FRIEDRICH WÖHLER AND THE CENTENARY OF SYNTHESIS.*

BY OTTO RAUBENHEIMER.

The pyramids themselves, doting with age, Have forgotten the names of their founders.—Fuller.

However, we do not have to go back 5000 years to the pyramids, as frequently, very frequently, events of great importance to Pharmacy, only one hundred years ago, are forgotten. It remains an everlasting credit to the Scientific Committee of the New Yorker Deutscher Apotheker Verein, the oldest pharmaceutical society in the United States, established 1851, to remember the birth of the great Liebig (1803) and to celebrate its centenary in 1903 and also the one of the master of pharmacy, Hager (1816), and to arrange celebration in 1916, in which, by the way, the Historical Section of the A. PH. A. also took part. This year marks the centenary of the Synthesis of Urea and a "Wöhler Symposium" was arranged on February 2, 1928, in which the writer and Prof. C. P. Wimmer and Dr. Fr. Klein took part. The N. Y. Apotheker Zeitung published part of these papers in German but for the presentation in English my entire paper is contributed to the Section on Historical Pharmacy at the Portland meeting of the AMERICAN PHARMACEUTICAL ASSOCIATION.

BIOGRAPHY OF WÖHLER.

Born in Eschersheim, Hesse-Cassel, on July 31, 1800, he entered the school at his native place, then at Roedelheim and the "gymnasium" in Frankfurt-on-the-Main. Here he obtained an excellent education under such eminent teachers as Karl Ritter, E. Grotefund and F. C. Schlosser. Through the publication of that epoch-making masterwork by Leopold Gmelin, "Handbuch der Theoretischen Chemie" in Frankfurt, 1817-1819, Wöhler became interested in chemistry. After graduation from the high school in the spring of 1820 he entered the University of Marburg with the intention to study medicine. Then he changed to Heidelberg so as to furthermore study chemistry and mineralogy under Gmelin. On September 2, 1823, he was promoted as Doctor Medicinae, sumna cum laude. Upon Gmelin's advice he abandoned medicine to study chemistry for another year under the master of chemistry, Jons Berzelius, in Stockholm. When Wöhler reached Lübeck he found that the next boat for Stockholm did not leave for six weeks. He spent this time in the laboratory of Apotheker Kindt, the discoverer of pinene chloride or artificial camphor. With Berzelius, Wöhler chiefly worked on mineral analysis and it is reported that the master's opinion of his assistant's analysis frequently was: "Schnell aber schlecht!" In 1824 Wöhler also accompanied Berzelius and the celebrated French geologist, Brongniart, on a two-month scientific trip through Scandinavia. Upon his return to the Fatherland and through the recommendations of the former students of Berzelius, Heinrich Rose and Eilhard Mitcherlich and especially of Leopold von Buch, a friend of the great Humboldt, Wöhler was appointed teacher of chemistry at the Technical School in Berlin, in 1825, which rank was increased to Professor in 1827. Here he became intimately acquainted with such scientists as Mitcherlich, the two brothers Rose, Poggendorff and Magnus.

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In 1831 he was called to Cassel to organize the newly erected Technical High School at which he was made Professor of Chemistry and Technical Chemistry. In 1836 he received a call to the University of Göttingen as the successor of Friedrich Stromeyer (1778–1835), who in 1817 discovered cadmium and determined its compounds. Here as Professor of Medicine and Chemistry, Director of the Chemical Institute and Inspector of Apothecary Shops in Hanover he exercised great influence and remained a bright ornament of the "Georgia-Augusta" until his death on September 23, 1888.

WÖHLER AND LIEBIG.

As early as 1820, as a medical student at Marburg, Wöhler began his researches on cyanogen compounds. These were continued at Heidelberg, 1821–1823, and as the result he published two papers on Cyanic Acid in Gilbert's Annalen. At the same time Liebig worked on fulminic acid with Gay-Lussac in Paris. In the laboratory of Berzelius, 1823-1824, Wöhler discovered that his cyanic acid and Liebig's fulminic acid had the very same chemical composition H.CNO. Liebig doubted this statement, but Wöhler persisted and at an interview in Frankfurt in 1825 convinced Liebig. This discovery led to a lifelong friendship and also caused Berzelius to coin "isomerism" (from the Greek isos, alike). Shortly afterwards Wöhler and Liebig again had different opinions as to the composition of "Kohlenstickstoffsäure," our pieric acid, or better, trinitrophenol of to-day. Wöhler at the time wrote to Liebig, "Es muss ein böser Demon sein, der uns immer wieder unvermerkt mit unsern Arbeiten in Zusammenstoss oder Kollisionbringt und das chemische Publikum glauben machen will, wir suchten dergleichen Zankäpfel als Gegner absichtlich auf. Ich denke aber es soll ihm nicht gelingen! Wenn sie Lust dazu haben, so können wir uns den Spass machen, irgend eine chemische Arbeit gemeinschaftlich vorzunehemen, und das Resultat unter unserem gemeinschaftlichen Namen bekannt zu machen. Versteht sich, Sie würden in Giessen und ich in Berlin arbeiten nachdem wir uns den Plan eingeteilt und uns von Zeit zu Zeit über den Fortgang Nachricht gegeben hätten. Ich überlasse die Wahl des Gegenstands ganz Ihnen."

Just as Göthe found in Schiller a friend and companion of similar taste, so gained Liebig in Wöhler a collaborator with whom he performed epoch-making discoveries. A very touching and generous reference to Wöhler is made by Liebig in his Autobiographical Sketch, in a passage which displays a side of his character that all his pupils know well—his unselfishness and kindness of heart. "I had the great good fortune, from the commencement of my career at Giessen, to gain a friend of similar tastes and similar aims, with whom, after so many years, I am still united in the bond of warmest affection. While in me the predominating inclination was to seek out the points of resemblance in the behavior of bodies or their compounds, he possessed an unparalleled faculty of perceiving their differences. A keenness of observation was combined in him with an artistic dexterity and an ingeniousness in discovering new means and methods of research and analysis such as few men possess. The achievement of our joint work upon urie acid and oil of bitter almond has frequently been praised—it was his work!

"I cannot sufficiently highly estimate the advantage which the association with Wöhler brought to me in the attainment of my own as well as our mutual aims, for by that association were united the peculiarities of two schools—the good

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that was in each became effective by coöperation. Without envy and without jealousy, hand in hand, we pursued our way; when the one needed help, the other was ready. Some idea of this relationship will be obtained if I mention that many of our smaller pieces of work which bore our joint names were done by one alone; they were charming little gifts which one presented to the other."

The first joint investigation, in 1829, was the determination of the composition of mellitic acid, $C_6(COOH)_6$, which was discovered by the apothecary Klaproth in 1789. Then followed, in 1830, a thorough research on cyanic acid H.CNO together with the discovery of new compounds. After Wöhler's removal to Cassel, in 1831, the classic investigation of benzoic acid was commenced and its composition determined as C_6H_5 .COOH. In 1832, Liebig and Wöhler published their immortal work "*Das Radikal der Benzoesāure*," in which they demonstrated the doctrine of radicals, which were capable of living transferred unchanged from one compound to another, in a manner analogous to inorganic elements. They found that the benzoyl radical C_6H_5CO could be combined with Cl, Br, I, S, NH₄, etc., always retaining its own individuality. Berzelius was so struck by this discovery that he considered it the "Anfang eines neuen Tages in der vegetabilischen Chemie" and he suggested the name "Proine" or "Orthrine," meaning "dawn" for "benzoyl."

Robiquet and Boudron, in 1830, discovered amygdalin in bitter almonds and Liebig and Wöhler, in 1837, determined its composition and also explained its decomposition through the ferment emulsin and water into benzaldehyde, hydrocyanic acid and glucose—therefore the name "glucoside." Amygdalin is therefore the prototype of the glucosides. Each time the writer prepares Infusion or Syrup of Wild Cherry or Emulsion of Bitter Almond, he cannot help but think of Liebig and Wöhler. In the same year (1837) they also prepared benzaldehyde in the pure state and studied its relation to benzoic acid.

In 1776, the apothecary Carl Wilhelm Scheele discovered uric acid and in 1837–1838 Liebig and Wöhler made their famous investigation and discovered fifteen new derivatives. Liebig in his Autobiographical Sketch narrates the following incident of this period: "During our joint research on uric acid, Wöhler one day sent me a crystalline body which he had obtained by the action of lead peroxide upon this acid. I immediately wrote to him with great joy and without having analyzed the substance, that it was allantoin. Seven years before I had this body in my hands; since that time I had not seen it again." It was a wonderful instance of what Liebig called "sight of eye memory," as this identification of allantoin was ultimately found to be correct. The memorable research on uric acid was the foundation of all future work thereon. Liebig said, "Schwerlich is je eine Arbeit der Art ausgeführt worden, welche schwieriger und reicher an Resultaten war; um alles ins Klare zu bringen dazu gehört ein Menschenalter."

The uric acid research was the last joint work of the two scientists, as Liebig turned more to physiological chemistry. Liebig and Wöhler were the inaugurators of what von Noorden calls the qualitative period of metabolism experiments. The two investigators will remain inseparable in the history of chemistry. Liebig himself gives expression to this in one of his last letters to Wöhler, dated December 31, 1871, in the following beautiful terms: "Even after we are dead, and our bodies returned to dust, the ties which unite us in life will keep our memory green, as an instance—not very frequent—of two men who wrought and strove in the same

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field without envy or ill feeling and who continued in the closest friendship throughout."

WÖHLER'S DISCOVERIES.

As early as 1820, when a student at the University of Marburg, Wöhler improvised a chemical laboratory in his rooms, in which he began his great life work on the cyanogen group. Here he prepared mercuric sulphocyanide, cyanogen iodide and other carbon-nitrogen compounds. This work was continued in the laboratory of Professor Leopold Gmelin at the University of Heidelberg during 1821–1823, resulting in the publication of two papers on cyanic acid in *Gilbert's Annalen*. As already pointed out Wöhler's discovery of the isomerism of his cyanic acid and Liebig's fulminic acid in the laboratory of Berzelius led to the intimate friendship of both scientists.

At Stockholm Wöhler just missed discovering vanadium, as he worked with its oxide. Sefström, in the same laboratory, in 1830 discovered the element which he named after the Scandinavian goddess, Vanadis. Soon afterwards Wöhler proved that Sefström's vanadium, and erythronium, discovered in 1801 by Del Rio in the mineral vanadite are identical. In the laboratory of Berzelius, Wöhler chiefly worked on mineral analysis, and in the course of his researches prepared tungsten oxychloride, WOCl₄, and sodium tungstate or tungsten sodium-bronze, $Na_2W_3O_9$. In 1823 Wöhler discovered amorphorous silicon (Si), the crystalline variety not being prepared until 1854 by Sainte-Claire Deville. Wöhler analyzed the deposits in the Swedish iron blast furnaces and determined their composition as Ti_5N_4C .

Besides the discovery of Si, Wöhler is responsible for the isolation of the following elements: Aluminum, in 1827, beryllium or glycium and the rare earth element yttrium in 1828. He obtained Al in the form of a gray metallic powder by the fusion of aluminum chloride with potassium, but in 1845 he succeeded in obtaining the metal in the form of beads. It is to be regretted that Wöhler's "aluminium" has been anglicized to aluminum! The element boron was prepared in a pure state by Davy in 1807 and Gay-Lussac and Thenard in 1808, but it remained for Friedrich Wöhler to further investigate it.

In 1838 he showed the difference between the official or infusible NH₂HgCl and the fusible (NH₃)₂HgCl₂—their composition being determined by Kane. In the same year he discovered parabanic acid. He discovered the dimorphism and isomorphism of the oxides of antimony and arsenic acid showed that dimorphous bodies possess different melting points in the amorphous and crystalline condition. He discovered two methods for the preparation of hydroquinone, C6H4-(NH)₂, in 1848, and in 1862 discovered calcium carbide CaC, decomposed same with water and evolved acetylene gas, C₂H₂. He examined castoreum finding phenol and salicin. In the analysis of Bezoar Stone he isolated bezoar acid and lithofellic acid. He discovered allantoin in the urine of suckling calves, prepared mandelic acid from amygdalin and cumarin from tonka bean. He decomposed narcotin into cotarnin and opianic acid. He discovered silicum hydride, SiH4, and arti-In 1860 he prepared cocaine in pure ficial chrome red or basic lead chromate. crystalline form from coca leaves, and besides its bitter taste found that it renders the parts of the tongue with which it comes in contact insensitive. The practical

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use of cocaine as an anæsthetic was not developed until 1884 by the ophthalmogist Carl Koller of Vienna.

In 1863 he called attention to the analogy of C and Si in organic compounds. In the latter part of his life he devoted himself principally to inorganic compounds, namely, the hydrides and halides of non-metals and the sub and peroxides of metals. He is responsible for the elaboration of a number of processes for use in pharmacy, chemistry, metallurgy and the arts.

In physiological chemistry Wöhler made two discoveries. First he found that alkali citrates and tartrates are changed in the metabolism and excreted in the urine as alkali carbonates. In 1824 Wöhler made, and in 1842 confirmed, a discovery which became the starting point of the modern chemistry of metabolism, namely, that benzoic acid taken internally was excreted through the urine as hippuric acid. This was the death nail to the old idea still current in Wöhler's time, that while plants can synthesize their complex materials, animals, on the other hand, have to receive their constituent substances already synthesized from plants or other animals. These two facts, together with the synthesis of urea, caused the celebrated Prof. Aug. Wilhelm Hofmann to write in his Biography of Wöhler: "The present generation, which is constantly gathering such rich harvests from the territory won for it by Wöhler, can only with difficulty transport itself back to that remote period in which the creation of an organic compound within the body of an animal or plant appeared to be conditioned in some mysterious way by the vital force, and they can hardly realize the impression which the building up of urea from its elements made on men's minds."

WÖHLER THE TEACHER.

Not the least of Wöhler's activities was his teaching! His influence as a teacher especially at Göttingen may be described as enormous. Like his friend Liebig, he laid the greatest stress upon a thorough grinding in the rudiments of chemistry. But unlike Liebig, who represented the romantic school, who restlessly rushed from one problem to another, Wöhler was a true representative of the classic school, who quietly but thoroughly investigated the various subjects. Owing to this quiet disposition he survived the younger Liebig by almost ten years.

Wöhler willingly imparted his chemical knowledge gained under Gmelin and Berzelius to his students, who flocked to him at Göttingen. He made the chemical department of the "Georgia-Augusta" famous and thousands of students from all over received their training in his celebrated laboratory. Out of this long list I will call attention to a few who continued to teach in the spirit of their master and who became famous themselves:

Heinrich August Wiggers (1803-80), editor of "Jahresbericht der Pharmazie," honored through the "Wiggers Scholarship" established 1870; Joh. Florian Heller (1813-71), physiological and pathological chemist at Vienna, whose name continues to live by his tests for albumin and sugar in urine; William Knop (1817-90), agricultural chemist and founder of the "Chemische Centralblatt in 1830;" W. H. Kolbe (1818-84), Professor in Marburg and Leipzig, noted for the synthesis of acetic acid (1845), formic acid (1861) and salicylic acid (1873); Andreas Georg Städeler (1821-71), apothecary and professor; Heinrich Limpricht (1827-1909), Professor in Greifswald and author of "Organische Chemie," Braunschweig, 1855; Anton Geuther (1833–89), director of the chemical laboratory, University of Jena; Rudolf Fittig (1835–1910), Professor in Tübingen and reviser of Wöhler's "Grundriss der Organischen Chemie;" Friedr. K. Beilstein (1838–1906), Professor in St. Petersburg and author of "Handbuch der Organischen Chemie."

Among his most influential pupils here in the United States, I beg to point out: Charles F. Chandler (deceased), of Columbia University, Ph.D., Göttingen, 1856; Edgar Fahs Smith (deceased), former Provost of University of Pennsylvania, father of the history of chemistry in the United States; Ira Remsen (deceased), of Johns Hopkins University, Ph.D., Göttingen, 1870; Otto Grothe (deceased), a mineral and organic chemist in Jersey City, who studied under Wöhler, but obtained his Ph.D. in Kiel under Ladenburg.

Wöhler, as a teacher, was revered by his students; as a man he was above reproach and as a scientist he was honored everywhere. The AMERICAN PHARMA-CEUTICAL ASSOCIATION and the Philadelphia College of Pharmacy, and numerous other organizations made Wöhler an honorary member.

WÖHLER'S LITERARY WORK.

The literary productions of Wöhler are very numerous, about three hundred of his own and fifty together with Liebig. They were published in *Gilbert's Annalen*, *Poggendorff's Annalen* and the *Annalen der Pharmazie*, whose title was changed to *Annalen der Chemie und Pharmazie* (*Liebig's Annalen*) of which he was for many years one of its editors. While at Stockholm he started the translation of Berzelius' Jahresbericht der Chemie from 1821 to 1847. He also undertook the collossal work of a German translation of Berzelius' Lehrbuch der Chemie, four volumes, 1825. The fifth edition in five volumes was published 1843-48. Wöhler is the author of the following books: 1. "Grundriss der anorganischen chemie," 1831. The 15th (last) edition was published in 1873 by his former student H. Kopp. 2. "Grundriss der organischen Cheme," 1840. Beginning with the seventh edition (1868) this book was revised by his former student, Rudolf Fittig. The eleventh edition was published in 1837. This book was also translated into English by Prof. Ira Remsen. 3. "Schwefelwasser-quellen zu Neundorf," 1836. 4. Praktische Uebungen in der Chemischen Analyse, 1853. 5. Mineralanalyse in Beispielen, 1861.

He was also collaborator on the following works:

"Handwörterbuch der Reinen und Angewandten Chemie," 1837, by Liebig, Wöhler and Poggendorff. Since 1838, together with Liebig, editor of the Annalen der Pharmazie.

Wöhler's papers and books are written in a clear, forcible but simple style and are remarkable for the depth of their contents. His letters to both Berzelius and Liebig prove that he had plenty of humor. It has been stated that Wöhler was the most prolific chemist produced by Germany!

LITERATURE ON WÖHLER.

Aug. Wilh. Hofmann: "Zur Erinnerung an Friedrich Wöhler," Berlin, 1883. Aug. Wilh. Hofmann: "Aus Justus von Liebig's und Fried. Wöhler's Briefwechsel," two volumes, Braunschweig, 1888, 1901.

Otto Wallach: "Briefwechsel zwischen J. Berzelius und Fr. Wöhler."

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Last, but not least, let me give a brief account of Wöhler's most important and epoch-making discovery, which was the beginning of a new era, namely, the newer organic chemistry.

SYNTHESIS OF UREA.

Organic chemistry, as late as 1827, was defined by the "master of chemistry" at that time, Berzelius, as "Die Chemie der Pflanzen- und Tiersubstanzen, oder der Stoffe, welche unter dem Einfluss der Lebenskraft gebildet werden." Berzelius and his contemporaries made a sharp distinction between the products of the mineral kingdom and the organic substances of animal or vegetable origin which were said to be formed under the influence of a "vital force." This force in living bodies, it was said, could never be imitated in the laboratory. It was even doubted if the elements composing organic substances were subject to the same laws as those of the mineral kingdom. This "vital force" theory received its death nail in the synthesis of urea by Friedrich Wöhler in February 1828 and the new organic chemistry was born.

Urea, carbonylamide, carbamide or carbonyldiamide, $CO(NH_2)_2$, was discovered by the French chemist Hilaire Marie Rouelle in 1773, a brother to the more celebrated Guillaume, François. He isolated a crystalline salt by extracting urine previously evaporated to a syrupy consistence with alcohol, and named the substance Extractum Saponaceum Urinæ. In 1799 Louis Nicolas Vauquelin, director of the École Superieure de Pharmacie in Paris from its foundation in 1803 until his death in 1829 together with his teacher Antoine François Fourcroy (1755–1809) obtained this substance in the pure state and named it "Urée" or "Urea."

During the time (1823–24) when Wöhler worked in the laboratory of Berzelius, he discovered that a combination of cyanic acid and ammonia resulted in a crystalline substance which, however, did not respond to the tests for both components. (*Poggendorff's Annalen der Physik und Chemie*, III (1825), 177.) In a letter from Wöhler to Berzelius dated Berlin, February 22, 1828, he called attention to renewed experiments forming ammonium cyanate from lead cyanate and ammonia and also from silver cyanate and ammonium chloride. Upon evaporation and crystallization Wöhler obtained a new substance, containing neither ammonium or cyanic acid. He had performed his master work—the synthesis of urea!

$NH_4.CNO + \Delta = CO(NH_2)_2.$

He joyfully writes to Berzelius: "Ich kann mein chemisches Wasser nicht halten und muss Ihnen sagen, dass ich Harnstoff machen kann, ohne dazu Nieren, oder überhaupt ein Thier, sey es Mensch oder Hund noethig zu haben. Das cyansaure Ammoniak ist Harnstoff."

Further on he states: "Nun war ich au fait, und es bedurfte nun weiter nichts alseine vergleichenden Untersuchung mit Pisse—Harnstoff, den ich in jeder Hinsicht selbst gemacht hatte und dem Cyan-Harnstoff. Wenn nun, wie ich nicht anders sehen konnte, bei der Zersetzung von cyansauren Blei durch Ammoniak kein anderes Product als Harnstoff, enstanden war, so musste endlich zur völligen Bestätigung dieser paradoxen Geschichte der Pisse-Harnstoff genau dieselbe Zusammensetzung haben, wie das cyansaure Ammoniak. Und dies ist in der That, nach Prout's Analyse

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der Fall, nach welcher der Harnstoff ist 4N + 2C + 8H + 20 (the old way of expressing the formula) das heisst, cyansaures Ammoniak."

The entire subject under the heading "Ueber die Kuenstliche Bildung des Harnstoffs" is published in *Poggendorff's Annalen der Physik und Chemie*, XII (1828), 253-256. It is also abstracted in Graebe: "Geschichte der Organischen Chemie," I, 52 and 55. Thanks are due to the "Father of History of Pharmacy" in the United States, Prof. Dr. Edward Kremers of Madison, Wis., for publishing Wöhler's letter and also Berzelius' answer in the New Yorker Apotheker Zeitung, March 1928, 1-3, after the Wöhler celebration by the New Yorker Deutscher Apotheker Verein in February 1928.

In conclusion I will present a bit of history of synthesis:

1783, Scheele: Hydrocyanic acid by the combination of carbon, carbon dioxide and ammonia.

1786, Scheele: Oxalic acid by the oxidation of sugar with nitric acid (therefore its synonym "Zuckersäure").

1822, Döbereiner: Formic acid (the acid from ants) by the oxidation of tartaric acid.

1824, Wöhler: Oxalic acid from cyanogen compounds.

1825, Gmelin: Croconic acid from potassium and carbon monoxide.

1828, Faraday and Hennell: Alcohol from ethylene.

1828, Wöhler: Synthesis of urea.

1831, Pelouze: Formic acid from hydrocyanic acid.

1832, Winckler: Mandelic acid from oil of bitter almond and hydrocyanic acid.

After this synthetic chemistry advanced at a rapid pace and attained its highest point in the work of Emil Fischer. However, do not let us forget the synthesis of urea in 1828, the starting point of modern chemistry and also that this year marks its centenary.

There is nothing new, except what is forgotten!

SOLUBLE BISMUTH SALTS.

In the course of a series of investigations undertaken with the object of preparing new salts of bismuth which are soluble in oil, and also in organic solvents, M. Picon (Journal de Pharmacie et de Chimie, September 1, 1928) succeeded in preparing two new compounds, bismuth hexahydrobenzoate and bismuth camphocarbonate. The former is prepared by heating hexahydrobenzoic acid and yellow oxide of bismuth in molecular proportions on a waterbath, with constant stirring, until a solid white salt is obtained. To remove the residue of yellow oxide of bismuth which escapes transformation, the product, reduced to a powder, is dissolved in hot benzol, filtered, and the benzol removed by distillation in vacuo. This salt is soluble in organic solvents which do not contain oxygen, and also in oils. Bismuth camphocarbonate is prepared in the same way from camphocarbonic acid and yellow oxide of bismuth. However, to effect combination it is necessary to add a very small amount of water to the mixture.

CAPSOL.

Unguentum Capsici Co.—Capsol, formula of the Association of Norwegian Pharmacists:

| Ethereal extract of capsicum | 100 | Gm. |
|------------------------------|-----|-----|
| Croton oil | 50 | Gm. |
| Camphor | 50 | Gm. |
| Oil of turpentine | 75 | Gm. |
| Oil of cajuput | 100 | Gm. |
| | 100 | Gm. |
| Soft paraffin, white | 300 | Gm. |
| Wool fat | 225 | Gm. |

Melt the paraffins and wool fat, add the croton oil and extract of capsicum, then dissolve the camphor in the mixture. Finally add the oil of turpentine and oil of cajuput and stir until cold.—Through Chemist & Druggist.

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