# LIFE SKETCH OF PROFESSOR SALIMUZZAMAN SIDDIQUI, F.R.S.

Born on 19th October 1897, Dr. Siddiqui had his early schooling in his home town, Lucknow, and graduated from Aligarh in 1919. Subsequently, he studied for over a year at University College, London, and then at the University of Frankfurt-on-Main, from 1921-27. After doing his First and Second Verband Examinations, he carried out his doctoral work under the guidance of Prof. Julius von Braun and was awarded the D. Phil. Nat degree on his thesis entitled, "Beitrage zur Kenntnis des Kodeins und der $\beta$-$\gamma$-ungesattigten Reste", in November 1927. On his return home he was entrusted by the late Hakim Ajmal Khan with the task of establishing the Drug Research Institute in Tibbi College, Delhi. During the ten years of his association with this Institute as its Director, he carried out intensive fundamental studies with his associates on the isolation and chemical structure of alkaloids and other physiologically active constituents of a number of medicinal plants.

In 1940, Dr. Siddiqui's services were taken over by the Council of Scientific and Industrial Research which was constituted on the outbreak of World War II. In this new position he had to devote a good deal of his attention, away from his basic researches to many applied problems relating to urgent supply demands in both civil and defence sectors. Early in 1947 in India, Dr. Siddiqui was selected as Director of the National Chemical Laboratories.

In 1951, Dr. Siddiqui was entrusted by the Government of Pakistan with the onerous assignment of establishing the Council of Scientific and Industrial Research. Starting from scratch in 1953, it has come to occupy a position which is recognised as representing the largest single research complex in the country.

On the establishment of Pakistan National Science Council in 1961, Dr. Siddiqui was appointed as its first Chairman in addition to being the Chairman of the Pakistan Council of Scientific and Industrial Research. After his retirement from these organisations in 1966, his services were enlisted by the University of Karachi as Professor of Chemistry and Research Director, with the assignment of establishing a Research Institute of Chemistry for the advanced training of students for their diploma and doctorate degrees.

As a result of persistent efforts towards the establishment of the Research Institute of Chemistry on a fullfledged basis, with an international level of working facilities, the Executive Committee of the National Economic Council gave its approval to the project, and in appreciation of the status and prospects of the Institute the Husein Ebrahim Jamal Foundation gave a generous donation of 50 lakhs towards the construction of its buildings, and the running expenditure of the Institute. On a request from the Government the Federal Republic of Germany provided a grant of 4.8 million Marks to meet the foreign exchange element of the project. The Institute has come to be regarded as one of the finest in the developing countries.

With his life long devotion to researches in the chemistry of natural products with broad based interests in related scientific disciplines, Professor Siddiqui has also been deeply involved in the cultural goods of human heritage. He owes this largely to his family tradition of interests in poetry, literature, classical music and painting. With reference to his interest in the modern trends of painting, it may be mentioned that he had his first exhibition of drawings and paintings at the Gallery Schames in Frankfurt/Main, along with paintings of Emil Nolde and Mueller, way back in 1924. And somewhat ironically, his first earnings in life were from his paintings in the second one man show at Gallery Uzielli, also at Frankfurt in 1927. His interest in this sector has continued all along with creative panses, and on the occasion of his 85th birthday, a renowned firm of publishers in Pakistan is bringing out a folder of his selected drawings. On the other hand, his translations of selected poems of Rainer Maria Rilke were published for the first time in the Urdu language during the 1930's. All this reflects in the person of Professor Siddiqui the dual graces of the arts and sciences, and his oft repeated stress on the unity of human culture.

## ACADEMIC DISTINCTIONS

In recognition of his contributions to scientific research and its organisation on a national level, Dr.

Siddiqui has been the recipient of many national and international academic honours. He was awarded the large gold medal of the Soviet Academy and D. Med. Honoris causa from the Frankfurt University in 1958. Later, he received the distinction of being elected as Fellow of the Royal Society in 1961. He was elected as member of Vatican Academy of Sciences and appointed as Pontificial Academician in 1964. In 1967 he was honoured with the conferment of D.Sc. degrees h.c. by the Universities of Karachi and Leeds.

Dr. Siddiqui is a foundation member of the Indian and Pakistan Academies of Sciences, and was elected as President of the latter for a 2-year period in 1967. He has participated in many national and international science conferences, and led Pakistan delegations to symposia and conferences abroad on numerous occasions.

## AWARDS

Member of the order of the British Empire, MBE (1946); Tamgha-e-Pakistan (1958); Sitara-e-Imtiaz, Pakistan (1962); President's Pride of performance Medal, Pakistan (1966); Hilal-e-Imtiaz, Pakistan (1980); Prize of Islamic Medicine organization award by Kuwait Foundation for the Advancement of Sciences, 1981.

## SCIENTIFIC CONTRIBUTIONS

Professor Siddiqui's contributions to pure and applied researches are embodied in over a hundred and forty research papers and memoirs and about fifty patent specifications. His life long studies in chemistry which started in 1920's have been mainly based on the isolation of physiologically active constituents of medicinal plants, elucidation of their structures and correlation of structure and pharmacological activity. These investigations have served the dual purpose of bringing up new medicinally important substances, and providing a basis for chemotherapeutic studies directed towards the synthesis of useful drugs modelled on the chemical structure of the natural products. Following is a brief sketch of some of his contributions, as it is not possible to give a full coverage to his published work in the limited space.

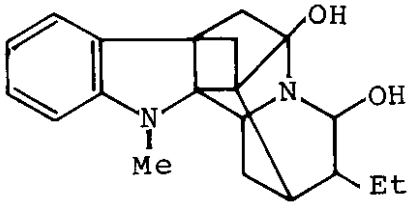
## ALKALOIDS

His studies on alkaloids go back to 1920's when he started his research career as a pupil of Prof. von Braun and worked on codeine derivatives with particular reference to the correlation of structure and activity.[1,2] This was a follow up of the observation of von Braun etal., that the degree of unsaturation in β-γ-position with respect to the basic nitrogen atom in codeine and morphine vitally affects their physiological activity. Thus the substitution of N-methyl with allyl group in these bases results in the reversal of their activity, functioning as stimulants instead of inhibitors of respiration.

Soon after his return home in 1928 he was entrusted with the task of establishing a Research Institute at the Ayurvedic and Unani Tibbi College Delhi, where he carried out intensive fundamental studies on the isolation and chemical structures of the alkaloids of various medicinal plants, among which *Rauwolfia serpentina, Holarrhena antidysenterica* and *Cassia absus* Linn. deserve special mention.

### *Rauwolfia Alkaloids*.[3-15]

As a result of his pioneering contributions to studies in the alkaloidal constituents of *Rauwolfia serpentina* during the 1930's, he reported the isolation of nine new alkaloids namely, ajmaline, ajmalinine, ajmalicine, isoajmaline, neoajmaline, serpentine, serpentinine and two weak bases m.p. 220°, and 234°. Some of these alkaloids have become world famous in the treatment of cardiovascular diseases and mental ailments. He named the main alkaloid 'ajmaline' in memory of Late Hakim Ajmal Khan who had used the drug for nearly two decades in the treatment of mental ailments. His work on the structure of ajmaline was later extended by Robinson, whose comprehensive studies in the base led to its formulation as I which was subsequently modified by Woodward as II.

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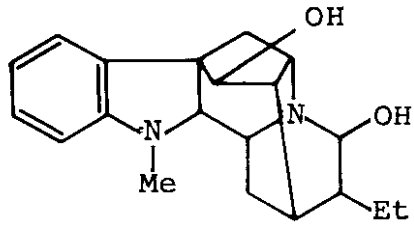


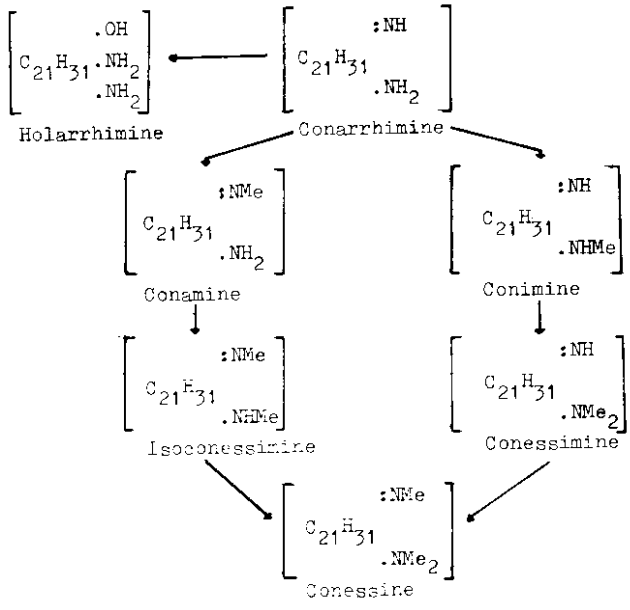

II

More recently, in view of increasing importance of ajmaline as a drug of choice in the treatment of cardiac arrhythmias, Prof. Siddiqui carried out studies in the correlation of structure and activity in the ajmaline series of bases which led to the hypothesis, that any chemical intrusion that stabilizes either the carbinol-amine or the aldehyde-imine structure of ajmaline, greatly reduces its cardiac activity. Thus diacetyl ajmaline in which the carbinol-amine ring is stabilized as well as N-methyl ajmaline in which the aldehyde group is fixed, show very meagre anti arrhythmic activity, indicating that the carbinol-amine aldehyde-imine tautomeric structure =N–CH–(OH) ⇌ NH–CHO serves as what may be termed as a cardiophoric grouping.

As a result of an observation in the course of these studies that mono-nitroajmaline is about twice as active as the mother base, he extended the nitration studies to reserpine.[131] Following highly critical experimental conditions, he was able to obtain 1-, 12-and 9-mononitro derivatives, the first two in fairly good yields. Pharmacological studies have shown that 1-nitroreserpine has the same order of activity as reserpine and is free from its undesirable side effects. On the other hand, 12-nitroreserpine has lower hypotensive action, but is much longer lasting when administered to unanaesthetised dogs.

*Holarrhena Alkaloids*:[16-33]

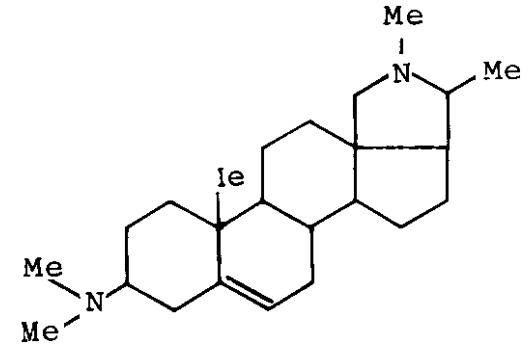
During the same period (1930's) he carried out systematic studies in the alkaloids of *Holarrhena antidysenterica* on account of its long reputed importance in the treatment of amoebic dysentery and other intestinal ailments. As a result of these investigations he communicated the isolation of six new crystalline bases, in addition to the alkaloid conessine which had been isolated as far back as 1858 by Haines. As a result of structural studies their mutual relationship with respect to conessine was established on the following pattern.




According to this, conarrhimine was considered as the mother base which leads on the one hand, to conessine and its various nor bases through methylation, and on the other, to holarrhimine through the hydrolytic fission of its nitrogen ring. These conclusions were mainly based on the studies in the action of BrCN on conessine, which led to the mono- and di-nor bases identical with isoconessimine and conimine both being convertible to conessine on methylation with formaldehyde/formic acid. It may be noted in this context that the methylation of the petroleum ether soluble fraction of the total alkaloids raised the yield of conessine to 0.4% as against a maximum of 0.1% reported by earlier workers. This finding greatly helped in subsequent structural studies.

Following the observations of Simonsen and Spaeth relating to Hofmann and Emde degradation of conessine, Prof. Siddiqui extended these studies to isoconessine. He noted that while conessine gives apoconessine on Hofmann degradation which on further degradation after Emde gives a hydrocarbon, later identified by Haworth as pregna-3,5, 20-triene, isoconessine is quantitatively recovered on its Hofmann degradation as a ditertiary base with the elimination of two molecules of methyl alcohol. Correlating this contrast between the behaviour of conessine and isoconessine with the observation of von Braun et.al. on the negative influence of β-γ- unsaturation on the N-stability of radicals attached to the nitrogen atom, he considered it probable that the isomerization of conessine was due to a shifting of the double bond.

He further observed that on heating conessine and isoconessine hydroiodides at 310° and 325° respectively, an identical hydrocarbon $C_{21}H_{30}$ with three double bonds, is obtained which he named as conessen. Comprehensive studies in conessen subsequently carried out by Haworth led to the establishment of the structure of conessine as III. On the other hand, the structures of iso- and neo- conessine, obtained by Prof. Siddiqui on the treatment of conessine with sulphuric acid, were established by Janot as IV and V.

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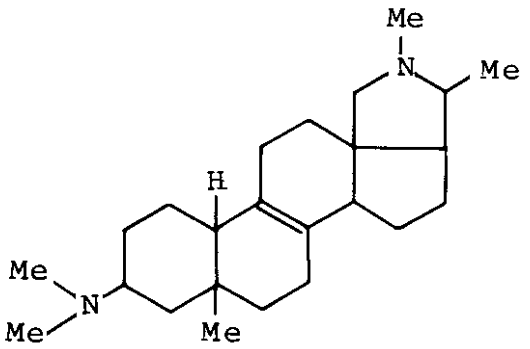


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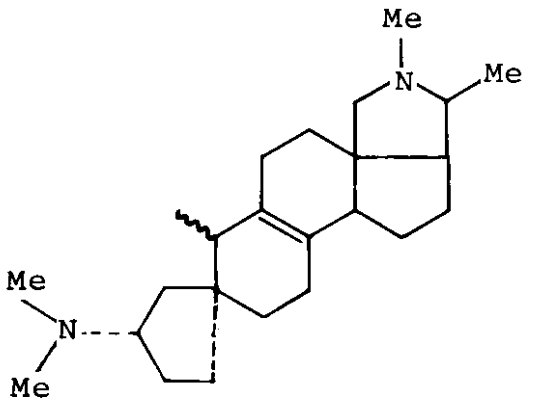


V

Some Extensions of von Braun BrCN Reaction:-

Reverting to his early work on the reaction of cyanogen bromide with conessine and isoconessine, Professor Siddiqui more recently contrived through highly critical experimental conditions to convert the monocyanamides of the two bases to their amides on mild acidic hydrolysis, diamines on reduction with zinc and hydrochloric acid, and carbinol amines from the latter on reaction with nitrous acid. On the other hand, they also furnished the guanido derivatives on treatment with concentrated ammonia.[122]

Later on, with the object of ascertaining the applicability and limitations of these reactions, he extended the studies to a number of bases including *Harmala*,[125] *Rauwolfia*[132] and *Ephedra*[133] alkaloids, as well as simpler aliphatic and aromatic amines, and arrived at the following conclusions:

1. The extension of von Braun BrCN reaction, which was employed for the conversion of tertiary into secondary amines, has a wide range of applicability and provides a new route for obtaining pharmacophoric derivatives of various organic bases in good yields.
2. The cyanamides in which the nitrogen atom occurs in a ring system are generally irresponsive to these reactions.
3. The urea derivatives could be prepared from all types of simpler bases, but in the case of alkaloids, the formation of such derivatives is sterically conditioned. Thus the alkaloids sandwicine, isosandwicine, ajmaline and isoajmaline which have tautomeric carbinolamine and aldehyde-imine structure do not yield urea derivative on mild hydrolysis. Further, attempts to obtain the amido derivative from the cyanamide of ephedrine led instead to a product identified as 2-imino-3,4-dimethyl-5-phenyl-oxazolidine. Work in this direction is being further pursued by Prof. Siddiqui's group.

*Cassia Alkaloids*[34,35,37,42,43,47,49]

Another significant contribution of Prof. Siddiqui in the field of alkaloids is the isolation of chaksine and isochaksine - the quaternary bases of *Cassia absus Linn.* As a result of structural studies in chaksine he noted that the thermal degradation of the base at 310-20° leads to p-iso-propylbenzoic acid and phthalic acid,

indicating the presence of a terpenic carbon skeleton, and accounting for ten out of the eleven carbon atoms of the molecule. Subsequent studies carried out by his and other groups of workers led to the formulation of tentative structures containing lactonic and guanidinic functions. With reference to these and other earlier findings, Wiesner proposed a lactonic structure for chaksine, further work on which through x-ray crystallography is awaited. Particular significance is attached to the isolation of chaksine as the first alkaloid carrying a monoterpenic nucleus.

*Harmala Alkaloids* :[46,47,52,126,130,135,140,]

In continuation of studies in the correlation of structure and activity relationships, he and his associates undertook extensive pharmaco-chemical studies on the main alkaloidal bases of *Peganum harmala* namely harmine and harmidine/harmaline and reported a whole series of new potentially pharmacophoric derivatives of harmine, harmidine, harmol, harmidol, tetrahydroharmine and amino tetrahydroharmine. Further studies in this direction are being persued.

## NON ALKALOIDAL CONSTITUENTS

*Semecarpus anacardium Linn.* [56-66,103,104]

Owing to the medicinal importance attributed to *Semecarpus anacardium* (Bhilawan), Prof. Siddiqui and his associates undertook in 1931 a systematic investigation of the resinol occurring in the pericarp of its nuts, and communicated the isolation of a phenolic substance Bhilawanol through fractional distillation. Studies in the structure of Bhilawanol revealed it as a catechol derivative with a $C_{15}H_{27}$ unsaturated straight side chain, carrying an average of two double bonds. It was later established that it is closely related to uroshiol obtained by Majima from the Japanese lacquer varnish ki-urushi (Rhus vernicifera) and differs from it only in the number and position of the double bonds in the side chain. These findings subsequently led to the industrial development of the resinol for the production of stoving enamels, varnishes, and moulding compositions during the World War II. In a recent reinvestigation of the petroleum ether insoluble, alcohol soluble component of Bhilawan nuts, two dimeric flavonoids have been isolated, the exact structure of which is under investigation.

*Melia azadirachta Linn.*[69-79,120]

In view of the curative properties attributed in folklore and traditional medicine to *Melia azadirachta* (Nim), it has been subjected to chemical and therapeutic studies from about the beginning of the current century. These studies were mainly concerned with the fatty acid components and other mostly amorphous substances isolated from the oil. However, it was only as late as 1942 when Prof. Siddiqui reported the isolation of two crystalline triterpenoids, nimbin and nimbinin, from the oil through solvent partitioning technique, without involving any chemical intervention. Their structures were subsequently established through structural work carried out by him and other groups of workers. These investigations stimulated a great deal of interest among many groups of workers in the constituents of various parts of the Nim tree, but it was first in 1972 when Prof. Siddiqui undertook a systematic study of the fresh undried fruit pulp, as a result of which he and his associates reported the isolation and structure of three new crystalline triterpenoids namely, nimolicine, nimoline and 17 $\beta$-hydroxyazadiradione. Studies in the various parts of "nim" tree are being further pursued in view of the exceptional interest in its constituents throughout the world.

*Cicer arietinum Linn*[94-97, 100,101,]

Taking into consideration the importance of *Cicer arietinum* (Vern. chana; Bengal gram) in the diet of the subcontinent Prof. Siddiqui worked on the fresh, undried germination sprouts and communicated in 1945 the isolation of three crystalline constituents which he named as Biochanin-A, Biochanin-B Biochanin-C. As a result of comprehensive structural studies, Biochanin-A was formulated as $C_{16}H_{14}O_5$ as a new isoflavone which was subsequently confirmed through synthesis by Baker. On the other hand Biochanin-B was identified with formononetine and Biochanin-C with the amino acid asparagine. Considerable significance has been recently attached to Biochanin-A on account of its oestrogenic activity and a potential antifertility agent along with genestein.

In conclusion, a brief reference may be made to Prof. Siddiqui's mode of work in the isolation of plant constituents and the experimental management of chemical reactions. As will be seen from some of the

examples noted above, he lays great stress on the use of fresh, undried plant materials for studies in their constituents, in order to avoid structural changes likely to occur in them through aerial oxidation and enzymatic action in the process of drying and storage. Furthermore, he continues to employ classical methods of isolation in view of the fact that once a method for obtaining uniform constituents has been ultimately worked out, they can be obtained in bulk quantities. This affords the possibility of comprehensive pharmacological investigations, and studies in the structure and activity relationship based on their derivatives.

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