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## **Curriculum Vitae and Scientific Research of Ulrich Hofmann (1903 - 1986)**

Ulrich Hofmann was born in 1903 in Munich as the son of Karl Andreas Hofmann (1870 - 1940) who led the Group of Inorganic Chemistry in the Institute of Chemistry. The head was Adolf von Baeyer (1835 - 1917). Seven years later K. A. Hofmann accepted the chair of Inorganic Chemistry at the Technical University in Berlin.

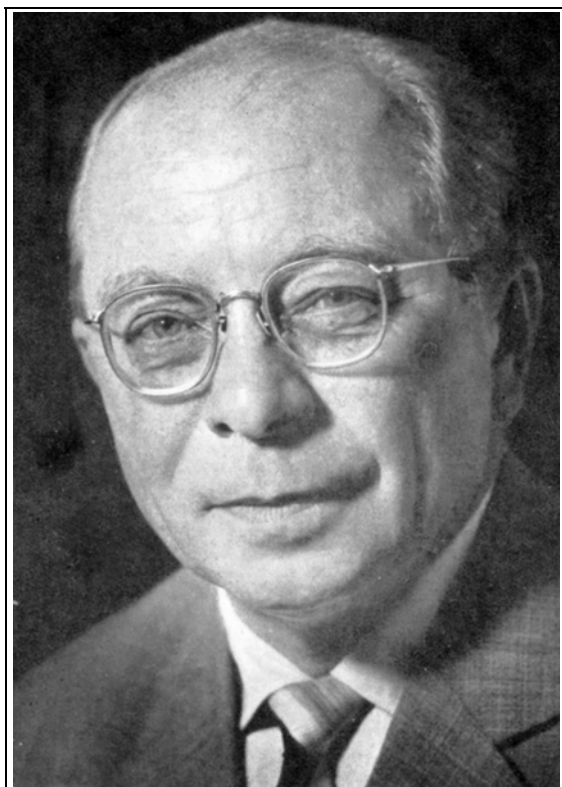
Ulrich Hofmann received the Diploma degree in civil engineering in 1925 in Berlin and the Ph. D. in 1926 for studies on lustrous carbon (HOFMANN, 1926). The postdoctoral lecture qualification (Habilitation) in 1931 was based on the contribution "Die Bildung und Konstitution der Graphitsäure (formation and structure of the graphitic acid)". In 1937 Ulrich Hofmann became Professor and Head of the Chemistry Institute of the University in Rostock and taught in inorganic and physical chemistry.

His time in the German army was very short because he was exempted to work on subjects important for the wartime economy. In 1942 Ulrich Hofmann became the head of the Institute of Inorganic and Analytical Chemistry at the Technical University (Technische Hochschule) in Vienna. Just before this change he bought one of the first electron microscopes which he transported to Vienna. Ulrich Hofmann had to leave Austria in 1945 and worked a certain time (1947/48) as a gardener for Süd-Chemie Comp. in Moosburg, Bavaria.

He started the second part of his academic carrier in 1948 when he was asked to teach inorganic and physical chemistry, later also technical chemistry, at the Philosophical and Theological High School (Philosophisch-Theologische Hochschule) in Regensburg. As most of the universities and technical universities in Bavaria were destroyed, the institution in Regensburg was transformed into an university-type high school. One of the problems was to find laboratories for the

students of chemistry. The solution was simple: the students with their teachers completed rooms and workplaces by themselves. Among the students were Armin Weiss who eventually established the practical course in physical chemistry, Hans-Peter Boehm and Theobalt Nemetschek who made their thesis under the supervision of Ulrich Hofmann.

When Ulrich Hofmann moved to the Eduard-Zintl-Institute of Inorganic and Physical Chemistry of the Technical University in Darmstadt (1951), he received an electron microscope from the Deutsche Forschungsgemeinschaft (DFG). In 1952 Ulrich Hofmann became the first President of the German Society for Electron Microscopy (Deutsche Gesellschaft für Elektronenmikroskopie).



Ulrich Hofmann

He did not follow calls of the Technical Universities in Aachen and Vienna (1956) but accepted the position as the Head of the Institute of Inorganic Chemistry at the University of Heidelberg. He retired 1971. One of the last conferences he visited was a joint General Meeting of the Kolloid-Gesellschaft and the German Clay Group (DTTG) in Kiel (1977).

Ulrich Hofmann received many awards. In 1955 he was awarded with the Alfred-Stock-Prize of the Society of German Chemists (GDCh). He became a Member of the Academy of Sciences in Heidelberg (1961) and Leopoldina in Halle (1962). The German Ceramic Society honoured him with the Hermann-Seger-Medal (1964). In 1965 he was awarded with the Wolfgang-Ostwald-Prize of the Kolloid-Gesellschaft in

recognition of his studies on the intracrystalline swelling of montmorillonite, polyphosphates, and collagen. In 1968, he received the Dr. h. c. from the University Munich (BOEHM, 1987; BENEKE, 1995).

When Ulrich Hofmann began to study the lustrous carbon (Glanzkohlenstoff), only graphite and diamond were distinguished. In 1917, Peter Debye (1884 - 1966) and Paul Scherrer (1890 - 1969) studied small particles of carbon which were considered amorphous by X-ray techniques. They observed similar X-ray diffraction patterns as in graphite and concluded that carbon forms only two modifications, graphite and diamond (DEBYE AND SCHERRER, 1917). Presently, four modifications are known: graphite, diamond, lonsdaleite, and fullerenes.

Lustrous carbon (Glanzkohlenstoff) forms by pyrolysis of hydrocarbons at low pressure on the smooth surfaces of materials like porcelain and quartz (pyrocarbon).

Hofmann applied X-ray diffraction which was not very abundant and known in this time (around 1925). He used the equipment of the Physical Chemistry Group of Karl Hermann at the Technical University (Technische Hochschule) in Berlin. Lustrous carbon appeared to be intermediate between amorphous carbon and graphite. The structure of the layers is as in graphite but the superposition of the layers is not regular, the layers are shifted and rotated one against the other. U. Hofmann and K. A. Hofmann used the term "Kreuzgitter" before the term "turbostratic disorder" from B. E. Warren in 1930 became more popular (HOFMANN AND HOFMANN, 1926; WARREN, 1990). Other types of carbons showed similar structures. U. Hofmann derived the dimension of the coherently scattering domains from the broadening of the X-ray reflections.

Graphite oxide (Graphitoxid) was known as "graphitic acid". Sir B. C. Brodie Jr. (1817 - 1880) reported in 1859 the molecular mass of graphite and graphitic acid. Graphitic acid is obtained by oxidation of graphite, e. g. by potassium chlorate. Its composition is variable, and the water content depends on the water vapour pressure. The X-ray studies of Ulrich Hofmann revealed that graphite oxide also shows a turbostratic disorder with distances between the layers being higher than in graphite or "Glanzkohlenstoff". He also observed that the distance between the layers increased with increasing water vapour pressure. The publication in 1932 was the first description of the intracrystalline swelling (innerkristalline Quellung) (HOFMANN ET AL., 1932).

Ulrich Hofmann described different types of carbons which were obtained by decomposition of graphite oxide, disproportionation of carbon monoxide on iron catalysts, and decomposition of hydrocarbons on iron (HOFMANN, 1928a,b). He observed the high adsorption of phenol by these carbons with high specific surface areas (HOFMANN ET AL., 1931). This was the reason of his interest in interface phenomena.

Further studies on the growth of graphite crystals at high temperatures brought him into contact with the industry, first with Siemens-Plania in Berlin, after the Second World War with Sigri Elektrographite in Meitingen near Augsburg. U. Hofmann always liked to cooperate with industry, not only from a financial point of view but also to promote exchange between research and application.

A decisive step was the electron microscopic observation of carbon black (Ruß) together with Manfred von Ardenne (1907 - 1997). They observed that carbon black consists of chains of spherical carbon particles (ARDENNE, HOFMANN, 1941).

In 1938 U. Hofmann and Walter Rüdorff described the intercalation of nitric and sulfuric acids into graphite with formation of positively charged graphite layers and intercalated anions. They also described the one-dimensional swelling which accompanies this process and the different types of stages. Depending on the degree of oxidation layers with intercalated anions alternate with one, two, three etc. unreacted layers (HOFMANN, RÜDORFF, 1938; RÜDORFF, HOFMANN, 1938).

It was in the Akademische Alpenverein zu Berlin (the club for walking and mountain climbing in the Alps) where U. Hofmann met Kurd Endell (1887 - 1945). Kurd Endell led a laboratory of building technology (since 1919) and was very much interested on clay minerals. He asked U. Hofmann to make X-ray diagrams of several clay minerals. U. Hofmann used a Debye-Scherrer camera with the exit-tube replaced by a narrow screen of lead so that the reflections at small angles could be easily observed, with  $\text{CuK}\alpha$  up to d-values of 38 Å. In the usual Debye-Scherrer cameras the large hole in the film for the emission screen made impossible to detect such high spacings.

Studying the montmorillonite samples of Kurd Endell he observed that the d-value of the first reflection was dependent on the water content: this was the first observation of the intracrystalline swelling of a clay mineral (HOFMANN, ENDELL, WILM, 1933).

Ulrich Hofmann further observed that montmorillonite heated to 700-800 °C did not longer swell and that the X-ray powder diagram was very similar to that of pyrophyllite (WILM, HOFMANN, ENDELL, 1934) the structure of which was proposed, in 1930, by Linus Carl Pauling (1901 - 1994) (PAULING, 1930). The two (hk) bands also corresponded to reflections of kaolinite. The structure of kaolinite was determined by J. W. Gruner in 1932 (GRUNER, 1932). Based on this knowledge Hofmann and Endell developed 1933 the structure model of montmorillonite (HOFMANN, ENDELL, WILM, 1933, 1934). The contribution "Kristallstruktur und Quellung von Montmorillonit. (Das Tonmineral der Bentonittone)" by Ulrich Hofmann, Kurd Endell and Dietrich Wilm 1933 was one of the most cited papers in clay science (BOEHM, 1987; BENEKE, 1995; BERGAYA, BENEKE, LAGALY, 2001).

A few years later, Cornelis Hendrik Edelman (1903 - 1964) (1936) and J. C. L. Favejee proposed (1940) a different structure with the corners of neighbouring  $(\text{Si,Al})\text{O}_4$  tetrahedra pointing up and down so that the cation exchange capacity could be related to acidic silanol groups (EDELMAN, 1936; EDELMAN AND FAVEJEE, 1940). Ulrich Hofmann proved that this model was not in agreement with the amount of water bound and the basal spacings. In agreement with studies of C. Edmund Marshall (1935) and Walter Noll (1907 - 1987) in 1936 U. Hofmann (1936) explained the cation exchange as a consequence of the Si-by-Al substitution in the tetrahedral sheet and cation defects in the octahedral sheet (MARSHALL, 1935; HOFMANN, BILKE, 1936; NOLL, 1936).

Bentonite was used in foundry moulding sands but only the bentonite of Wyoming was suitable; bentonites from Germany could not be used in this application. U. Hofmann and K. Endell studied many bentonites and detected that the decisive difference was the type of the interlayer cation, mainly sodium in the Wyoming bentonites but calcium (and also magnesium) in the bentonites from Germany. By adding soda Hofmann and Endell transformed the German bentonites into materials similar to the Wyoming bentonites which could be used in foundry

moulding sands. As described in the German Patent (1934) and the British Patent (1935) the calcium bentonite and 2 - 2.5 % (w/w) sodium carbonate ( $\text{Na}_2\text{CO}_3$ ) were dispersed in water. The dispersion was allowed to stand for a certain time, then the sediment was washed, separated and dried (HOFMANN, ENDELL, 1935).

U. Hofmann and K. Endell changed the process of soda-activation in a way it is still used today: The bentonite was mixed with the (dry) sodium carbonate, then water was added to reach a solid content of 20-30% (w/w). The mass was homogenized by kneading and grinding to obtain the soda-activated bentonite (Patents in 1936) (BENEKE, LAGALY, 2002).

In 1937 Ulrich Hofmann and Edmund Maegdefrau described a mica-type clay mineral from Sárospatak in Hungary. Because of its small particle size it contained larger amounts of water and showed plasticity. Maegdefrau and Hofmann (1937) wrote:

*„Ein besonders reines Material erhielten wir in dem sogenannten Kaolin von Sárospatak (Ungarn), auf den uns Herr Mathyasovszky [Chemieingenieur László Mattyasovszky (10.02.1912 - 17.02.1992)] aufmerksam machte. Dieses Mineral, das eine rein weiße Brennfarbe besitzt, wird in Ungarn wegen seiner hohen Plastizität und Trockenfestigkeit zur Verbesserung der Formbarkeit der Porzellanmasse verwendet und ersetzt zugleich durch seinen Kaligehalt Feldspat. Da die hier beschriebene Untersuchung zeigt, daß das Mineral glimmerartige Struktur besitzt, nennen wir es im folgenden „Glimmer von Sárospatak“.*

Furtheron:

*„1. Glimmer von Sárospatak: Dieses Mineral kommt in etwa waggongroßen Nestern im Liparit im Hegyalgebirge [=Tokaj mountains], im Nordosten von Ungarn vor. Es ist rein weiß und derbstückig. Die feinsten Teilchen lassen bei 200facher Vergrößerung noch keine Kristallform erkennen.*

*Durch Aufschlännen und Sedimentieren des fein gepulverten Materials in destilliertem Wasser konnte die im Röntgenbild glimmerartige Substanz vollständig von ihren geringfügigen Verunreinigungen Kalzit und Quarz befreit werden. Weder röntgenographisch noch mikroskopisch waren nach der Reinigung fremde Bestandteile erkennbar“.*

László Mattyasovsky (1885 - 1935), the uncle of László Mattyasovsky (1912 - 1992; Kossuth-Prize-Laureate) was designer and painter in the famous porcellain industry of Vilmos Zsolnay in Pécs. Ceramics of this company are seen at many buildings in Hungary, especially in Budapest (Mathias church, covered market, buildings of the Institute of Geology). In 1999 Zsolnay Porcelángyár was split into three companies (VICZIAN, 2002; HARASZTI, 2002).

U. Hofmann and E. Maegdefrau observed a relatively high cation exchange capacity which is related to the cations at the external surface. The potassium ions in the interlayer spaces are not exchangeable but are acting as fluxes in ceramic processes. The Russian author I. D. Sedletzky proposed 1940 the mineral to be

named “Sarospatakit” (SEDLITZKY, 1940) but Maegdefrau (MAEGDEFRAU, 1941) and Hofmann, Endell and Maegdefrau (HOFMANN, ENDELL, MAEGDEFRAU, 1943) used “Sarospatit”. However, the name “Illite” (from the state Illionois in USA) became more popular. Today the mineral is known as Illite from Füzérravány (VICZIÁN, 2000; BERGAYA, BENEKE, LAGALY, 2001).

Very fruitful was the conception of clay mineral aggregates in form of the house-of-cards which Ulrich Hofmann proposed first in 1942 (HOFMANN, 1942, 1952, 1956). This idea certainly developed during card games which Ulrich Hofmann liked very much. It was shown that the volume occupied by the clay mineral platelets did not change when a thixotropic gel was freeze-dried (WEISS, FAHN, HOFMANN, 1952).

An important result of Hofmann’s studies on the swelling of montmorillonite was that the maximum amount of interlamellar water depends on the type of the interlayer cation. Even still more significant was the discovery that the montmorillonite layers separate completely when the interlayer cations were  $\text{Li}^+$  or  $\text{Na}^+$  and the salt concentration was low, typically below 0.1 M for NaCl. This process (delamination, exfoliation) is one of the most striking properties of smectites. U. Hofmann explained this behavior on the basis of the DLVO theory (Derjaguin-Landau-Verwey-Overbeek-Theory) (FRÜHAUF, BURCK, HOFMANN, 1962; HOFMANN, 1962a, 1964).



Ulrich Hofmann ahead (1963)

It is understandable that Ulrich Hofmann in several papers studied the ability of clay masses to be moulded or shaped at certain water contents. He explained the plasticity of kaolins by the attraction between the kaolinite particles in the presence of divalent cations, mainly  $\text{Ca}^{2+}$  or  $\text{Mg}^{2+}$ . When these cations are replaced by sodium ions after addition of sodium carbonate or oxalate (which leads to precipitation of the earth-alkali cations as carbonates or oxalates), the plastic mass of kaolin at the same water content liquefies to a slip which is used for the production of porcellain in moulding forms of porous gypsum (CZERCH, FRÜHAUF, HOFMANN, 1960; HOFMANN, BURCK, 1961; FRÜHAUF, BURCK, HOFMANN, 1962).

The influence of the type of cations on the mechanical properties of the green and fired bodies was studied by U. Hofmann in a series of papers (HOFMANN, 1949; HOFMANN, CZERCH, SCHARRER, 1958; HOFMANN, 1959; HOFMANN, 1962a,b; HOFMANN ET AL, 1962).

U. Hofmann and A. Hausdorf described in 1942 the effect of interlayer spaces with different water contents and thicknesses within the crystals, i. e. the interstratification on the X-ray powder diagrams.

Another well-known effect was observed by Ulrich Hofmann and Richard Klemen in 1950 (Hofmann-Klemen-Effect). The experiments were done 1944 in Vienna.  $\text{Li}^+$  montmorillonite lost the cation exchange capacity and the swelling behavior when it was heated to 125 °C. Hofmann and Klemen explained this effect as a consequence of the diffusion of the lithium ions into the octahedral sheet of the montmorillonite layer (HOFMANN, KLEMEN, 1950).

Ulrich Hofmann also studied many other processes and systems. In 1933 and 1934 Max Fink and U. Hofmann reported on tribo-chemical reactions, in particular on the oxidation of metals during friction and the consequences for the abrasion of gear-wheels (FINK, HOFMANN, 1933a,b; FINK, HOFMANN, 1934a,b, BENEKE, 2000).

The use of salt to stabilise sand streets not covered with asphalt (often used in the North-European countries) was proposed by Ulrich Hofmann (HOFMANN, 1962c; HOFMANN, SCHEMBRA, 1967). He also recommended the use of clays to stabilise bitumen emulsions (HOFMANN, MATULL, 1968; MATULL, HOFMANN, 1968) and studied the effect of inorganic fillers on the properties of rubber and polymers (KUNOKOWSKI, HOFMANN, 1955).

U. Hofmann begun the investigation of ultramarine in Rostock but completed it in Heidelberg. The color is mainly due to the unusual anions  $\text{S}_3^-$  (blue) and  $\text{S}_2^-$  (yellow) which are stabilised in the zeolitic structure (HOFMANN, HERZENSTIEL, 1964; HOFMANN, 1968; HOFMANN ET AL, 1969; SCHWARZ, HOFMANN, 1970).

The studies of Ulrich Hofmann in Heidelberg were devoted to the ancient techniques of production and decoration of greek, mainly attic vases and Terra Sigillata, the abundant roman everyday dishes. It was a coincidence that many Terra Sigillata ceramics were dug out very near the Institute of Inorganic Chemistry in Heidelberg. With his permanent interest on the classical antiquity Ulrich Hofmann was fascinated by these studies (HOFMANN, 1962d; HOFMANN, 1965; HOFMANN, THEISEN, 1965a,b; HOFMANN, 1966; HOFMANN, THEISEN, YETMEN, 1966). One can imagine that the detection of the ancient techniques of decoration of the vases was the highlight at the end of Ulrich Hofmann's academic carrier (BENEKE, LAGALY, 2003).

#### Acknowledgement

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