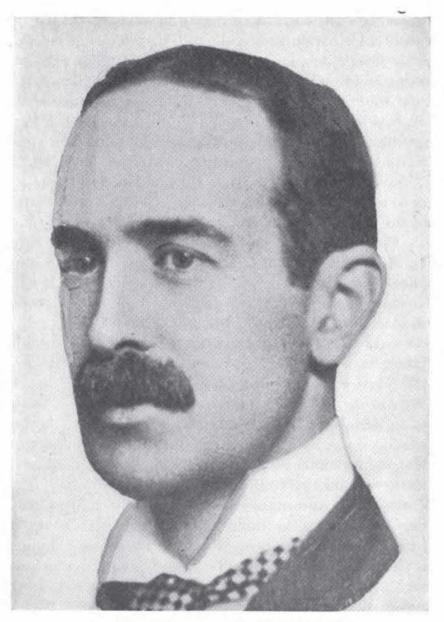


FIG. 65. GEORGE A. SOPER (1868-1948)

First American engineer to investigate ozonation of water; undertook research as subject for doctorate at Columbia University shortly after completing work as operator of Warren Filter at Louisville Filter Testing Station (From photograph taken about 1896)

Digitized by Google

of public water supplies had hardly gained a foothold up to 1941. On the continent of Europe it fared better from the start, but only in France, with its strange complex of ideas on water purification, have many ozonation plants been established.

Handicapped by cost of both installation and operation, ozonation cannot compete with the equally efficient, simpler and far less expensive chlorination. One advantage of ozonation over chlorination is that it does not create new and increase existing taste-and-odor troubles. In America, taste-and-odor control has of late given promise to ozonation which it lacked or lost during the earlier years of its trial and abandonment. Even so, it must be regarded as an uncertain competitor of ammonia compounds and activated carbon. This phase of ozonation is reviewed by John R. Baylis in his monograph on the whole subject of taste-and-odor control (80). His section on ozone contains a succinct review of the history of ozonation with descriptions of the various types and makes of apparatus promoted up to 1935. This section therefore will be confined chiefly to brief mention of the earlier papers introducing ozonation to American water works men; to a chronological summary of American trials of the process; and to a brief summary of the scanty available figures on the number of ozonation plants in various other countries.

A few British and American patents on the production of ozone preceded by an early mention of ozone as a bactericide follow:

C. J. Fox, in 1873, reported experiments showing that ozone destroyed bacteria in fluids containing organic matter [Citation from Baylis (80)].

E. H. C. Monckton, in a British patent granted January 21, 1874, states "Water is purified by ozonizing it by passing electric currents through it in tubes or channels of special construction, which is at the same time being acted on by electric current."

In the American aeration process patent granted to Albert R. Leeds of Hoboken, N.J., May 6, 1884 (No. 298,101; application filed November 3, 1883), the claims are for purifying water by "saturating it with oxygen or ozone by causing the water to come in contact, while under artificial pressure and in motion, with compressed air." The specifications mention "destroying any deleterious substances."

On December 29, 1896, a British patent was granted to J. Y. Johnson (Electric Rectifying & Purifying Co.) on the production of ozone by passing oxygen from a reservoir through a cooling vessel into space between two connecting cylindrical vessels, across which there was a brush discharge of electric current. Experiments and Comments.-The first American investigator of ozone as a water disinfectant was George A. Soper. As a part of work for his doctor's degree in the late 1890's he made laboratory tests at Columbia University, reviewed the literature of ozonation and visited the Tindal plants applying ozone to water at Blankenberg, on the coast of Belgium, and at Joinville le Pont, in the environs of Paris. His conclusion was that "drinking water can be sterilized, and that unpleasant colors and odors arising from organic impurities can be removed by ozone" (81).

Very soon after the appearance of Soper's thesis, Robert Spurr Weston published an abstract of a German paper on ozonation, read in 1899 by Theo. Weyl before the German Society of Gas and Water Works Engineers (82). The abstract summarized results at a small test plant at Charlottenburg, near Berlin. In appended critical comments, directed pointedly at Weyl's assertion that "sand filtration is uncertain" while ozonation renders water "germ free," Weston said that both Weyl and Dr. Soper "assume that the enginemen and electricians in charge of an ozone plant would be more careful than men with the same degree of training in charge of a filter plant."

Sharp dissent to Weyl's paper was voiced in the discussion which followed and which was abstracted by Allen Hazen for the benefit of American readers (83). William Lindley, English-born and -trained engineer located for many years at Hamburg and at Frankfort, Germany, speaking for those in charge of large water works, differed strongly with Weyl's contention that "sand filters are among the most dangerous appliances to be found in the control of cities." He added: "In scientific circles it will not do to set up the results of a little first experiment against a well-proved system which, on an enormous scale and through decades, has been carried out with success."

Another early contribution to the literature of ozonation, written for the particular benefit of American water works men, was made early in 1901 by the Russian engineer, Nicholas Simin, who was chief engineer of the water works at Moscow (84). After visiting experimental plants in Europe, the Russian engineer suggested the use of rapid filters and a small amount of coagulant to prepare water for ozonation. In discussing the paper, Allen Hazen aptly summed up the role of ozonation by saying that it must be considered as an auxiliary to sedimentation and filtration, "simply removing the relatively small number of bacteria remaining after these processes had



been employed, together with the odor and, if enough ozone were used, the color."

In 1906, a masterly review of ozonation to that date was submitted to the American Water Works Association by George C. Whipple (57). In 1910, the subject was reviewed at length by *Engineering News* (85). This article described foreign apparatus, including the Vosmaer system, the American rights of which had been acquired by a Philadelphia company. A few foreign municipal installations, all small, were described, as were also three plants, also small, on this side of the Atlantic.

Shortly after the appearance of this article, two New York engineering journals described the ozonation plant completed in 1910 by the city of St. Petersburg [Leningrad] (86, 87). This seems to have been the largest installation up to that time in any country. Its capacity was 11 mgd. It consisted of 126 Siemens and Halske ozonators, five Otto emulsifiers and five sterilizing towers. Current stepped up to 7,000 volts was employed.

In America the earliest significant commercial attempts to apply ozone to municipal water supplies were made by the United Water Improvement Co. of Philadelphia. It acquired the American rights to the Vosmaer system, which had been previously installed at Schiedam and Nieuwersluis, Holland, and had been tested at the latter place, but soon took up the American patented system of J. Howard Bridge, who became associated with the company.

Ozonation tests were made in 1906 at an experimental filter plant operated by the Department of Water Supply at Jerome Park Reservoir, New York City. Apparatus was installed at the expense of the United Water Improvement Co. of Philadelphia. It included a Hungerford mechanical filter for the removal of bacteria, a refrigerator to remove moisture from the air used in the production of ozone, and an ozonizer of the "H. Blanken System," in which 60-cycle, 10,000-volt a-c. was used. It was operated May 7–31, 1906, by engineers acting for the city. This appears to have been the first disinterested American experimental test of water ozonation. A summary of the conclusions, by I. M. DeVarona, Chief Engineer of the Department of Water Supply, New York City, approved by Rudolf Hering and George W. Fuller, consulting engineers (88), stated that apparently the color in the water might be reduced from about 15 to 5 and the bacteria from about 100 to 7, at a cost of about \$20 per mil.gal., but the plant "did not run one single day without stopping, a fact of itself sufficient to demonstrate the impracticability of using the proposed method of ozone purification as an adjunct of our filtration plant, regardless of its cost."

Five ozonation plants on municipal water supplies were installed in the United States and Canada within the 1908–12 period. All were eventually abandoned. As nearly as can now be determined, dates of service of the first three plants were: Lindsay, Ont., 1908– 1910; Ann Arbor, Mich., 1909– or 1910–1914; Baltimore County Electric Water Co., near Baltimore, Md., 1910–1918. Not all were used continuously. There were some reconstructions. In 1927, an ozonation plant was installed by the borough of Ogdensburg, N.J., and in 1928, by the village of Delhi, N.Y. The first of these was given up in 1930, on the completion of rapid filters. The second was used until destroyed by a flood in 1935, and was followed by a chlorinating apparatus (89–96).

A year's experimental work at Milwaukee, Wis., in 1919, led to the conclusion that apparatus for water sterilization by ozone had not yet been developed to the point where it could compete with chlorine. These studies (97) were made at the Milwaukee filter testing station by Ernest F. Badger, under the direction of J. W. Ellms, Consulting Engineer, who reported to H. P. Bohman, Superintendent of Water Works. The ozone apparatus was made by the Ozone Co. of America, apparently a Milwaukee concern. Good bactericidal results were obtained on both raw and filtered water but the cost was considered prohibitive.

Recent American Installations and Experiments.—A new phase of ozonation in the United States was begun with the completion of plants in 1930 at Hobart, Ind., and in the spring of 1932 at Long Beach, Ind., for small privately owned water works controlled by a holding company interested in both water and lighting companies. Control of tastes and odors as well as bacteria were objectives of these ozonation plants. An official statement, written late in 1939, regarding these ozone plants, their designer, the company that built them, and its successor is here summarized by the director of trade relations of the new company (98):

The Hobart and Long Branch ozonation apparatus was designed by J. M. Daily, who "spent several years in France with the leading ozone company there." In his apparatus Daily incorporated "several

major improvements over its European prototypes." The equipment at the two Indiana towns was built by the American Ozone Co. The patents of that company were taken over in 1938 by Ozone Processes, Inc., organized "as a member of a group of operating companies owned by Welsbach Street Illuminating Co. Ozone Processes, Inc., is adequately financed and equipped to do necessary research and engineering work . . . never done previously in this country."

Besides an experimental laboratory at Moorestown, N.J., Ozone Processes has operated small "pilot plants" at Whiting, Ind., in 1938-39, and at the New Brighton station of the Beaver Valley Water Co. of Beaver Falls, Pa. A 1-mgd. testing plant was put into use early in 1941 at the Lower Roxborough station of the Philadelphia water works. Filtered water from the Schuylkill River was being ozonated there in cooperation with the Philadelphia Bureau of Water, of which Seth M. Van Loan was Chief. Manganese was quantitatively removed at a pH of 6.8 to 7.0 (99). The tests at New Brighton were given up at the request of the water company because it concluded that ozonation, like other processes tried, was unable to cope with the peculiarly troublesome tastes and odors of Beaver River water. At Whiting, the tests were followed by a contract with the city under which the ozone company installed a 3.5mgd. ozonation plant that was put into use in July 1940. A 0.3-mgd. ozonation plant was completed at Denver, Pa., in March 1940.

Of the three Indiana towns, Long Beach is a small summer resort on Lake Michigan. It is supplied with water from the lake, collected by open-joint tile buried a few feet in lake sand. Ozonation apparatus was installed in 1930. After a series of tests by a private laboratory the water company began applying chloride of lime, in batches, to the pump well. The chlorinated and ozonated water had always met Treasury Standards up to the close of 1939. Occasional unsatisfactory samples had been attributed to the makeshift method of chlorination.

Hobart had a population of 5,800 in 1930, when the water works were constructed. The supply is taken from Deep River, a sluggish stream into which the sewage of Crown Point (4,000 population) is discharged 15 mi. above a small impounding reservoir. The water is coagulated, settled, filtered, ozonated and chlorinated. Ozonation was begun in the spring of 1932 because of odors in the water due to algae. At first, chlorine was applied to the filtrate before the water went to the ozonator but soon the chlorine was added to the raw water. At both Long Beach and Hobart ozonation was adopted some years ago, when the Daily apparatus was in its early stage. The water works at both towns are controlled by the Northern Indiana Public Service Company.

Whiting is located at the southern end of Lake Michigan. The population of about 11,000 is supplied with water from the lake which is badly polluted with domestic sewage and the wastes from oil refineries, steel mills, chemical plants, soap works and corn products factories. Coagulation, filtration and chlorination are said to have made the water hygienically safe but chlorination increased taste-and-odor troubles despite ammoniation. Encouraged by the ozonation trials of 1938–39, the city contracted October 23, 1939, with Ozone Processes, Inc., for the installation of an ozonation plant. This was put into use in July 1940. Contrary to what might be expected, ozone is applied to the raw water, which then passes through the old treatment plant.

The treatment plant, which had proved itself unable to control taste and odor before pre-ozonation was introduced, had a designed capacity of 8 mgd., but the abnormal and increasing pollution had limited it to a maximum of 4 mgd. The normal consumption is 2 mgd., with hot-weather peaks up to 4 mgd. This plant included: ammoniation just ahead of prechlorination; coagulation with alum; baffled flocculation; baffled sedimentation in two basins; rapid filtration; and postchlorination as needed. Activated carbon was not used in normal plant operation because no effective dosage was possible. The output was "generally very unpalatable, due to kerosene and phenolic tastes accentuated by heavy chlorine dosages necessary to cope with the normal heavy primary pollution" (100).

The pre-ozonating plant includes: electrostatic air filter, air dryer, air compressors, ozonators, meters and two ozonizers. Raw water is pumped to the top of the ozonizers. Ozonated air is delivered to the bottom of the ozonizers through porous tubes and passes up through the raw water. Once it has been ozonized, the water is dosed with chlorine, ammonia and alum. It then passes through a bubble-type flocculator and through two settling basins between which the activated carbon is or may be applied, and finally it is put through gravity rapid filters. The filter effluent is then chlorinated on its way to the clear-water well (98, 100).

Arthur W. Consoer, Consulting Engineer of Chicago, and James C. Nellis, Whiting City Engineer (101), state that six months' operation (October 1940–March 1941) showed a cost of \$5.49 per mil.gal. as contrasted to \$3.50 for chemicals alone for six previous months—but capital charges at 6 per cent would add about \$2.00 to the cost of ozonation. The chemicals used were alum, chlorine and ammonia.

During the six-month period, activated carbon as well as ozone was used quite extensively, but with little reduction in threshold odor numbers over ozone alone, although the pilot tests indicated that the combination would be of value. Results of the six months of operation as well as comments of water consumers "indicate quite clearly that ozonation has done more than any other treatment in reducing tastes and odors in the water supply."

The contract between the city of Whiting and Ozone Processes, Inc., contained a guarantee centering on reduction of odor during a three-year period, backed by a surety bond. It provided that the contractor should not be released from the bond by "entry of any lawful order of the Board of Health of Indiana" during the three-year period "requiring a discontinuance of ozonation" (101). This, it may be interjected, reflected the fact that the board had withheld a permit for ozonation other than as a trial of the process.*

At Denver, Pa., a 0.30-mgd. ozonation plant was completed for the borough in March 1940, by Ozone Processes, Inc. The water supply is from Cocalico Creek, a small stream draining an agricultural area. Before ozonation the water is coagulated, settled and passed through a rapid filter. The borough adopted ozonation for sterilization, removal of tastes and odors and reduction of color. The Pennsylvania

•The industrial pollution of water in the lower end of Lake Michigan, and especially at Whiting, is extremely heavy. Changes in oil refinery practices, derived from the increased production of high octane gasoline during and after World War II, introduced new wastes into the lake and thereby lessened the degree of satisfactory removal of tastes and odors effected by ozonation. In April 1948 B. A. Poole, Director, Bureau of Environmental Sanitation, Indiana State Board of Health, reported that up to that time the board "had not been shown by plant results that ozone alone could be depended upon for disinfection of water to be used in a municipal distribution system." At the time of this report, Whiting was using ozonation to improve tastes in its raw water supply only when threshold odors reached a high point. Meanwhile, at Hobart, the ozonators, which had been rebuilt after the war, were not in routine use, primarily because escape of ozone into the plant limited operation to periods when the windows could be opened for ventilation; and at Long Beach, where two ozonators were available, ozonation had been discontinued, at least temporarily.—Ed.

THE QUEST FOR PURE WATER

Department of Health authorized the installation of the ozone plant for demonstration purposes, provided the final effluent was chlorinated. Tests were run until July 1, 1941, jointly by the borough and Ozone Processes, with checks by the State Department of Health. Late in September 1941, the Department approved the ozone plant as an agency for removing tastes and odors and the reduction of color, but insisted on a continuation of postchlorination (99–100).

Summary.-Despite promotion work and a few experiments, no ozonation plant was built in America until 1908. In the period 1908-1942, plants were built at eight water works in the U.S. and one in Canada. The first five of these have been abandoned. In the U.S. the general attitude of health departments that have had occasion to pass upon ozonation projects for municipal supplies has been to regard them as experimental and to grant tentative permits only.

Recent Ozonation Plants in Europe

In England, ozonation of municipal supplies did not begin until 1936. There were only four such plants in use early in 1940. At least two of the four British plants treat only the water of some of the sources drawn upon. The first and largest of the plants was put into use at Brighton in 1936 and has a capacity of 5 mgd. [All the capacities here given are in U.S. gallons.] In January 1937, the South Staffordshire Waterworks Co. put in use a 1.2-mgd. ozone plant of the Van der Made type at its Huntington pumping station, one of seven stations lifting water from the red sandstone (102). In April 1937, a 1.7-mgd. plant was put into operation by the Ashton-under-Lyne, Stalybridge and Dunkinfield Water District to treat water from the Knott Hill impounding reservoir, which was subject to seasonal infestations of algae causing tastes and odors (103). The fourth ozonation plant in England was completed in 1939 by the Colne Valley Water Co. It has a capacity of about 4 mgd. Early in 1941, Lt.-Col. G. Ewart Morgans, of the Water Purification Department of British "Otto" Ozone (104), stated that he had just made a contract with the city of Manchester, England, for a 3.6-mgd. filtration and ozonation plant from which water will be supplied in bulk to Salford. This plant was to be used for "exhaustive tests." It was expected that it would be extended to a capacity of 90 mgd. "This," says the representative of the "Otto" process, "is my fourth big ozone plant to be installed in Great Britain."



Prefatory to his paper on the Ashton-under-Lyne, Lancashire, ozonation plant, M. T. B. Whitson, Engineer and Manager there (103), reviewed the development of European ozonation processes and illustrated the main features of the various methods used to mix ozone and water, particularly the Otto process of 1900, 1914 and 1940, the Siemens-DeFrise process at Paris, of 1900, the Marmier-Abraham process at Chartres, of 1908, and the Van der Made process of 1940, installed at Ashton-under-Lyne. So far as known to him, Whitson said, "of the many types of ozone producers developed during the last 50 years, only two [were in use in 1940] on a commercial scale." Both had been developed from the Werner von Siemens experimental ozonizer of 1857 and depended for the production of ozone on the discharge of high voltage electricity between stationary electrodes. The companies formed by Siemens and DeFrise, Marmier and Abraham and Dr. M. P. Otto were amalgamated in 1910 and 24 plants were erected in France in the next four years. In 1940, said Whitson, there were in use on public water supplies (apparently meaning all types still in service), 90 installations in France, fourteen in Italy, five in Belgium, four in England, three in Roumania, two in the United States and one in Russia (103). "Ozonation, in France," Whitson said, "has come to stay." He believed it deserved more consideration in England than it had received.

The four largest ozonation plants in France, early in 1940, were: Toulon, 26.4 mgd. (U.S.), installed 1910-35, treating impounded surface water after filtration; Nice, 21.17 mgd., installed 1906-30, treating unfiltered water from Vesubie Canal; Nancy, 21.17 mgd., installed 1933, treating water from a filter gallery and from the Moselle River, filtered; Villefranche-sur-Mer, installed in 1910, treating filtered water from Vesubie Canal (105). Under construction for the city of Paris were plants with a contract capacity of 79.26 mgd., designed to ozonize filtered water from the River Marne. A. Gury, formerly Chief of the Paris Municipal Water Service (106), states that ozonation of the Paris supply at St. Maur was stopped during the first World War. Lt.-Col. C. H. H. Harold, Director of Water Examinations on the Metropolitan Water Board, London (17), said that Paris, in 1910, decided that 24 mgd. (U.S.) of Marne water should be ozonated, half by the DeFrise and half by the Otto process. He also mentioned an inspection, made in 1934, of ozonation plants in France, from Nancy in the East to

353

Finistère in the West and along the Riviera from Mentone to Toulon. The majority of the old installations in France, he stated, were located where hydroelectric power could be used. His general conclusion was favorable to ozone treatment, "provided that at all times an absence of breaches in the filter barrier can be guaranteed. On the other hand, chloramine offers an all-round security under every possible condition and is the Safety First water works valve."

In Germany, wrote Dr. Karl Imhoff early in 1941 (107), ozonation was used at some water works many years ago but not at all now.

Ultraviolet Rays

Shortly after chlorination got its running start and while ozonation was entering the lists as a rival, experimental trials of ultraviolet rays as a means of killing bacteria in public water supplies were inaugurated in France. No permanent adoptions for public water supplies in France or elsewhere in Europe have been found on record. In the United States, there were four adoptions on municipal supplies in the period 1916–28, and as many more by industrial concerns for their plants and villages more or less under their ownership. All four municipal installations and two of the industrial-communal plants have been abandoned. In Central America, ultraviolet ray apparatus was reported as installed in 1926, but its capacity was not given. Hundreds of swimming pools and other private or semi-private establishments in the United States have been equipped with ultraviolet ray lamps.

"The earliest positive mention" of the successful use of the bacterial action of ultraviolet rays, stated Kenneth C. Grant, civil engineer in Pittsburgh (108), in 1910, "[was] in 1877, by two English scientists, Downs and Blunt." He adds: "The first attempt to put this method of purification into use is at Marseilles, France, where an experimental plant is being operated in competition with several types of filtration and purification works." Grant's article was chiefly concerned with ultraviolet ray experiments he had seen at the physiological laboratory of the Sorbonne, in Paris. In those tests there was used a quartztube mercury arc lamp of the Westinghouse type.

An article on the Marseilles tests (109) makes it apparent that in 1909 or 1910 the Marseilles authorities invited proprietors of water purification apparatus to install and operate, at their own cost, plants to treat 200 cu.m. (52,840 U.S. gal.) of water per day. The Westinghouse-

354

Cooper-Hewitt Co. of Paris installed ultraviolet ray apparatus which treated water that had been passed through Puech-Chabal filters. During a month's run at the rate of 600 cu.m. or 158,500 U.S. gal. per 24 hours, the article states, the treated water showed no coliform organisms. The effluent from the Puech-Chabal filters, however, is said to have contained only 22 to 24 bacteria per ml. Details of all the competitive tests at Marseilles were given early in 1911 in an article by Walter Clemence, London agent of the Puech-Chabal filter interests (110).

Prompted by an article in *Eau et Hygiene*, Engineering News (111) said, "If the merciless hunt goes on, bacteria will become as scarce in France as snakes in Ireland." It noted that the water company at Rouen, although supplying water of good repute from springs back of the city, was carried away by "the current French craze for absolute sterility in water-supplies" and had experimented with ultraviolet rays from Westinghouse-Cooper-Hewitt mercury lamps. These were applied to the effluent from Puech-Chabal filters. All the tests showed that the filters alone gave a high reduction in total bacteria, a complete removal of coliform organisms, and a complete absence of bacteria in the water subjected to ultraviolet rays. Commenting editorially on the tests, Engineering News said: "Rouen turns ultraviolet rays on a corporal's guard of bacteria that, now and then a day, manage to pass the tortuous zoogloea-lined channels of a multiple battery of Puech-Chabal filters." The editor then asks: "Is this worth while? . . . have Rouen and other French cities which are so hard on the trail of the last solitary germ in their water-supplies ever approached a like state of perfection in all other or any other matter affecting public health? Are their milk supplies absolutely pure? . . ."

In the United States, the first known installation of ultraviolet ray apparatus on a municipal supply was put in use at Henderson, Ky., toward the end of 1916 (112). It was given up in 1923 or 1924. The population of Henderson in 1920 was about 12,000. This was the largest city in America to use ultraviolet rays. Berea, Ohio, installed ultraviolet ray apparatus in August 1923, and abandoned it in July 1936 (113, 114). Similar treatment was also begun in 1923 at Horton, Kan.; it has since been abandoned (115). The fourth and last of the municipal water works known to use ultraviolet rays was Perrysburg, Ohio, where apparatus was put into use in 1928 and abandoned in 1939 (114).

All four of these plants were equipped by the R.U.V. Co., Inc., of New York. In 1931 that company sent the author a list that included a considerable number of other water supply installations. of which a few were for industrial plants and related communities and others for hotels and institutions. Of four industrial-communal plants, information from company officials and state health departments shows that one installed in 1926 at Parr, S.C., by the Broad River Power Co., was in use in 1940; one completed in 1919 at Mascott, Tenn., by the American Zinc Co., was apparently in use in October 1939. Of the two not in use, one was installed in 1918 and given up in 1932 by The Washington Mills Co., at Fries, Va.; the other at the Gill Hill, Kan., Camp of the Cities Service Oil Co., was operated from 1919 to 1929. Reasons for abandoning six of the eight municipal or industrial-communal plants were: cost of operation compared with other means of water disinfection available; the belief of state officials that chlorination would have greater sanitary efficiency (116), and unsuitableness of the electric current in the case of Henderson, Ky. Late in 1941, an R.U.V. representative stated that no attempt to obtain municipal water supply installations had been made for some years, due to the small size of quartz lamps available.

A few vessels on the Great Lakes have used ultraviolet rays for water sterilization since 1916 or 1917. In 1928, when Frank H. Shaw took charge of U.S. Public Health Service control of water supplies in the Great Lakes district, bacterial samples were collected from 230 of 643 interstate vessels in use. Seventeen had R.U.V. installations, eleven used chlorination, two used ozone, 167 used stills and 33 took water from certified shore sources. During 1933, ozone on the two vessels was replaced by chlorination. Writing early in 1940, Shaw (116) stated that ultraviolet rays were still being used to sterilize lake water that had previously been filtered but probably some of the R.U.V. installations had been replaced by other methods of disinfection since 1928. Meanwhile, the number of vessels using chlorination had increased to 31.

356

- 49. PELIGOT, EUGENE. Études sur le Composition des Eaux. Comptes Rendus Hebdomadaires des Séances, 58:729-738 (1864). [D]
- 50. GARDNER, L. H. Discussion of "Methods of Filtration of Water in Use in the United States and Europe," by JOHN JAMES ROBERTSON CROES. Proc. A.W.W.A., p. 74 (1883).
- 51. ———. The Clarification and Purification of Public Water Supply. Scientific American Suppl., 20:8146 (1885). [D]
- 52. DRAGENDORFF, G. Purification of Water Containing Organic Impurities. *Pharmaceutical Jour.*, 3rd Se ries, 2:1049 (1872). [D]
- GWINN, DOW R. The Use of Iron as a Coagulant in Connection With Mechanical Filtration. Proc. A.W. W.A., pp. 94-99 (1900).
- 54. WOLMAN, ABEL; DONALDSON, WELL-INGTON; & ENSLOW, LINN H. Recent Progress in the Art of Water Treatment. Jour. A.W.IV.A., 22:1161-1177 (1930).

CHAPTER XIV

Disinfection

- 1. HERODOTUS. History. English version, ed. by GEORGE RAWLINSON. D. Appleton & Co., New York (1859). [D]
- 2. HIPPOCRATES. The Genuine Work of Hippocrates. Tr. from the Greek by FRANCIS ADAMS. Williams & Wilkins Co., Baltimore (1939). [D]
- 3. PLACE, FRANCIS EVELYN, Water Cleansing by Copper, Jour. Preventive Medicine (London), 13:379 (1905). [A]
- SUS'RUTA. The Hindu System of Medicine According to Sus'ruta. Tr. from the Sanskrit by UDOY CHAND DUTT. Vol. 95, Chap. 45, pp. 215-222, Bibliotheca Indica, Royal Asiatic Society of Bengal. J. W. Thomas, Calcutta (1883). [D]
- PLINY THE ELDER. The Natural History of Pliny. Tr. by JOHN BOSTOCK & H. T. RIPLEY. (Bohn Classical Library) H. G. Bohn, London (1855-57). [D]
- 6. PLUTARCH. Vitae Parallelae. The tr. called Dryden's, corrected from the Greek and rev. by A. H. CLOUGH. Little, Brown & Co., Boston (1859). [D]
- SEMPLE, ELLEN CHURCHILL. Domestic and Municipal Waterworks in Ancient Mediterranean Lands. Geog. Review, 21:466-474 (1931).
 [D]

- 8. AEGINETA, PAULUS. The Seven Books of Paulus Aegineta. Tr. from the Greek with a commentary by FRANCIS ADAMS. In Greeks, Romans and Arabians on All Subjects Connected With Medicine and Surgery. Sydenham Society, London (1844-47). [D]
- 9. IBN SINA, HUSSAIN IBN ABD ALLAH. A Treatise on the Canon of Medicine of Avicenna. Incorporating a tr. of the "First Book" by G. CAM-ERON GRUNER. LUZAD & CO., LONdon (1930). [D]
- BOFRHAAVE, HERMANN. Elements of Chemistry. Tr. from the original Latin by TIMOTHY DALLOW. J. & F. Pemberton, London (1735). [D]
- 11. DUNGLINSON, ROBLEY. Human Health; or Influence of Atmosphere, Food . . . on the Healthy Man. Philadelphia (1814). [D]
- 12. RIDEAL, SAMUEL. Water and Its Purification. C. Lockwood & Son. London (1902). [D]
- & ERIK. Chemical Disinfection and Sterilization. E. Arnold & Co., London (1921). [D]
- BECHMANN, M. Purification of Water for Domestic Use. French Practice. *Proc. A.S.C.E.*, Part D, 44: 183–190 (1904). [E]

- IMBEAUX, EDOUARD. Annuaire Statistique et Descriptif des Distributions d'Eau de France, Algerie, Tunisie. C. Dunod, Paris (3rd ed., 1931). [D]
- 16. HINMAN, JACK J., JR. Water Supply in the Field. Vol. 1, No. 4, Univ. of Iowa Studies in Medicine, Iowa City (1918). [D]
- 17. LONDON METROPOLITAN WATER BOARD. 29th Annual Report. London (1934). [X]
- RIDEAL, SAMUEL & BAINES, E. The Suggested Use of Copper Drinking Vessels as a Prophylactic Against Water-Borne Typhoid. Royal San. Inst. Jour. (London), 25:591-595 (1904). [E]
- 19. KRAEMER, HENRY. Use of Copper in Destroying Typhoid Organisms and the Effects of Copper on Man. *Amer. Jour. Pharmacy*, 77:265-281 (1905). [K]
- 20. (a) AMY, JOSEPH. Nouvelles Fontaines Domestiques, Approuvées par l'Academie Royale des Sciences. J. B. Coignard, Paris (1750). [B]
 (b) —. Suite du Livre Intitulé "Nouvelles Fontaines Filtrantes Approuvées par l'Academie Royal des Sciences." Antoine Boudet, Paris (1754). [B]
- 21. MOORF, GEORGE THOMAS & KELLER-MAN, KARL F. A Method of Destroying or Preventing the Growth of Algae and Certain Pathogenic Bacteria in Water Supplies. Bulletin No. 64. Bureau of Plant Industry, U.S. Dept. of Agriculture, Govt. Printing Office, Washington, D.C. (1904). [D]
- 22. Copper as an Algicide and Disinfectant in Water Supplies. Bulletin No. 76. Bureau of Plant Industry, U.S. Dept. of Agriculture, Govt. Printing Office, Washington, D.C. (1905). [D]
- 23. JACKSON, DANIEL D. The Purification of Water by Copper Sulfate. Eng. News, 54:307-308 (1905). [E]
- 21. MOORE. GEORGE THOMAS; ET AL. The Use of Copper Sulfate and Metallic Copper for the Removal of Organisms and Bacteria From Drinking Water. Jour. N.E.W. W.A., 19:474-582 (1905).

- 25. BROWN, C. ARTHUR. The Practical Sterilization of Public Water Supplies by Means of Copper-Iron Sulfate and Filtration. Proc. A.W. W.A., pp. 239-252 (1905).
- NAGELI, CARL VON. Über Oligodynamischen Erscheinungen in Lebenden Zellen. Schweizerische Naturforschenda Gesellschaft, 33:1-59 (1893). [D]
- THIELE, HERMANN & WOLF, K. Uber die Bakterienschädigenden Einwirküngen der Metalle. Archive für Hygiene, 34:43-70 (1899). [D]
- SACL, P. Über die Keim Kotende Firnwirkung von Metallen Oligodynamische Wirkung. Klinische Wochenschaft (Vienna), 30:714 (1917). [X]
- 29. SUCKLING, E. V. The Sterilization of Water by Catadyn Silver. Water & Water Eng. (London), 34:15-18 (1932). [E]
- MOISEEV, S. V. The Sterilization of Water by Silver Coated Sand. Jour. A.W.W.A., 26:217-238 (1934).
- JUST, J. & SZNIOLIS, A. Germicidal Properties of Silver in Water. Jour. A.W.W.A., 28:492-506 (1936).
- ANON. Silver Process Effective in Water Sterilization. Chem. & Metallurgical Eng., 41:372 (1934). [C]
- SHAPIRO, ROBERT & HALE, FRANK E. An Investigation of the Katadyn Treatment of Water. Jour. N.E. W.W.A., 51:113-124 (1937).
- DORROH, J. H. The Design and Operation of Swimming Pools. Jour. A.W.W.A., 27:100-107 (1935).
- 35. Manual of Water Quality and Treatment. American Water Works Assn., New York (1940).
- RACE, JOSEPH. Chlorination of Water. J. Wiley & Sons, New York (1918). [E]
- 37. HOOKER, ALBERT HUNTINGTON. Chloride of Lime in Sanitation. J. Wiley & Sons, New York (1913). [E]
- AMERICAN PUBLIC HEALTH ASSN. COMMITTEE ON SEWAGE DISPOSAL. Unpublished report for 1933. [X]

496

- CRIMP, WILLIAM SANTO. Sewage Disposal Works. Griffin, London (1894). [E]
- 40. FULLER, GEORGE WARREN. Sewage Disposal. McGraw Hill Book Co., New York (1911). [E]
- 41. METCALF, LEONARD & EDDY, HARRI-SON P. Disposal of Sewage. Vol. 111, American Sewerage Practice. McGraw-Hill Book Co., New York (1914–15). [D]
- ANON. A Review of Attempts to Use Electricity in Sewage and Water Purification. Eng. News, 67: 534-536 (1912). [E]
- 43. Patents for Inventions-Class 46: Filtering and Otherwise Purifying Liquids. H.M. Stationery Office, London (1676-1866). [E]
- BAKER, M. N. Sewage Purification in America. Eng. News, New York (1893). [X]
- 45. BACHE, R. MEAD. Possible Sterilization of City Water. Am. Philosophical Soc. Proc., 29:26-39 (1891).
 [D]
- 46. ANON. The Electrical Purification of Water. Eng. Rec., 29:110 (1894).
 [E]
- GAYOL, ROBERTO. Enforme el Sistema de Saneamiento por Medio de Soluciones de Chloruros Electrolizados. A. N. Ayuntamiento, Mexico City (1893). [X]
- IMBEAUX, EDOUARD. Personal Letters. Nancy, France (August 20 & 30, 1934).
- DROWN, THOMAS M. Electrical Purification of Water. Jour. N.E.W. W.A., 8:183 (1893-94). [E]
- 50. FRANKLAND, EDWARD. On the Metropolitan Water Supply During the Year 1866. Chem. News (London), 15:151 (1867). [C]
- 51. WANKLYN, J. ALFRED. Report on the Organic Pollution of Drinking Waters. British Med. Jour., p. 351 (March 30, 1872). [X]
- 52. ANON. Second Report Dealing With the Atkins Moulded-Carbon Filter. British Med. Jour., p. 338 (September 21, 1872). [X]

- 53. JEWELL, WILLIAM M. Personal Letters. Chicago (1933–35).
- 54. FULLER, GEORGE WARREN. Report on the Investigation Into the Purification of the Ohio River Waters at Louisville, Ky. D. Van Nostrand, New York (1898). [B]
- 55. VAUGHAN, VICTOR C. Report on Analyses of Water From the Adrian (Mich.) Water Co. Before and After Passing Through a Jewell Mechanical Filter, April, 1899. From papers of JOHN A. COLE, Consulting Engineer to the Adrian Water Co. (1899). [X]
- 56. SMART, F. B. Personal Letters. Adrian, Mich. (1934-35).
- 57. WHIPPLE, GEORGE C. Disinfection as a Means of Water Purification. *Proc. A.W.W.A.*, pp. 266–288 (1906).
- 58. BOBY, WILLIAM. The Early Use of Chlorine. Jour. A.W.W.A., 23:283– 284 (1931).
- 59. ——, Personal Letter, London (April 1931).
- 60. HOUSTON, ALEXANDER CRUIKSHANK. Studies in Water Supply. Macmillan, London (1913). [E]
- 61. JOHNSON, GEORGE A. Hypochlorite Treatment of Public Water Supplies-Its Adaptability and Limitations. Am. Jour. Pub. Health, 1: 562-574 (1911). [E]
- 62. (a) LEAL, J. L. The Sterilization Plant of the Jersey City Water Supply Co. at Boonton, N.J. Proc. A.W.W.A., pp. 100-109 (1909).

(b) FULLER, GEORGE WARREN. Description of the Process and Plant of the Jersey City Water Supply Co. for the Sterilization of the Water of the Boonton Reservoir. Proc. A.W.W.A., pp. 110-134 (1909).

(c) JOHNSON, GEORGE A. Description of Methods of Operation of the Sterilization Plant of the Jersey City Water Supply Company at Boonton, N.J., and Discussion of Results of Analyses of Raw and Treated Water, With Notes on the Cost of the Treatment. *Proc. A.W.W.A.*, pp. 135-162 (1909).

- 63. LEAL, J. L.; FULLER, GEORGE WAR-REN; & JOHNSON, GEORGE A. Defendants' Testimony. Jersey City *vs.* Jersey Water Supply Co. In Chancery of New Jersey. (February-October 1909). [X]
- 64. ANON. The Hypochlorite Plant. Eng. News, 63:686 (1910); 64:483 (1910). [E]
- 65. GORMAN, ARTHUR E. Chronology of the Bubbly Creek Chlorination Plant. Based on data in his file. [X]
- 66. JENNINGS, CHARLES A. Personal Letters. (September-October 1936).
- 67. Отія, Јонм С. The Poughkeepsie Water Works. Jour. N.E.W.W.A., 23:283-301 (1909).
- 68. POUGHKEEPSIE BOARD OF PUBLIC WORKS. Annual Report. Poughkeepsie, N.Y. (1909). [E]
- 69. WEST, F. D. Disinfecting 200,000, 000 Gallons of Water a Day-Experience With Chloride of Lime and Liquid Chlorine at Torresdale Filtration Plant (Philadelphia). Jour. A.W.W.A., 1:403-455 (1914).
- BALDWIN, ROBERT T. History of the Chlorine Industry. Jour. Chem. Education, 4:313-319 (1927). [C]
- KIENLE, JOHN A. The Use of Liquid Chlorine for Sterilizing Water. Proc. A.W.W.A., pp. 267-289 (1913).
- U.S. PUBLIC HEALTH SERVICE. National Census of Water Treatment Plants of the United States. Water Works Eng., 96:63-117 (1943).
- 73. ANON. Water Sterilization by Gaseous Chlorine. Paterson Engineering Co., Ltd., London (1928). [E]
- 74. LONDON METROPOLITAN WATER BOARD. 31st Annual Report. London (1936). [E]
- ANN. Chlorination of Water Supplies. (British Waterworks Assn. Questionnaire.) The Surveyor, 97: 518 (1940). [E]
- BUNAU-VARILIA, PHILIPPE. L'Auto Javellization Imperceptible. Paris (1926). [X]

- 77. WEST, FRANCIS D. Personal Letter. Arlington, N.J. (1931).
- MCCONNEL, S. Verdunization. Water & Water Eng., 37:310-311 (1935).
 [E]
- 79. IMHOFF, KARL. Personal Letter. Essen, Germany (April 20, 1941).
- 80. BAYLIS, JOHN ROBERT. Elimination of Taste and Odor in Water. Mc-Graw-Hill Book Co., New York (1935). [E]
- SOPER, GEORGE A. The Purification of Drinking Water by the Use of Ozone. Eng. News, 42:250-253 (1899). [E]
- ANON. Sterilization of Water by Ozone. Eng. News, 42:340 (1899).
 [E]
- ANON. Relative Value of Ozone and Slow Sand Filtration as a Means of Purifying Water. Eng. News, 43:92-95 (1900). [E]
- 84. SIMIN, NICHOLAS. Ozonized Air. Proc. A.W.W.A., p. 41 (1901).
- ANON. Production and Utilization of Ozone With Especial Reference to Water Purification. Eng. News, 63:488-496 (1910). [E]
- 86. ANON. Ozone Water Purification Plant at St. Petersburg, Russia. Eng. Rec., 63:473 (1911). [E]
- 87. ANON. Ozone Plant at St. Petersburg, Russia. Eng. News, 66:783 (1911). [E]
- ANON. Report on the Filtration of the Croton Water Supply, New York City. Eng. News, 58:561-562 (1907). [E]
- 89. HEAD, WILLIAM C. Personal Letter. Ann Arbor, Mich. (1933).
- 90. PRYER, R. W. Water Purification by Ozone, With Report on the Ann Arbor Plant. Jour. Ind. & Eng. Chem., 6:797-800 (1914). [C]
- CASWELL, H. H. Development of Ann Arbor Water Supply. Jour. A.W.W.A., 32:1577-1586 (1940).
- 92. RAY, D. Personal Letter. Lindsay, Ont. (August 29, 1933).

498

- 93. WALDEN, A. E. & POWELL, S. T. Sterilization of Water. *Proc. A.W. W.A.*, pp. 128–149 (1910).
- 94. POWELL, S. T. The Use of Ozone as a Sterilizing Agent for Water Purification. Jour. N.E.W.W.A., 29: 87-93 (1915).
- 95. ——, Personal Letter. Baltimore, Md. (1933).
- 96. Cox, CHARLES R. Significant Experiences in the Treatment of Water in New York State. Jour. N.E. W.W.A., 53:444-457 (1939).
- ANON. Milwaukee Testing Station Results on Filtration of Water. Eng. News-Rec., 85:257-259 (1920).
- JENKS, STANLEY. Personal Letter. Philadelphia, Pa. (October 7, 1941).
- 99. HANN, VICTOR. Ozone Treatment of Water. Jour. A.W.W.A., 35:585-591 (1943).
- 100. BARTUSKA, JAMES F. Ozonation at Whiting, Ind. Jour. A.W.W.A., 33: 2035-2050 (1941).
- 101. CONSOER, ARTHUR WARDEL & NELLIS, JAMES C. Ozone Reduces Water Odors and Tastes. Eng. News-Rec., 127:367-369 (1941). [E]
- 102. ANON. Huntington Pumping Station of the South Staffordshire Waterworks Co. Eng., 143:651-653, 711-713 (1937). [E]
- 103. WHITSON, M. T. B. The Treatment of Water by Ozone. Paper read before Manchester and District Assn. of Inst. C.E. (March 20, 1940). [X]
- 104. MORCANS, G. EWART. Personal Letter. London (May 23, 1940).
- 105. FRENCH FOREIGN OFFICE. Information obtained by French Consul at Philadelphia (1940). [X]
- 106. GURY, A. L'Eau à Paris. (Extraits d'un Manuscrit, écrit en 1913.) Paris (1939). [Mimeographed Abstract.] [E]
- 107. IMHOFF, KARI.. Personal Letter. Essen, Germany (April 20, 1941).
- 108. GRANT, KENNETH C. Sterilization of Polluted Water by Ultra-violet Ray. Eng. News, 64:275 (1910). [E]

- 109. ANON. Sterilization of Polluted Water by Ultra-violet Rays at Marseilles, France. Eng. News, 64:633 (1910). [E]
- 110. CLEMENCE, WALTER. The Water Supply of Marseilles and Trials of Filtering and Sterilizing Apparatus. Eng., 91:106-108, 139-142 (1911). [E]
- 111. ANON. Another French Quest for the Last Germ in City Water Supplies. Eng. News, 66:686 (1911).
 [E]
- 112. SMITH, A. T. Ultra-violet Rays Finish Treatment of Henderson (Ky.) Water Supply. Eng. News-Rec., 79: 1021-1022 (1917). [E]
- BLOCHER, JOHN MILTON. The Purification of Water by Ultra-violet Radiation. Jour. A.W.W.A., 21: 1361-1372 (1929).
- 114. WARING, F. H. Personal Letter. Columbus, Ohio (October 18, 1939).
- 115. BOYCE, EARNEST. Personal Letter. Lawrence, Kan. (October 23, 1939).
- 116. SHAW, FRANK R. Personal Letters. Chicago (1933 & 1940).
- 117. WALDEN, A. E. & POWELL, S. T. Sterilization of Water by Ultraviolet Rays. Proc. A.W.W.A., pp. 341-346 (1911).
- 118. VON RECKLINGHAUSEN, MAX. Purification of Water by Ultra-violet Rays. Jour. A.W.W.A., 1:565-588 (1914).
- 119. Ultra-violet Rays for Water Purification. Jour. N.E.W.W.A., 29:202-213 (1915).
- 120. SPENCER, R. R. Ultra-violet Rays; Their Advantages and Disadvantages in the Purification of Drinking Water. Jour. A.W.W.A., 4:172– 182 (1917).
- 121. FAIR, GORDON M. Elements of the Theory Underlying the Disinfection of Water by Ultra-violet Light. Jour. A.W.W.A., 7:325-342 (1920).
- 122. PERKINS, ROGER G. & WELCH, HENRY. Sterilization of Water by Ultraviolet Light as Emitted by the Carbon Arc. Jour. A.W.W.A., 22:959-967 (1930).