

## FERDINAND MÜNZ: EDTA AND 40 YEARS OF INVENTIONS

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### Abstract

This paper describes the life and accomplishments of the Austrian chemist Ferdinand Münz, who synthesized EDTA (ethylenediaminetetraacetic acid) in 1935. He was also the author of many patents in the textile field whose compounds are still in production today. Numerous textile treatment methods are ascribable to his name. He has been virtually unknown to the history of science because of persecution for his Jewish origins and his tragic personal issues.

### Biography The Viennese Period

Ferdinand Münz was born in Krakow on June 23, 1888, the son of Michael and Bertha Münz. At the time of his birth Krakow was one of the most populous cities of the Austro-Hungarian Empire. He had three siblings: Stefan, Ernest and Amelie. Little is known about Stefan. Amelie was sent to a concentration camp later in life, and she perished there (1). Ernest was a lawyer, and later emigrated to New York. Among the scanty biographical documents available there exists a picture of Joseph, Ferdinand's uncle, who was born in 1880 in Krakow and died in 1933 in Tel Aviv (2).

When Ferdinand was 10 years old, his family moved to Vienna (3). After the collapse and fall of the Austrian Empire (1919) he opted for Austrian citizenship; in a patent, in fact, we can read: "... and Ferdinand Münz, a

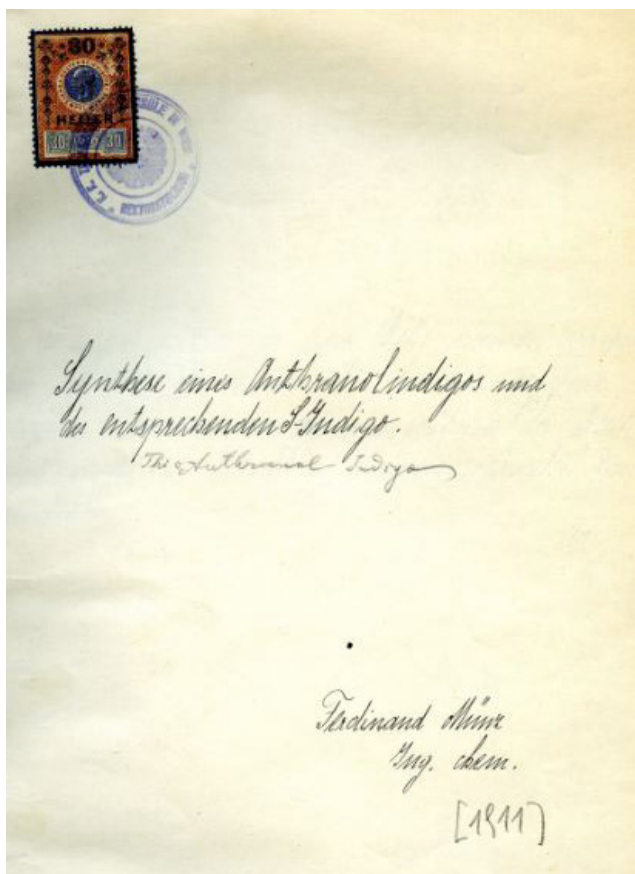
subject of the Austrian Republic" (4). In the archives of the Technical University of Vienna (TU Wien) (5) it is reported that his father, Michael, worked as a commercial employee in the same city in 1906. During his university studies, Münz moved three times. His last known address was Kaiserstraße 34, district of Neubau, Vienna. His first language was Polish, not German as his surname would suggest. The family had settled in the Kingdom of Galicia and Lodomeria (6) for several generations.

Münz attended the k. k. Staatsrealschule (7) of the V district of Vienna and he passed his matura (i.e. the final exam) on July 11, 1906. That same year he enrolled at Chemisch-technische Fachschule, faculty of technical chemistry, of the Technische Hochschule of Vienna, now TU Wien. TU Wien is still considered one of the most prestigious academic institution in the world. When he started his university studies, he had already fulfilled his military service (5).

On February 27, 1909, he passed the first Staatsprüfung (literally, state examination) and on July 13, 1910, he obtained the title of "Ingenieur," that we can read next to his name on the first page of the thesis (Figure 1). This title was necessary before he could apply for the Dr. techn. degree.

In October 1910, once his laboratory duties were complete, he began to work on his thesis. He first attempted to obtain the title of Dr. techn. (technical doctor, a title that does not correspond to Ph.D.) in July 1911. At

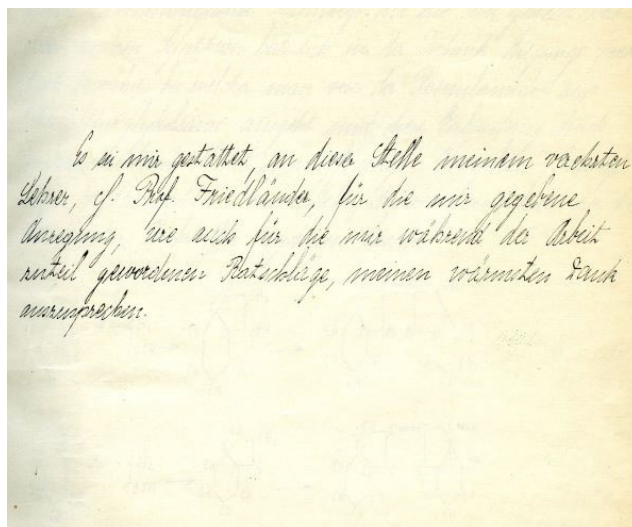
that time he failed, receiving inadequate grades from his teachers, Prof. Eugen Bamberger (1857-1932) and Prof. Hermann Suidas (1887-1973), son of Wilhelm Suidas (1853-1922), the future rector of the Technischen Hochschule Wien. On December 21, 1911, at age 23, he finally passed the exam and achieved the above-mentioned academic title (5).



**Figure 1.** First manuscript page of Ferdinand Münz's thesis (courtesy of TU Wien).

The thesis, “Synthese eines Anthranolindigos und des entsprechenden Schwefel-Indigo [Synthesis of an Anthranol Indigo and of the corresponding thioindigo]” (Figure 1), was focused on the chemistry of thioindigo, an organosulfur compound used to dye wool and cotton, related to the plant-derived dye indigo. An intellectual and technical inclination to the textile sector and dyeing processes were, then, already evident; these were subjects of study that would accompany him for the rest of his life. In the second page of his manuscript (Figure 2) there is an important acknowledgment to Prof. Paul Friedländer (1857-1923) that can be translated “At this point I would like to thank my esteemed teacher, Prof.

Paul Friedländer, for the suggestion of the topic and for his advice during the work.”



**Figure 2.** Dedication of the thesis to Prof. Paul Friedländer (courtesy of TU Wien).

Friedländer had studied chemistry at Königsberg in the laboratories of Carl Gräbe (1841-1927), and later in Strasbourg (8). There he became assistant to the future Nobel laureate Adolf von Baeyer (1835-1917), who was the first German chemist to synthesize indigo (9). Friedländer's research followed that of his teacher, focusing on the colors derived from indigo. For example, he discovered thioindigo, the subject of Münz's thesis. He also studied tyrian purple and *bolinus brandaris*, the type of mollusk from which it is possible to extract this dye. From 1895 to 1911 (the year when Münz finished his studies) Friedländer worked at the Technologisches Gewerbemuseum of Vienna, where he became chief of the chemistry department. In 1911 he moved to Germany and he began to work at the University of Darmstadt (10).

It is interesting to note, at this point, that the *fil rouge* linking these important scientists is indigo dye and its derivatives. We can therefore also include Ferdinand Münz in this group of inventors: although he was no longer involved in dye synthesis, he kept studying how to set them on fabric.

### First Years in Germany

After Münz obtained his degree, he moved to Viersen, near Düsseldorf, and he started working for Pongs und Zahn. On May 1, 1914, he moved to Leopold Cassella & Co., a company that was involved in textile chemistry and the pharmaceutical and cosmetic industries, and which

later merged into I. G. Farben. He then worked at the main plant in Fechenheim. At the outbreak of World War I he was enlisted in the Austrian Army, and in 1916 he was seriously wounded. When he returned to work for Cassella & Co., Münz dedicated himself to problems related to the research and synthesis of dyes for textile use. Over the years he penetrated deeper and deeper into this field and in 1927 he was promoted and transferred to the main scientific laboratory of the company, directed by Georg Kalischer (1873-1938). During this time he could devote himself actively and exclusively to industrial research.

In 1921 he was domiciled in Haingrabenstrasse 10, in Fechenheim-On-The-Main (4, 11). Today this place is part of the town of Maintal (12). At the beginning of the seventeenth century Fechenheim was a small village. During the second half of the nineteenth century, with the arrival of Cassella & Co., the city's population grew, and in 1914 the company employed 3,000 workers. From 1938, Fechenheim was incorporated as a district in Frankfurt (13).

Thanks to the patents it is possible to follow Münz's movements. Until the 1940s the city of Frankfurt was listed as his place of residence (14). Other patents and sources, perhaps more specifically, cite Mainkur, or Fechenheim-Mainkur (11), which is an area located slightly north from the center of Fechenheim, where Leopold Cassella & Co. had its headquarters.

Some time after 1934-1935 (15), but surely by 1938, he lived in Reuterweg 57, Frankfurt. That address was found in a document of the American Jewish Joint Distribution Committee (JDC) that might indicate Münz's attempt to emigrate to the United States of America (16). The document contains as depository the Lt. Julien D. Goell (1912-1944) (17). The same address is found in a transit document to the Buchenwald concentration camp in 1938, where Münz had been interned from November 25 to December 1, 1938 (18).

Comparing data from the JDC archives (19), Goell's year of death and when Münz was released from Buchenwald, it is reasonable to think that Münz tried to flee in a period between the first months of 1939 and 1944, when the growing Jewish persecution was manifested in occupied Europe. From 1942 to 1945, as the tide of World War II was turning for the worse for the Axis forces, biographical and scientific information about Münz faded away until it ceased.

In sum, Münz lived first in Haingrabenstrasse, about 4 km away from the Cassella & Co. factory in Mainkur,

and later in Reuterweg, near the Farben headquarters in Frankfurt.

In the period in which he lived near Frankfurt, in the late thirties, Münz was married to Maria Ewald (1897-1964). A document dated June 20, 1950, reads, "Married with Maria Münz, born Ewald (aryan)" (18). They had two children: Ferdinand Münz (junior) and Ilse Münz (married Mörschel) (20).

He may have been released after only a month from Buchenwald and a month before the liberation of Theresienstadt thanks to his wife (21).

## The Golden Decade

The years 1930-1939 constituted the golden decade of this inventor in terms of productivity. It was his most profitable period in an otherwise dark era, since he synthesized numerous commercial compounds which are still in production today.

One of these is EDTA, for which Münz filed a patent in Germany in 1935. It was published in 1942 without a named inventor (22) because a judge did not recognize him as an important inventor, despite the fame and high esteem he received in those years (3). He filed a patent for the same compound in the United States in 1936, in an attempt to give greater visibility to his discovery (23). Münz was considered a Jew by the Nazis (24) because of his origins, and at the time, in Nazi Germany a Jew could not perform scientific research freely. Because of those persecutions and deprivations, several publications and patents appeared in the literature without his name. For example, a couple of patents (22, 25) were issued nameless and later mentioned in his obituary (3).

In the same years he met Otto Bayer (1902-1982) in Frankfurt. Together they produced several patents concerning wetting agents, dyeing processes and polyurethane synthesis. The first one (26) presumably dates to the first years after they met in Frankfurt. Otto was not related to the Bayer family of the company of the same name, but he worked there, and later for I. G. Farben after the two companies merged (27). Münz and Bayer had a long and fruitful collaboration on many patents, from the aforementioned until the 1960s (28).

## Theresienstadt

On February 18, 1945, Münz was interned in the concentration camp of Theresienstadt (29), in today's Czech Republic. It was used as a prison camp and as

a place of transit to the eastern “death camps.” He was released on April 9, 1945 (18). The camp was set free by the Red Army on May 8 of the same year.

Two years before the end of the World War II Arthur von Weinberg (1860-1943) was deported there, despite his old age, and he died there, at age 83. He was the co-owner of Cassella & Co. The causes of his death can be attributed to his old age and the horrible hygienic conditions following cholecystectomy surgery (30).

### After “the Hell” and Later Years

The Theresienstadt concentration camp was described by Otto Bayer as “the hell” in Münz’s obituary. After returning from it Münz kept working at Farben. In 1945 he lived in Kaiserstrasse 20, Frankfurt (18). At the end of that year he moved near Cologne, in the Stammheim district (28, 31). There he worked in the Farben offices in Leverkusen, where his colleague Bayer, who was entrusted with the direction of the main research laboratory, had been transferred. The two men were apparently friends and esteemed colleagues since they first met in the Cassella laboratories in Frankfurt. In fact, Bayer writes (32):

Since there were already personal ties between us at Mainkur [at the Cassella laboratories], Münz came to work at the end of 1945 at the main laboratory at Leverkusen, where he worked on a whole series of interesting problems.

During those years Münz came into contact with the future Nobel laureate Kurt Alder (1902-1958). In 1949 they published a paper on diene synthesis and additions (33). Alder became director of the Bayer research laboratories in Leverkusen, where he became interested in rubber manufacturing processes from butadiene. Later he also obtained a professorship at the University of Cologne (34). In the industrial field EDTA, or more specifically chelate iron Fe(II)EDTA, is used in the polymerization process of styrene butadiene rubber (SBR) (35). It is possible that Alder called Münz since he was among the most experienced chemists in the field.

On January 1, 1956, Münz retired to private life.

A passenger list (36) of the Dutch steam ship *Rijndam* (also known as *Ryndam*) suggests that he visited the United States. He or another Ferdinand Münz sailed from Rotterdam and arrived in New York on May 28, 1957. The document shows his name and surname, his nationality (Austria), his passport number (565009), and travel class (tourist). If this was Münz, it is very likely that

he went to his brother Ernest who lived in New York at the time. His stay in the US was however not permanent.

He went back to Europe and he passed away on August 16, 1969, in the city of Glashütten, West Germany, at the age of 81. He was buried in the Hauptfriedhof (main cemetery) of Frankfurt (20).

### Inventions

The publications and patents of Ferdinand Münz cover forty years: from the first publication (37) with Richard Haynn in 1922, to the last patent with Otto Bayer, dated 1964 (28).

According to Bayer (3) the fundamental inventions attributed to Münz include the registered trademarks Solidegal, Humectol, and Trilon, as well as studies on conservation and treatment of cotton and wool. Humectol and Solidegal are alkyl phenol ethoxylates (APEOs), modified phenols having the phenolic OH replaced with long aliphatic chains, such as  $\text{Ar-O-CH}_2(\text{CH}_2)_n\text{CH}_3$ . Usually they are surfactant agents. They are used as auxiliary chemical agents in the textile dyeing process. They are leveling agents, able to level out a dye on a fabric, without which the final product would be defective (unevenly colored). For example, Solidegal can be used in conjunction with indanthrene (38), the trade name for a group of synthetic organic vat dyes derived from anthraquinone. Trilon is nitrilotriacetic acid (NTA, Figure 3), a surfactant. It removes metal cations that could interfere with textile processes.

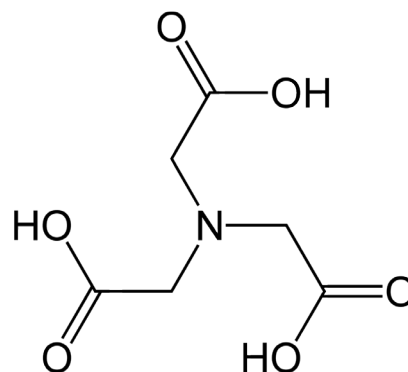


Figure 3. NTA structure

It is appropriate to add EDTA (Figure 4) and its salts to the list of Münz’s important inventions. The first registered trademark for EDTA was “Trilon B” by I. G. Farben and its successor BASF. Other commercial names are sequestrene (Ciba-Geigy), Versene (Dow), Chelest



(Chelest, Japan), Rexene (Grace, Sweden) and Hampene (Grace, USA).

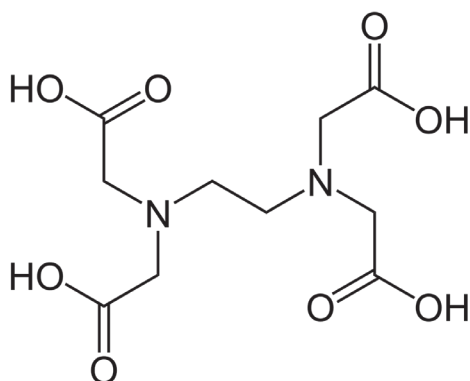


Figure 4. EDTA structure

### Synthesis of EDTA

Münz designed and synthesized EDTA as a substitute for citric acid in order to reduce the dependency of the German government on the import of foreign chemicals. He noted that an aminocarboxylic acid worked much better than citric acid as a chelating agent, and therefore reasoned that a polyaminopolycarboxylic acid would have worked even better. The main purpose was to remove calcium ions from water in order to improve the dyeing process on the fabric. His method involved the reaction between hot ethylenediamine with monochloroacetic acid and sodium hydroxide (23). The final product was, however, contaminated with sodium chloride (Figure 5).

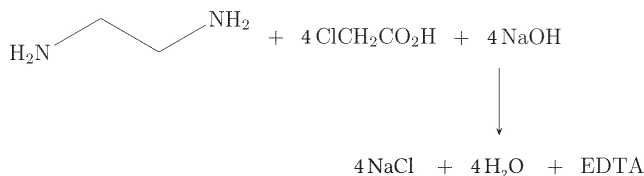


Figure 5. Münz's first reaction scheme to produce EDTA.

In 1941 Frederick C. Bersworth synthesized EDTA in the United States using a different and more convenient synthesis (39). This process is still used today. It is an alkaline cyanomethylation of ethylenediamine: disodium EDTA and ammonia are produced by reacting ethylenediamine with formaldehyde and sodium cyanide (Figure 6). Much of the  $\text{NH}_3$  evaporates, but part of it reacts with the starting materials to form NTA (Figure 3). To separate and purify the product, the solution needs acidification with sulfuric or hydrochloric acid, which al-

lows the formation of insoluble EDTA and consequently its separation from NTA, which remains in solution (35).

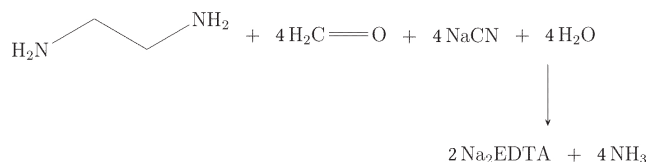


Figure 6. Bersworth's reaction scheme to produce EDTA.

In 1962 a third EDTA synthesis pathway was developed by John Singer and Mark Weisberg (40), that produces a very pure form of its disodium salt. In this process the cyanomethylation is separated from the hydrolysis. The strategy is to use ethylenediamine, formaldehyde and hydrocyanic acid, to produce EDTN, (ethylenedinitrilo)tetraacetonitrile (solid) which is then separated, washed and hydrolyzed with sodium hydroxide, to form  $\text{Na}_2\text{EDTA}$  and ammonia. By performing this synthesis, the majority of secondary reactions that lead to formation of impurities are removed. The yield is greater than 96%. This method, moreover, is well suited for synthesizing NTA with a high purity grade (and yield).

Other EDTA production processes not commonly used in the commercial field also exist, for example catalytic oxidation of tetra(hydroxyethyl)ethylenediamine (41).

### Uses of EDTA

EDTA is widely used in everyday life in formulations such as shampoos and shower gel. It is used to complex ions that contribute to the hardness of the water. In this way, no traces of salt remain on the skin or hair. It is also present as an additive and preservative agent in drugs, foods and cosmetics and as an antibacterial agent, since it manages to destroy the cell wall of bacteria, even resistant, gram-negative bacteria (35).

It is largely used in analytical chemistry for the analysis of water or for the quantitative identification of metal ions present in a solution. One widely used method is based on the Eriochrome Black T (EBT) as an indicator: at the equivalence point the chelating agent removes the cation from the  $\text{EBT-M}^+$  complex, making the solution change color from red to blue.

EDTA has also other uses in coloring and textile finishing, being found in many commercial dyes. Traces of contaminating ions from the fibers, reagents and water, can be introduced during the machining process, and

could otherwise alter colors. It is also very effective in products for cleaning metal surfaces, as it can solubilize oxides coated on the surface, improving the smoothness and uniformity of the metal.

### Chelating Therapies

At about the same time Bersworth developed his synthesis, Martin Rubin, a professor at Georgetown University, discovered that EDTA had biological effects on calcium homeostasis. This led his laboratory to begin using this molecule as an anticoagulant. In fact, EDTA can be used for diagnostic purposes in blood analysis (for example in Vacutainer<sup>®</sup> test tubes (42), as anticoagulant).

EDTA is widely used in the so called “chelating therapies.” Injected intravenously into the patient, it can complex cations of toxic metals present in the bloodstream. Subsequently, the metal-EDTA complex is expelled with the urine. It was used after the end of the World War II to counter widespread lead poisoning among prisoners or naval personnel employed in painting ship hulls (using lead-based colorants). Not long afterwards, Dr. Harry Foreman (University of Minnesota) studied and developed new therapies based on EDTA, both for lead poisoning (43) and for radioactive substances, such as plutonium (44). Since the 1950s the use of EDTA was also extended to the treatment of lead poisoning in children (45).

From 1954 Foreman led several studies on the risks to chelating therapies, determining the lethal dose of EDTA for humans. In a publication from 1956 (46) he and his coworkers reported that high doses of calcium disodium EDTA administered for small periods of time can lead to kidney disorders. This discovery led to the creation of new security protocols in therapeutic treatments with EDTA.

### Conclusion

Ferdinand Münz undoubtedly contributed to the progress and advancement of chemistry. He would have left a huge mark in the history of science and chemistry, if only he had not suffered the continuing persecution by the Nazis as a member of the Jewish religion. His name, in fact, was doomed to be eclipsed from the EDTA patent, thereby destroying all biographical traces from that document. Probably the fact of being married to an “Aryan” woman was an element on his favor, which allowed him to survive interments in concentration camps.

I would conclude this Austrian chemist's biography with the same sentence written in his obituary by Otto Bayer (47):

The memory of Dr. Ferdinand Münz must not disappear, because he had created something that will last forever, and he was a wise and intelligent man of great kindness and modesty.

As a concise reminder of the importance of his discoveries to the field of chemistry, it would be appropriate if his epitaph simply read “EDTA” in the same way the formula of entropy was carved on Boltzmann's tomb (48).

### Acknowledgments

I wish to recall all the people who have collaborated in the writing of this article with suggestions, criticisms and comments: they deserve my gratitude. Thanks to Dr. Marco Fontani, chemist and chemistry historian, University of Florence; Ludovico Pavesi, Politecnico di Torino; Jessica Lamantia, University of Illinois at Chicago; Dr. Nicolò Marnoni, chemist, for the part on APEOs; and the Austrian historian Dr. Paulus Ebner, TU Wien. Thanks also to all the people who answered my numerous emails and letters: Desiree Wolny, archivist of Bayer; the historian Stephan Münz; Ursula (Sue) Foster, for the history of Fechenheim; the library staff of the Polo Scientifico of the University of Florence; Cinzia Augelli, library “Arturo Graf” of the University of Turin; the Institut für Stadtgeschichte Frankfurt; Joerg Hausmann, Grünflächenamt Frankfurt; the association Yad Vashem for the copious material provided to me; the Theresienstadt Martyrs Remembrance Association; and the civil registers and cemeteries staff of Frankfurt, Cologne, Maintal, Vienna, and Krakow. Finally, thanks to my parents for their ongoing support.

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47. Die Erinnerung an Ferdinand Münz darf nicht verblassen, denn er hat Bleibendes geschaffen und war ein kluger, weiser Mensch von großer Güte und Bescheidenheit (3).
48. That is,  $S = k \log W$ .

### About the Author

Matteo Paolieri was born in Pistoia, Italy, in 1996. He is currently studying at the faculty of chemistry at the University of Florence. He is an independent scholar in history of chemistry. He is also a member of the *Gruppo Nazionale di Fondamenti e Storia della Chimica* (Italian National Society of History of Chemistry, [www.GNFSC.it](http://www.GNFSC.it)) and a journalist specialized in history of science for the popular Italian blog *Italia Unita per la Scienza* ([www.italiaxlascienza.it](http://www.italiaxlascienza.it)).

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