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REVIEW ARTICLE

Nikolai Nikolaevich Zinin (1812-1880) – eminent Russian chemist-organic, discoverer of the method of receiving aniline by nitrobenzene reduction

Nikolai Nikolaevich Zinin (1812-1880) - eminente químico orgánico ruso, descubridor del método de recibir anilina por reducción de nitrobenceno

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ABSTRACT

It is a survey of literature concerning both life and scientific activity of Nikolai Nikolaevich Zinin. Special attention is paid to his epoch-making discovery made by him in 1842 and his chemical works published in 1839-1880. This survey is supplemented with: 1) names of the authors of Zinin's biographies, biographical notes, and references to him, 2) information about Zinin's works bibliography, 3) description of the selected facts of his life.

Keywords: N. N. Zinin; organic chemistry; Zinin's chemical works; nitrobenzene reduction; aniline; Russia; XIX century.

RESUMEN

Es un estudio de la literatura sobre la vida y la actividad científica de Nikolai Nikolaevich Zinin. Se presta especial atención a su descubrimiento que hizo época en 1842 y sus trabajos químicos publicados en 1839-1880. Esta encuesta se complementa con: 1) nombres de los autores de las biografías de Zinin, notas biográficas y referencias a él, 2) información sobre la bibliografía de obras de Zinin, 3) descripción de los hechos seleccionados de su vida.

Palabras clave: N. N. Zinin; química orgánica; trabajos químicos de Zinin; reducción de nitrobenceno; anilina; Rusia; siglo XIX.

INTRODUCTION

Nikolai N. Zinin's epoch-making discovery made in 1842

The results of the epoch-making discovery Nikolai Nikolaevich Zinin (Fig. 1) published in the Bulletin de l'Academie des Sciences de St. Pétersbourg in 1842 (Poray-Koshits, 1943, cited by Zinin, 1982, p. 211) and later in the Journal für Praktische Chemie (Zinin, 1842). In

his paper entitled Beschreibung einiger neuer Basen, dargestellt durch die Einwirkung des Schwefelwasserstoffes auf Verbindungen der Kohlenwasserstoffe (Description of Some New Organic Bases, Represented by the Action of Hydrogen Sulphide on Hydrocarbons with NO2); Zinin described first of all the action of hydrogen sulphide on the alcohol solution of 1-nitrophthalene. As a result of this reaction Zinin obtained a new compound, which he named "naphtalidam" (1-naphthylamine). He defined its chemical composition. Zinin observed that nitronaphthalene dissolved better in alcohol with addition of ammonium. Therefore, he improved his method and started to dissolve hydrogen sulphide not in the alcohol but in the naphthalene solution initially saturated with ammonia. "In this way, the classic method of reduction nitro compounds with ammonium sulphide came into being – Zinin's method" (Poray-Koshits, 1943, cited by Zinin, 1982, p. 211). Zinin going to generalization of the discovered reaction reduced consecutive compound – nitrobenzene. Similarly to the nitronaphthalene reduction, he saturated purified nitrobenzene alcohol solution with ammonium, and next passed hydrogen sulphide through it. This reaction resulted in the colorless alkaline liquid, which he named "benzidam". Zinin also defined its chemical composition and properties (Poray-Koshits, 1943, cited by Zinin, 1982, p. 211; Figurovskii & Solov'ev, 1957, p. 134; Zinin, 1842).

Different ways of writing an equation of nitrobenzene reduction to aniline by Zinin's method

Basic two problems faced by the chemistry in the XIX century included a way of calculating atomic and molecular weights as well as defining compounds compositions. There were significant controversies in this field among followers of the different chemical schools. These controversies were also related to the writing of the chemical compounds formulas and reaction equations. Therefore, the authors of chemical books published in France, Germany, Russia, and Great Britain between 1852 and 1890 used different methods to show a conversion of nitrobenzene to aniline by Zinin's method. Examples below illustrate this best.

German chemist Leopold Gmelin (1788-1853), professor at the University in Heidelberg, in his book entitled Handbuch Der Organischer Chemie (Handbook of Organic Chemistry), published in 1852, receiving aniline (C12NH7) with Zinin's method presented as follows: C12NH5O4 + 6HS = C12NH7 + 4HO + 6S (Gmelin, 1852, p. 672).

French chemist Faustino Malaguti (1802-1878) in his book entitled Leçons Élémentaires De Chimie (Basic Chemistry Lessons), published in Paris in 1863, receiving aniline (C12H7Az), reducing nitrobenzene (C12H5AzO4) solution saturated with ammonium (AzH3) with hydrogen sulphide (HS), presented in such an equation:

C12H5AzO4 + 6AzH3 + 12HS = 6S + 4HO + 6(AzH3,HS) + C12H7Az (Malaguti, 1863, p. 165).

Eminent Russian chemist Dmitri I. Mendeleev (1829-1896), discoverer of his "periodic law", in his book entitled Organicheskaya khimiya (Organic Chemistry), II edition, published in 1863, presented the following equation of aniline receiving reaction from nitrobenzene with Zinin's method: C6H5NO2 + 3H2S = C6H7N + 2H2O + 3S (Mendeleev, 1863, cited by Mendeleev, 1948, p. 517).

German chemist Friedrich August Kekulé (1829-1896), professor of chemistry at the Bonn University, in his textbook of the organic chemistry published in 1866, reaction of nitrobenzene reduction with hydrogen sulphide showed in this way:

2C6 H5.NO2 + 6H2S = 2C6H5.NH2 + 4H2O + 3S2 (Kekulé, 1866, p. 584).

In 1890, English chemist Sir Henry Enfield Roscoe (1833-1915) and German chemist Carl Schorlemmer (1834-1892) in their book entitled A Treatise on Chemistry. The Chemistry of the Hydrocarbon and Their Derivatives or Organic Chemistry, published in 1890, Zinin's reaction illustrated with the following equation: C6H5.NO2 + 3H2S = C6H5.NH2 + 2H2O + 3S (Roscoe & Schorlemmer, 1890, p. 196).

References to Zinin's method of nitrobenzene reduction to aniline in the chemical literature

The authors of some books of chemistry, chemical technology, history of chemistry, and history of science and technology, published in France, Germany, Norway, Russia, and Great Britain in 1866-2009, informed their readers about Zinin's method of nitrobenzene reduction to aniline. Some authors mentioned this method for Zinin's life, i.e. M. Reimann (1866, p. 21), August Wilhelm von Hofmann (1875, p. 26), and Marcellin Berthelot (1876, p. 119). Other chemists also have seen an importance of this experimental method, e.g. Ernst von Meyer (1888/1898, p. 466); August Bernthsen (1899, p. 366); Theodor Posner (1903, p. 75); Gustav Bunge (1906, p. 196); Thorstein Hiortdahl (1907, p. 71); Richard Willstätter and Heinrich Kubli (1908, pp. 1936-1940); René Du Bois-Reymond and D. Carl Schaefer (1908, p. 466); Sir William A. Tilden (1913, pp. 169-170); Edvard Hjelt (1916, p. 170); Richard Meyer (1922, p. 218); Paul Walden (1952, p. 46); Yurii Ivanovich Solov'ev (1983, p. 237), Irina Yakovlevna Mittova and Alexander Mikhailovich Samoilov (2009, p. 102).

An effect of Zinin's discovery on the development of the organic chemistry and organic chemical industry in the XIX century

In 1943, Russian chemist-organic Aleksandr Yevgen'yevich Poray-Koshits (1877-1949) wrote: "One of the Russian discoveries, which made the highest impact on the development of the organic chemistry was transformation of the aromatic nitro compounds in several reduction products into primary amines. 100 years ago, this process discovered founder of the whole school of organic-synthetics, professor at the Kazan University Nikolai Nikolaevich Zinin.

It happened at the beginning of the organic chemistry, when classic theory of the organic compounds structure not even exist, and the chemist only started to examine interactions of comparatively few available their individual representatives with the simplest reagents" (Poray-Koshits 1943, cited by Zinin, 1982, p. 211).

Other methods of receiving aniline

Up to Zinin's discovery of receiving aniline from nitrobenzene as the source of this reaction, indigo was used for this purpose. In 1826, German chemist Otto Unverdorben (1806-1873) received aniline from indigo dry distillation and named the obtained compound

"crystalline". Fourteen years later, chemist Julius Fritsche (1808-1871), academic at the St. Petersburg Academy of Sciences, received this compound in reaction of the concentrated potassium hydroxide solution with indigo in the elevated temperature. He named obtained compound "aniline" from the Spanish word "añil" (indigo). He revealed also its empiric formula: C6H7N. In 1834, German chemist Friedlieb Ferdinand Runge (1794-1867) discovered this compound in the coal tar and named "kyanole". In 1843, German chemist August Wilhelm von Hofmann (1818-1892) found that Unverdorben's "crystalline", Frische's "aniline", Zinin's "benzidam", and Runge's "kyanole" is the same substance (Figurovskii & Solov'ev, 1957, p. 135; Trepka, 1960, p. 94; Walden, 1952, p. 47).

Because aniline content in the coal tar is scarce, Zinin's discovery in 1842, was epoch-making fact. Due to it, production of aniline in significant quantities with a simple and cheap method from nitrobenzene became possible as nitrobenzene content in the coal tar is very high (Figurovskii & Solov'ev, 1957, p. 135; Trepka, 1960, p. 95).

At the end of the XIX century fifties, acting faster reducers replaced ammonium sulphide for the reaction of nitrobenzene reduction. To receive low aniline quantities a method proposed by the Russian chemist Friedrich Konrad Beilstein (1838-1906), i.e. reaction of nitrobenzene with stannous chloride (Tin (II) chloride) solution in hydrochloric acid, proved useful: C6H5.NO2 + 3SnCl2 + 6HCl = C6H5.NH2 + 3SnCl4 + 2H2O (Roscoe & Schlorlemmer, 1890, p. 196).

In 1859, French chemist Antoine Béchamp (1816-1908) showed that aniline may be obtained in reaction of nitrobenzene with iron filings and acetic acid. Since 1864, acetic acid in this reaction was replaced by hydrochloric acid: C6H5.NO2 + 3Fe + 6HCl = C6H5.NH2 + 3FeCl2 + 2H2O (Roscoe & Schorlemmer, 1890, p. 196). This method was used not only in chemical laboratories but was also used to produce aniline on the industrial scale.

Information about bibliography of Zinin's works

Bibliography of works published by Nikolai N. Zinin in 1839-1880 includes 54 items (Zinin, 1982, pp. 249-257). The majority of these publications are the articles presenting his experiments (Figurovskii & Solov'ev, 1957, pp. 173-178). In 1957, Russian historians of chemistry Nikolai Aleksandrovich Figurovskii (1901-1986) and Yurii Ivanovich Solov'ev (born 1924) made detailed analysis of Zinin's chemical works (Figurovskii & Solov'ev, 1957, pp. 125-153). They distinguished three basic fields of his experimental works: "1) analysis of the substances extracted from the natural products; 2) investigation of chemical character of the substances with oxidation and reduction processes; 3) investigation of the condensation reaction and chemical properties of aromatic aldehydes and ketones, benzoic aldehyde derivatives" (Figurovskii & Solov'ev, 1957, p. 127). Supplementation of the above was chemical studies concerning i.a. fermentation and nitration processes as well as analysis of nitro compounds.

The results of 30 experimental studies Nikolai N. Zinin published in German in the following journals: 1) Justus Liebigs Annalen der Chemie (12 publications in 1840-1866); 2) Journal für Praktische Chemie (17 articles in 1842-1866), and 3) Annalen der Pharmacie (1 publication in 1839) ("Wiley Online Library", 2018). This last publication (1839) concerned the first Zinin's chemical work. It was entitled Beiträge zur Kenntnis einiger Verbindungen

aus der Benzoylreihe (Contributions to the Knowledge of some Compounds of the Benzoyl Series) (Zinin, 1839). Zinin published it during his training abroad (Figurovskii & Solov'ev, 1957, p. 46).

Literature and references to Zinin from the years 1874 – 2018

Biographies

Russian-language literature about Nikolai Nikolaevich Zininin the years 1880 – 1955 includes 37 items. There are both the books and articles in the journals, such as: Uspiekhi khimii (Advances in Chemistry), Zhurnal khimicheskoy promyshlennosti (Journal of Chemical Industry) and Zhurnal prik□adnoy khimii (Journal of Applied Chemistry) (Figurovskii & Solov'ev, 1957, pp. 179-180).

Out of the books in which Russian authors presented Zinin's biography in 1880-1965, one should list biographic sketch written immediately after his death by his former students, eminent Russian chemists Aleksandr Porfir'yevich Borodin (1833-1887) and Aleksandr Mikhaylovich Butlerov (1828-1886). They published it in the Zhurnal Russokogo fiziko-khimicheskogo obshchestva (Borodin & Butlerov, 1880, cited by Zinin, 1982, pp. 181-209). In 1881, an article of the same authors devoted to Zinin was published in the German journal Berichte der deutschen chemischen Gesellschaft in the part "Obituary" (Butlerov & Borodin, 1881).

In 1921, Russian chemistry historian Boris Nikolaievich Menshutkin (1874-1938) published biographic book about Zinin (Menshutkin, 1921). Twenty seven years later, Russian chemist Alexander Erminingel'dovich Arbuzov (1877-1968) briefly described Zinin's life and scientific activity in his Kratkiy ocherk razvitiya organicheskoy khimii v Rossii (A Short Essay of Organic Chemistry in Russia) (Arbuzov, 1948, pp. 28-34). In 1957, N. A. Figurovskii and Yu. I. Solov'ev published biographic sketch about Zinin (Figurovskii & Solov'ev, 1957). In 1965, Russian prosaic writer Lev Ivanovich Gumilevskii (1890-1976) published Zinin's biographic book (Gumilevskii, 1965).

Publications devoted to Zinin's life and scientific activity in English are not numerous. For example, 15 April, 1880, short article about Zinin was published in Nature (T.H.N., 1880, pp. 572-573). In 1940, Henry M. Leicester (1906-1991), American chemistry historian, published an article entitled N. N. Zinin, an early Russian chemist in the Journal of Chemical Education (Leicester, 1940, pp. 303-306). In 1995, Nathan M. Brooks, professor at the New Mexico State University (U.S.A.) published an article entitled Nikolai Zinin at Kazan University (Brooks, 1995, pp. 129-142), while in 2002, presented Zinin's biography in the Bulletin for the History of Chemistry (Brooks, 2002, pp. 26-36). In 2012, David E. Lewis, professor of chemistry at the University of Wisconsin-Eau Claire (U.S.A.), briefly described Zinin's life and selected results of his chemical works in the book entitled Early Russian Organic Chemists and Their Legacy in its chapter Nikolai Nikolaevich Zinin and His Chemistry (Lewis, 2012, pp. 42-46).

Biographic notes

Zinin's biographic notes may be found in some German, Russian, Rumanian, U.S.A., and British publications between 1874 and 2002. The following authors have written these notes: 1) Carl Frederking in German in the book published in Gòttengen (Frederking, 1874, pp. 271-272); 2) Karl Schaedler, also in German, in the book entitled Biographisch-litterarisches Handwörterbuch der wissenschaftlich bedeutenden Chemiker (Biographical Literature Handbook of Scientifically Significant Chemists) (Schaedler, 1891, pp. 161-162); 3) Sir William A. Tilden (1842-1912) in the II edition of his book entitled The Progress of Scientific Chemistry in Our Own Times with Biographical Notices (Tiden, 1913, p. 183); 4) Iosif Borisovich Fainboim in the Russian-language journal Priroda (Nature) (Fainboim, 1940, pp. 84-86); 5) Max Solomon in the book in Romanian language entitled Lumini în retortă (Lights in the Retort), published in Bucharest (Solomon, 1962, pp. 190-193); 6) Carl Graebe in his Geschichte der organischen Chemie (History of Organic Chemistry) (Graebe, 1972/1920, p. 118); 7) Yurii I. Solov'ev in the book entitled Istoria khimii (History of Chemistry) (Solov'ev, 1983, p. 237); 8) David E. Lewis in his paper entitled The University of Kazan – Provincial Cradle of Russian Organic Chemistry published in the Journal of Chemical Education (Lewis, 1994, pp. 39-40); 9) Georg Schwedt in the book entitled Liebig und seine Schüller – die neue Schule der Chemie (Liebig and His Students – the New School of Chemistry) (Schwedt, 2002, pp. 196-197).

References to Zinin and the results of his experimental works

English chemist Julius Berend Cohen (1859-1935) in his Practical Organic Chemistry for Advanced Students, describing one of the methods of receiving aniline cited biographic data from the article in which Zinin published his discovery in 1842. He mentioned Zinin's name and the method of receiving benzoin (C6H5CO.CH(OH).C6H5) (Cohen, 1900, pp. 129-169).

British chemist Thomas Percy Hilditch (1880-1965) in his book entitled A Concise History of Chemistry Zinin's name relates to a short information about his discoveries: a) aniline from nitrobenzene; b) α-naphtylamine; c) benzidine from nitrobenzene, and d) azoxybenzene (Hilditch, 1911, p. 219).

British chemist John Cannell Cain (1871-1921) paid attention to Zinin's investigations related to azobenzide (azobenzene – C6H5.N2. C6H5) (Cain, 1920, p.92; Zinin, 1845, pp. 93-107).

American chemist Frank Clifford Whitmore (1887-1947), professor of organic chemistry at Northwestern University, mentioned Zinin's discovery in 1855 concerning the discovery of receiving allyl mercuric iodide (CH2=CH-CH2-HgI) in reaction between allyl iodide (3-iodopropene: CH2=CH-CH2I) and mercury (Hg) (Whitmore, 1921, p. 140).

Zinin's name appear several times in the article devoted to the Russian historian of chemistry B. N. Menshutkin written by the historian of chemistry Tenney L. Davis (1890-1949), professor at the Massachusetts Institute of Technology (U.S.A.) (Davis, 1938).

Brian Halton – chemist of the New Zealand Victoria University mentioned Zinin's name in his latest article devoted to A. M. Butlerov, published in the Chemistry in New Zealand (Halton, 2018).

Selected facts from Zinin's life

Nikolai Nikolaevich Zinin was born on August 13, 1812 in Shusha – Caucasic city. In 1820, he began secondary school in Saratov. In 1833, he graduated mathematic department in the Philosophical Faculty at the University in Kazan with the degree of mathematic sciences candidate (Fainboim, 1940, p. 84). He stayed in the university and been lecturer of the analytic mechanic for two years. Since autumn 1834, Zinin was also lecturer of the hydrostatics and hydrodynamics, and since August 1835 □ chemistry (Figurovskii & Solov'ev, 1857, p. 32). In 1835, he passed examinations and defended master's degree thesis in the physic-mathematical sciences.

On January 4, 1837 Zinin was appointed the adjunct in the department of chemistry and on September 25 of the same year he went to Germany for two years to improve his skills in chemistry. First, he stayed in Berlin. There, he attended lectures of mathematics by Enno Heeren Dirksen (1792-1850), Johann Gustav Lejeune Dirichlet (1805-1859), Daniel Christian Lehmus 1780-1863), and Martin Ohm (1792-1872). He attended also lectures of technology, geology, and geography by professors Heinrich Wilhelm Dove (1803-1879) and Heinrich Gustav Magnus (1802-1870) as well as lectures of mineralogy and crystallography by Gustav Rose (1798-1873). Zinin attended lectures of theoretical and practical chemistry by professors Eilhard Mitscherlich (2794-1863), and Heinrich Rose (1795-1864). He also visited factories situated in the city and suburbia. Then, Zinin worked in the laboratory of Justus von Liebig (1803-1873) in Giessen for one year. He also attended his lectures of the experimental chemistry.

In September 1838, his foreign training was prolonged for another year. He was permitted to go to France to attend lectures of chemistry and physics by prominent French professors and visit factories and all remarkable industrial objects.

In September 1839, Zinin completed his work in the Liebig's laboratory in Giessen and went to Paris. There, he attended lectures by Joseph Louis Gay-Lussac (1778-1850), lectures of organic chemistry by Jean Baptiste André Dumas (1800-1884). He also attended lectures of analytical chemistry by Théophile-Jules Pelouze (1807-1867) and worked in his laboratory (Figurovskii & Solov'ev, 1957, p. 46).

In June 1840, Zinin went to England, to London and stayed there for about three weeks. He travelled through the Netherland, Belgium, Germany, and Berlin, and returned to Russia in September 1840 (Figurovskii & Solov'ev, 1957, pp. 46-47). Zinin went to St. Petersburg, where he defended his doctor's dissertation entitled Compounds of Benzoyl and the Discovery of New Substances Belonging to the Benzoyl Family (Arbuzov, 1948, p. 51).

On June 5, 1841 Zinin was appointed the professor of the chemical technology at the Kazan University. Seven years later, he left Kazan and moved to St. Petersburg, where he was appointed professor of chemistry and physics at the Medico-Surgical Academy. He worked there to 1874. In June 1855, Zinin was elected the adjunct of the St. Petersburg Academy of Sciences. Ten years later, he became full academician.

In 3-5 September 1860, Zinin participated in the first ever international chemical conference in Karlsruhe (Germany) (Figurovskii & Solov'ev, 1957, p. 202). He was one of its

organizers and his name is listed among 43 other chemists, who on June 10, 1860 signed a letter with information about this conference sent to the chemists of the entire world (Anschütz, 1929, p. 194).

On 19 June 1862, Zinin was appointed a foreign member of the Chemical Society of London (The Jubilee of the, 1896, p. 195). On November 24, 1873, Zinin became member-correspondent of the Academy of Sciences of Paris, and on December 15 of the same year he was elected honorary member of the German Chemical Society in Berlin (Figurovskii & Solov'ev, 1957, p. 120).

Zinin's last years of life

Zinin was frequently ill. His health was deteriorated at the end of 1878. Pain of the legs made walking difficult. Moreover, he lost an appetite, suffered from bowel pain, frequent nausea and vomiting. Unexpectedly, his health improved a little at the end of December 1879. However, on the 5 February 1880 morning the disease attacked with double force. Nikolai Nikolaevich Zinin died on February 6, 1880. His funeral took place on February 9. In the group of the participants in this ceremony were among other: Dmitri Ivanovich Mendeleev (1834-1907), Aleksandr Mikhaylovich Butlerov (1828-1886), and Aleksandr Porfir'yevich Borodin (1833-1887), and also scientists from the St. Petersburg Academy of Sciences and Medico-Surgical Academy. Professors carried his coffin on their shoulders alternately with the students. In the front of the coffin two wreaths were carried. One from the professors of the Medico-Surgical Academy. The second from the medicine students. Funeral oration delivered Aleksandr M. Butlerov on behalf of the Russian Chemical Society. Aleksandr P. Borodin described Zinin as the scientist and social activist (Figurovskii & Solov'ev, 1957, pp. 120-121).

Zinin's memory was devoted session of the German Chemists Society, which took place in Berlin on March 8, 1880. August Wilhelm von Hofmann informed participants about Zinin's death. Hofmann is his short speech did not list his entire scientific achievements, reminded one epoch-making discovery – transformation of "nitro compounds into anilines". He said also that he met Zinin personally in Giessen in Liebig's laboratory in the end of the XIX century thirties. He emphasized Zinin's modesty and "excitation with which he moved towards the science". Finally, Hofmann said: "Even if Zinin did not does nothing more except nitrobenzene reduction to aniline, his name would be written with letters of gold in the history of chemistry (Hofmann & Tiemann, 1880, p. 440; Borodin & Butlerov, 1880, cited by Zinin, 1982, p. 209).

Fig. 1. Nikolai Nikolaevich Zinin (1812-1880) ("Academician N. Zinin," 1860/1870)

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