Sophus Mads Jørgensen

A DANISH PLATINUM METALS PIONEER

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It is as Alfred Werner’s primary scientific adversary and the chemist whose work on co-ordination compounds provided the experimental basis for Werner's revolutionary co-ordination theory that Jørgensen is usually remembered. Yet in his work on what were then called “molecular compounds” he made valuable contributions to our basic knowledge of rhodium and platinum complexes.

Although unjustly neglected today, Sophus Mads Jørgensen was an important chemist during his lifetime, and his work aroused great interest among his contemporaries. In 1906 the French Académie des Sciences presented him with its Lavoisier Medal. Early in 1907, the first year in which Alfred Werner was nominated for the prize, Jørgensen received two nominations for the prize – by 1906 Nobel chemistry laureate Henri Moissan (1852–1907) and Jean-Gaston Darboux (1842–1917) of the Institut de France, Académie des Sciences (1). In his nomination Darboux cited “parmi les travaux de M. Jørgensen l’ensemble de ses recherches sur les bases ammoniées métalliques et plus particulièrement sur les bases du platine” (2). Moissan died later that year, and Jørgensen was never renominated.

The Werner-Jørgensen Controversy

Today most chemists, regardless of their fields of specialisation, are acquainted with the nature and importance of the work of Alfred Werner (1866–1919) (3–5). Indeed, almost every chemistry student is now introduced to at least the essentials of his co-ordination theory. Yet, except
for those who have had occasion to delve into the study of complex compounds, Jørgensen's name remains entirely unknown.

Few realise that Werner's new and revolutionary theory was based upon experimental data carefully and painstakingly accumulated over a number of years by Sophus Mads Jørgensen, Professor of Chemistry at the University of Copenhagen. It is perhaps not an exaggeration to say that Werner's theory might never have been proposed had not Jørgensen's work provided the observations requiring explanation.

Except for some early isolated organic and inorganic research, Jørgensen devoted himself exclusively to investigating the co-ordination compounds of cobalt, chromium, rhodium and platinum; this important work was carried out between 1878 and 1906. Jørgensen created no new structural theory of his own. His interpretations of the luteo (hexaammines), purpureo (halopentaammines), roseo (aquapentaammines), praseo (trans-diallotetraammines),violeo (cis-diallotetraammines), croceo (trans-dinitrotetraammines), flavo (cis-dinitrotetraammines) and other series of co-ordination compounds were made in the light of his logical extensions to and modifications of the famous chain theory of the Swedish chemist Christian Wilhelm Blomstrand.

For fifteen years Jørgensen's views remained the most acceptable of the numerous theories advanced to explain the properties and reactions of the so-called molecular compounds, which were not explicable in terms of the contemporary valence theory.

In 1893 Alfred Werner, an unknown twenty-six-year-old Privat-Dozent at the Eidgenössisches Polytechnikum in Zürich, challenged the old system with his radically new co-ordination theory (4b). The ensuing controversy between Jørgensen and Werner constitutes an excellent example of the synergism so often encountered in the history of science (6). Jørgensen's strong sense of history caused him to view Werner's new theory as an unwarranted break in the development of the theories of chemical structure. He regarded it as an ad hoc explanation insufficiently supported by experimental evidence.

The controversy between Jørgensen and Werner over the constitution of metal-ammine complexes provides us with an excellent illustration of the synergism so often encountered in the history of science. During the course of this competition, conducted without any trace of jealousy or rancour, each chemist did his utmost to prove his views, and in the process a tremendous amount of fine experimental work was performed by both. Although not all Jørgensen's criticisms were valid, in many cases Werner was forced to modify various aspects of his theory. However, the basic postulates were verified in virtually every particular.

Although Werner's ideas eventually triumphed, Jørgensen's experimental observations are in no way invalidated. On the contrary, his carefully performed experiments have proved completely reliable and form the foundation not only of the Blomstrand-Jørgensen theory, but also of Werner's theory. From the very beginning of the controversy, Werner continually acknowledged his great debt to the older man. For example, in 1913, on his return to Zürich from Stockholm where he had received the Nobel prize, Werner, addressing the Kemisk Forening in Copenhagen, acknowledged the important role that Jørgensen's experimental contributions had played in the development of the co-ordination theory. Unfortunately, Jørgensen's grave and final illness prevented the meeting of the two great adversaries.

Jørgensen's Life

Sophus Mads Jørgensen was born in Slagelse, Denmark on July 4, 1837, the son of master tailor Jens Jørgensen (1788–1862) and Caroline Grønning Jørgensen (1794–1855). His early schooling was obtained at Slagelse and later at the Søren Velvillie where his interest in chemistry was awakened by F. Johnstrup. This dedicated teacher allowed the older students to work in his own private laboratory on Sundays, and Jørgensen eagerly availed himself of the opportunity. After his graduation in 1857, Jørgensen entered the University of Copenhagen, where he
continued his studies with the encouragement of Professor Edward Augustus Scharling. In 1863 he received his Master of Science degree in chemistry. He then worked at the university until 1864 becoming Scharling's assistant.

Three years later he became a manager of the chemical laboratories and an instructor at the Polytekniske Laereanstalt (Polytechnic School). In 1869 he received his doctorate with the dissertation, "Polyiodides of Alkaloids". In 1871 he married Louise Wellmann, and in the same year he became a lecturer at the university. In 1887 he became Professor of Chemistry, a position which he held until his retirement in 1908. Jørgensen died on April 1, 1914 and is buried in the Holmen Cemetery in Copenhagen (6-10).

As a research worker, Jørgensen was methodical, deliberate and careful. Although he could have delegated much routine work to assistants, he insisted on personally performing all his analyses. He reserved one day per week specifically for this task (11). In spite of his passion for perfection, his output was tremendous, and we are indebted to him for many of the basic experimental facts of co-ordination chemistry.

Like many famous chemists, Jørgensen was not content merely to make original contributions in the laboratory but was also keenly interested in the historical development of chemistry, especially in Denmark. His studies in this field are characterised by the same enthusiasm and thorough workmanship shown in his laboratory investigations. Unfortunately, few of these studies have been published.

Considering the volume and quality of Jørgensen’s work, it is difficult to see how he found time to participate in the scientific and administrative activities that he undertook. He also served as an associate editor of the Zeitschrift für Anorganische Chemie from its founding in 1892 until his death.

Jørgensen also held many administrative positions. From 1871 until his death he was a member of the Board of Directors of the Rosenborg Brøndanstalt. From 1872 to 1908 he was technical consultant for the Customs Administration and Harbour Administration. In 1885 he was elected a member of the Board of Directors of the Carlsberg Foundation, and in 1909 he became chairman, a post which he held until 1913 when illness forced him to resign. From 1886 until his death he was a member of the Direktion of the Selskabet for Naturlaerens Udbredelse. In 1889 he was elected chairman of the Carlsberg Laboratories. He was also a member of commissions for sugar (1870), pharmacopoeia (1889) and gypsum (1893).

Jørgensen’s Research

Jørgensen’s research, which resulted in 76 articles and 19 books (8b) can be divided into seven groups. The first (1866–1878) includes a few isolated papers and his work on alkaloid polyiodides (the subject of his doctoral dissertation), while the other six (1878–1906) constitute the work for which he was famous – his investigations of metal-ammine complexes. These studies appeared in various publications of the Videnskabernes Selskab, then in the Journal für Praktische Chemie, and after 1892 in the newly founded Zeitschrift für Anorganische Chemie. Indeed, the fortunate subscriber had an opportunity to follow the scientific controversy between Jørgensen and Werner.

Platinum Compounds

Jørgensen’s first work on inorganic co-ordination compounds involved platinum rather than the cobalt-ammines which figured so prominently in the Werner-Jørgensen controversy. In the first article Jørgensen described platinosoplatinic oxide, Pt₂O₄, prepared by fusing Na₂[PtCl₄] with Na₂CO₃ and washing the black residue with water, dilute HNO₃, and aqua regia (12). The “compound”, which is reduced to platinum black by formic acid, hydrogen, or coal gas, was later shown to be a mixture of PtO and PtO₂ (13). In the second article Jørgensen described yellow Ag₂[PtCl₄] (formulated as 2AgCl·PtCl₄), prepared by metathesis of AgNO₃ and H₃[PtCl₄]; the compound reacted with water:

\[ \text{Ag}_2[\text{PtCl}_4] + 2\text{H}_2\text{O} \rightarrow \text{H}_2[\text{PtCl}_4(\text{OH})_2] + 2\text{AgCl} \downarrow \]

He also prepared PtCl₆(OH)₂ (the anhydride
of $H_2[PtCl_2(OH)_2]$ and $H_2[PtCl(OH)_3]$ (prepared only in solution and formulated as a solution of PtCl(OH)$_3$ (14). In his research on various complex cations of different metals Jørgensen routinely prepared $[PtCl_2]^{-}$ and $[PtCl_3]^{2-}$ salts, but these works need not be considered here.

In his first article on the platinum(II)-ammines Jørgensen attempted to elucidate the constitution and isomerism of compounds (15). In this classic work he prepared the cis and trans isomers of $[PtCl_2(NH_3)_2]$ (16), which he formulated in terms of the Blomstrand chain theory as:

$$\begin{align*}
\text{cis-NH}_3\text{NH}_3\text{Cl} & \quad \text{PtCl} \\
\text{Pt} & \quad \text{trans-NH}_3\text{Cl} \\
\text{cis-NH}_3\text{Cl} & \quad \text{PtNH}_3\text{Cl} \\
\text{Pt} & \quad \text{trans-NH}_3\text{Cl}
\end{align*}$$

He used the reactions of these isomers with pyridine to decide which formula belongs to the one and which to the other series, preparing in the course of this proof the following compounds (modern formulae in parentheses): platosemidipyrindine chloride ($cis-[PtCl_2py_2]$) (where py is pyridine) (17); platospyridine chloride ($trans-[PtCl_2py_2]$) (17); platodipyrindine chloride ($[Pt_{py_2}]Cl_23H_2O$); platopyryidine chloride-platinos chloride ($[Pt_{py_2}][PtCl_2]$); platopyryidineammine chloride, $\alpha$ ($cis-[Pt_{py_2}(NH_3)_2]Cl_2$); platopyryidineammine chloride, $\alpha$ with platinos chloride ($cis-[Pt_{py_2}(NH_3)_2]Cl_2$); and platopyryidineammine chloride, $\beta$ ($trans-[Pt_{py_2}(NH_3)_2]Cl_2$). He also prepared the chloride, iodide and tetrachloroplatinate(II) of the last compound and numerous similar compounds of PtCl$_2$ with CH$_2$NH$_2$, C$_2$H$_4$NH$_2$ and n-C$_2$H$_4$NH$_2$ (15). Ironically, in many cases Jørgensen's work bore the seeds of the Blomstrand-Jørgensen theory's destruction, for many of the compounds that were first prepared by Jørgensen later proved instrumental in demonstrating the validity of Werner's views. In the case now under consideration Werner used the above compounds and their reactions to prove today's generally accepted square planar configuration for platinum(II) (4c).

In the first of seven papers in the series "Ueber Metallamminverbindungen" Jørgensen was the first to prepare ethylenediamine complexes of platinum(II) and cobalt(III) and the first to recognise the bidentate nature of this organic ligand (18). He prepared platoseme-ethylenediamine chloride, ($[PtCl_2en]$) (where en is ethylenediamine); platoditetraethylammine chloride ($[Pt_{en_2}Cl_2]$); platodiethylammine platinos chloride ($[Pt_{en_2}][PtCl_2]$); and platodiethylammineammine (sic) chloride ($[Pt_{en_2}(NH_3)_2]Cl_2$). In a subsequent paper, taking into account Petersen's later published evidence (19) for monomolecular molecular weights by freezing-point and conductance measurements, Jørgensen halved his formulae of the first and second compounds, formulating them as Cl.Pt.en.Cl and Pt.en$_2$.Cl, respectively (20).

Jørgensen prepared the acid corresponding to Zeise's salt, K$[PtCl_6(C_2H_4)_2]$.H$_2$O, by heating sodium hexachloroplatinate(IV) with ethanol (21):

$$Na_2[PtCl_6] + 2C_2H_2OH \rightarrow H[PtCl_6(C_2H_4)_2] + 2NaCl + HCl + CH_2CHO + H_2O$$

By metathesis with KCl and NH$_4$Cl he used it to prepare Zeise's salt, an improvement over Zeise's original method (22), and the corresponding ammonium salt, respectively. In the same article he also reported the preparation of $[PtCl_2(C_2H_4)_2]$, $[Pt(NH_3)_2][PtCl_4(C_2H_4)_2]$ and the compound $[Pt(NH_3)_2][PtCl_2(NH_3)_2]$, first prepared by Alfonso Cossa (23).

Jørgensen described several methods for the preparation of platopyridinetrinamine chloride, $Pt_{py}(NH_3)_3Cl_2(H_2O)$($[Pt_{py}(NH_3)_3]Cl_2H_2O$), which on heating yielded $cis-[Pt_{py}(NH_3)_3]$, $trans-[Pt_{py}Cl_2]$ and $trans-[Pt_{py}Cl]$ (24). He also prepared $[Pt_{py}(NH_3)_3][PtCl_4]H_2O$, $[Pt_{py}(NH_3)_3][PtCl_4]$, and by various reactions he claimed that, contrary to Blomstrand's and Cleve's view, Peyrone's chloride ($cis-[PtCl_2(NH_3)_2]$) is the symmetrical compound Cl.NH$_3$.Pt.NH$_3$.Cl, while Reiset's (second) chloride ($trans-[PtCl_2(NH_3)_2]$) is the unsymmetrical compound Cl.NH$_3$.NH$_3$.Pt.Cl. He also
prepared trans-[Ptppy₃(NH₃)₂]Cl₂H₂O (15) and the platinum(IV) compounds, cis- and trans-[PtCl₂py₂] (15, 24).

Six years later Jørgensen (25) treated the ethylenediamine salt of Zeise's acid with cold water to yield yellow s-platoethylenediamine ethylene chloride:

\[
\text{C}_2\text{H}_4\text{(NH}_3\text{)}_2\text{(Cl}_2\text{PtC}_2\text{H}_4\text{Cl})_2 \rightarrow \text{C}_2\text{H}_4\text{(ClNH}_2\text{PtC}_2\text{H}_4\text{Cl})_2 + 2\text{HCl}
\]

The new compound has the structure:

\[
\begin{array}{c}
\text{Cl} \quad \text{NH}_2\text{CH}_2\text{CH}_2\text{NH}_2 \\
\text{C}_2\text{H}_4 \quad \text{Cl} \\
\end{array}
\]

Jørgensen also reiterated his contention about the structures of Peyrone’s chloride and Reiset’s second chloride based on the reaction of Peyrone’s chloride with dimethylamine to yield cis-[PtCl₂((CH₂)₂NH)₂]. He also prepared the salts [Pt(CH₂)₂NH(NH₃)₂]X₂, where X = Cl, Br or [PtCl₄].

In his last published experimental article (26) and the only paper to bear the name of a co-author, Jørgensen described a red isomer of one of the earliest, most famous, and most important co-ordination compounds to be named after its discoverer, Magnus’ green salt, tetraammineplatinum(II) tetrachloroplatinate(II), discovered in 1828 (27) by Heinrich Gustav Magnus (28). The compound was obtained by the metathetical reaction used to prepare the green salt:

\[
\text{[Pt(NH}_3\text{)}_4\text{]}\text{Cl}_2 + K_2\text{[PtCl}_6\text{]} \rightarrow \text{[Pt(NH}_3\text{)}_4\text{]}\text{[PtCl}_4\text{]} + 2\text{KCl}
\]

Conditions favourable for the production of the red isomer are the absence of K₂[PtCl₆] (traces of which are often present in K₂[PtCl₄]) and a very dilute neutral or slightly ammoniacal solution. The same isomer was also obtained by reaction of s-platoethylenediamine ethylene chloride (25) with [Pt(NH₃)₄]Cl₂ in the presence of a large excess of water. The red isomer is very stable when dry, but on boiling with water it is converted quantitatively into the green form. Jørgensen explained the isomerism, which has still not been satisfactorily explained (29), by the formulae:

\[
\begin{align*}
\text{Pt} & \quad \text{(red)} \\
\text{Pt} & \quad \text{(green)}.
\end{align*}
\]

Rhodium Compounds

Jørgensen’s first article on rhodium compounds is a 2-page preliminary communication (30) in which he thanked the Carlsberg Foundation for providing him with costly rhodium so that he could extend his studies of metal-ammines (Metallammoniakverbindungen). He announced that the rhodium complexes are completely analogous to the corresponding cobalt and chromium complexes, and he summarised the results of his experiments to be published in the future. The following year (1883), in his first detailed article, he reported the preparation of three new series of pentaammines and one new series of tetraammines (31). The pentaammines included Cl₅(Rh₂,₁₀NH₁₅)X₄ (the doubled formulas were still in use for cobalt, chromium and rhodium complexes), chloropurepurenorhodium salts ([RhCl(NH₃)₄]X₂, where X = Cl, Br, I, OH, NO₃, ClO₄, SO₄, SO₃, CO₃, SiF₄ or [PtCl₄]), Br₃(Rh₂₁₀NH₁₅)X₄ bromopurepurenorhodium salts ([RhBr(NH₃)₄]X₂, where X = Br, I, NO₃, OH, CO₃, SiF₄ or [PtBr₄]), and I₃(Rh₂₁₀NH₁₅)X₄ iodopurepurenorhodium salts ([Rhl(NH₃)₄]X₂, where X = I, Cl, Br, OH, NO₃, SO₄, SiF₄ or [PtI₄]). To the tetraammine series he assigned the formula Cl₂(Rh₂¹₈py)X₄, and the name dichlorotetrapyridinerhodium salts ([RhCl₂py]X₂, where X = Cl, Cl, HCl, Br, NO₂, OH, SO₄ or [PtCl₄]).

Because [RhCl(NH₃)₄]Cl₂ and [RhBr(NH₃)₄]Br₂ can be prepared readily in a state of high purity and because the elements, other than rhodium, that they contain have had their atomic weights accurately ascertained, Jørgensen
suggested that they could be used to determine the atomic weight of rhodium (31). By heating the chloride in air and afterwards in hydrogen and oxygen-free carbon dioxide, he obtained the atomic weight of 103.06 as the average of four determinations. Similar treatment of the bromide yielded a value of 103.03 as the average of two determinations. The currently accepted value is 102.91.

In his next article, “On the Relationship between Luteo and Roseo Salts” (32), Jørgensen cited dozens of examples to demonstrate that the luteo (hexaammine) and roseo (aquapentaammine) salts of cobalt, chromium and rhodium are analogous not only in composition (the luteo salts contain 2NH₃ in place of the 2H₂O of the roseo salts) but also in solubility, crystalline properties, appearance, formation and reactions. The structural formulae he proposed are given below, where R = cobalt, chromium, or rhodium:

### Roseo salts

\[
\begin{align*}
R_2 & : H_2O, X \\
H_N, H_N, H_N, X & \\
H_N, H_N, H_N, H_N, X & \\
H_N, H_N, & \\
H_N, X & \\
O_2 & \\
\end{align*}
\]

(modern, aquapentaammines, \([RH₂O(NH₃)₅]X₃\))

### Luteo salts

\[
\begin{align*}
R_2 & : H_N, X \\
H_N, H_N, X & \\
H_N, H_N, H_N, X & \\
H_N, H_N, H_N, H_N, X & \\
H_N, H_N, & \\
H_N, X & \\
\end{align*}
\]

(modern, hexaammines, \([RNH₆]X₃\))

In fact, in the case of the luteorhodium (hexaamminerhodium(III)) salts, Jørgensen’s proposal was a sheer prediction, for he did not succeed in preparing these compounds until the year 1891.

Jørgensen next described, in a 3-part article, the detailed preparation, properties and reactions of \([Rh₂O₁₀NH₃₂H₂O]X₃\), roseorhodium salts ((RhH₂O(NH₃)₅)X₃, where X = OH, NO₃, NO₃, HNO₃, Cl, Br, I, SO₄, (I₂SO₄), HPO₄, (NaP₂O₇), [Co(CN)₆]₃, [Fe(CN)₆]₃, (SO₄)[AuBr₄], [NO₃][PtCl₆] or (SO₄)[PtCl₆]) (33); (NO₃)₂(Rh₂₀₁₀NH₃₂X₄₃ nitratopurpureorhodium salts (nitratopentaammines, \([R(NH₃)₆]X₃\)) (34); and (NO₃)₂(Rh₁₀₁₀NH₃₂X₄₃ xanthorhodium salts (nitropentaammines, \([R(NH₃)₆]X₃\)) (35).

In 1891, Jørgensen finally succeeded in preparing the parent series of the rhodium-ammines - the long-sought luteorhodium (hexaamminerhodium(III)) salts (36), from sodium roseorhodium pyrophosphate ((RhH₂O(NH₃)₅)NaP₂O₇, 11H₂O) (31, 33) by a tedious process. A simpler process involved heating chlororpureorhodium chloride with concentrated ammonia for four days (36):

\[\text{[RhCl(NH₃)₅]Cl} + \text{NH₃} \rightarrow \text{[Rh(NH₃)₅]Cl}\]

Jørgensen (36) prepared the compounds \([Rh(NH₃)₅]X₃\), where X = NO₃, NO₂, HNO₃, Cl, Br, I, SO₄, I₂SO₄, BrSO₄, PO₄, NaP₂O₇, [PtCl₆], [Cl][PtCl₆] and \(\frac{1}{2}([SO₄]₂\ [PtCl₆])\). He also prepared \([RhH₂O(NH₃)₅]NO₃, HNO₃\) (37) and later, in his first article attacking Werner’s views (38), \([Rh(NH₃)₅][RhCl₃]\) and \([RhCl(NH₃)₅][RhCl₃]₂\). The hexaamminerhodium(III) salts, to which he assigned the monomolecular formula \((Rh₁₀₁₀NH₃₂X₄₃\) are very stable; after the passage of more than four decades the composition of Jørgensen’s samples was found to be unchanged (39).

In his last article on rhodium, published in 1903 (40), Jørgensen returned to his work of two decades earlier (31). Because of the analogous behaviour of rhodium and iridium he decided that the chloropentaamminerhodium(III) chloride used in his determination of the atomic weight of rhodium (31) might have contained iridium. Using [RhCl(NH₃)₅]Cl purified by two different methods, he repeated his atomic weight determination and obtained the previous value.

### Conclusion

Like Alfred Werner, Jørgensen was primarily interested in one of the most fundamental problems of chemistry, the nature of the chemical
bond and the constitution and configuration of chemical compounds. Co-ordination compounds, known in his time as Molekülverbindungen (molecular compounds), provided him with an ideal means to this end. Although he interpreted their structure in terms of the now defunct Blomstrand-Jørgensen chain theory, his pioneering experimental research on new series of platinum and rhodium compounds (as well as those of cobalt and chromium) has provided us with some of the most fundamental data of inorganic chemistry that are still valid today.

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