

## Milan Randić – Life and Work

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Professor Milan Randić is an important person in the history of Croatian chemistry.<sup>1</sup> It was he that first did research in quantum chemistry in Croatia and, thus, started theoretical chemistry research in our country. He founded the Theoretical Chemistry Group at the Rugjer Bošković Institute in Zagreb. Randić was so persuasive a proselytizer that he attracted to the Theoretical Chemistry Group a number of bright and hard working graduate students who, along with their own students, are currently the most productive scientists in Croatia. Randić is also widely recognized as one of the world leaders in the contemporary mathematical chemistry.

Randić recently summarized his research philosophy in the following words:<sup>2</sup> »There are no doubts that experiments are central to chemistry, as has been recently emphasized by Roald Hoffmann (*Amer. Scientist* **81** (1993) 15–17). One may continue then and argue that while central to chemistry are experiments, central to an experiment is theory, modeling. However, I hasten to add that while most experiments yield data that may eventually become of interest to theory, not every theory and every model necessarily stimulate experiments, or are of interest to an experimental chemist. This is true even when theory is sound and the model of a molecule is generally accepted. On the other hand, experiments without theory appear, as Lord Rutherford once reflected, like stamp collecting!«.

### CURRICULUM VITAE

Milan Randić was born in Belgrade (Serbia) on October 1st, 1930. His parents were Olga (née Medanić) (1900–1990) and Dinko Randić (1894–1971). His father Dinko Randić was a customs officer in Belgrade and thus Milan was born there. Randić and Medanić are very old Croatian families of

seafaring people. They have lived for centuries in Kostrena – Sveta Barbara, a small place on the northern Adriatic coast, close to Sušak and Rijeka.

Dinko Randić was a colorful character. He was a soldier in the Austro-Hungarian army during the First World War and was taken prisoner by the Russians. In due course, he participated with many other Croatian prisoners of war in the October Revolution of 1917, which brought Lenin and the Soviets to power in Russia. Years later this would bring him a decoration by the Soviet Government.

Since during his customs service, his father was transferred to several places in the period between 1938 and 1942, Milan completed his primary education in three cities: Belgrade (first grade), Kostrena (second grade) and Split (third and fourth grades). He completed his secondary schooling in the difficult war and post-war period from 1942 to 1949. He entered the University of Zagreb in 1949, first enrolling in the Department of Chemical Technology at the Technical Faculty. Randić soon found that this was a mistake and in the second semester he joined the Department of Physics at the Faculty of Science and Mathematics, being attracted to physics by weekly seminars organized by Professor Ivan Supek, a former student of the Nobel Laureate in Physics Werner Heisenberg (1901–1976). Milan Randić graduated in theoretical physics in 1954 after completing research for his B.Sc. thesis under Professor Supek. This work dealt with the electronic structure of benzene and represents the first piece of research in quantum chemistry ever done in Croatia. Professor Supek presumably gave this problem to Randić because of his one semester of chemistry studies.

Randić received his Ph.D. degree from Cambridge University in 1958 for his thesis entitled *Some Studies of Infrared Spectra of Molecules*, in which he reported his research on the analysis of infrared spectra of high resolution. This research was carried out under the guidance of Professor Norman Sheppard FRS. During his Cambridge days he also attended lectures given by a number of theoretical physicists and chemists, such as the Nobel Laureate in Physics Paul A. M. Dirac (1902–1984) and the leading English theoretical chemist of those days Charles A. Coulson (1910–1974). Randić also struck a life-long friendship with John N. Murrell, the future world-famous theoretical chemist and a Fellow of the Royal Society. Professor Murrell was later teacher to several Croatian theoretical chemists (e.g., Trinajstić, Bosanac, Živković) and thus has, besides Randić, most strongly influenced the early development of theoretical chemistry in Croatia.

Randić liked so much what he heard about theoretical chemistry that, after returning from Cambridge, he abandoned molecular spectroscopy for good and started doing research in theoretical chemistry. Soon after his return to Zagreb, Randić had to serve a year in the army. Upon his return to

Zagreb from the army service, he founded the Theoretical Chemistry Group at the Rugjer Bošković Institute. Randić first attempted to form a molecular physics group in the Department of Theoretical Physics, but without success. However, Professor Božo Težak (1907–1980), head of the Department of Physical Chemistry, was much more open-minded than the Institute's theoretical physicists and asked Randić to join his department and to form a theoretical chemistry group.



Milan Randić in the time of his most important paper on the connectivity index (1975) and his son Velimir.

Randić remained at the Rugjer Bošković Institute until 1965 as a research associate. But, he also spent two years abroad in this period: the first year with John N. Murrell at the University of Sheffield and the second year with David Bishop at the University of Ottawa. In 1965 Randić joined the Department of Chemistry at the Faculty of Science and Mathematics, as an associate professor soon to be promoted to full professorship. He was the

first professor of quantum chemistry in Croatia and was giving undergraduate and graduate lectures in quantum chemistry and molecular spectroscopy.

His undergraduate lectures were based on the books by Walter, Eyring and Kimball (H. Eyring, J. Walter, and G. E. Kimball, *Quantum Chemistry*, Wiley, New York, 1944) and by Coulson (C. A. Coulson, *Valence*, University Press, Oxford, 1952) while he also recommended the following two books for his graduate lectures: A. Stretwieser, Jr., *Molecular Orbital Theory for Organic Chemists* (Wiley, New York, 1961) and F. A. Cotton, *Chemical Applications of Group Theory* (Interscience-Wiley, New York, 1963). Randić did not write a book on quantum chemistry, but wrote an article entitled *Atom in Technical Encyclopaedia* (*Tehnička enciklopedija*, Leksikografski Zavod, Zagreb, 1963, Part 1, pp. 456–479) in which he introduced some quantum-chemical concepts, such as hybridization, crystal field theory, elements of the VB and MO theory, many-body wave functions, etc. He also wrote a chapter entitled *Molecular Quantum Mechanics* in Supek's book *Theoretical Physics and the Structure of Matter* (*Teorijska fizika i struktura materije*, Školska knjiga, Zagreb, 1962–1963; second part, third edition) in which he presented some elementary concepts of quantum chemistry. These were the first introductory texts on quantum-chemical topics in Croatia.

In 1971 Randić left Zagreb for good, but his influence on Croatian theoretical chemistry is felt even today, since his first graduate students are still very active in research. Leaving Zagreb also meant his abandonment of quantum chemistry research. In his American years he turned to mathematical chemistry. This was a very wise decision because while he was doing quantum chemistry, although very innovatively, he was just another quantum chemist, but his very creative and prolific research in mathematical chemistry launched him to the position of one of the world leaders in this field of theoretical chemistry.

Randić spent the next ten years (1971–1980) at various universities in the USA, such as Johns Hopkins University in Baltimore (Maryland), University of Utah in Salt Lake City, Massachusetts Institute of Technology in Boston, Harvard University in Cambridge (Massachusetts), Tufts University in Medford (Massachusetts), State University of New York in Buffalo, Iowa State University in Ames, and University of Nebraska in Omaha. He finally settled down in 1980 at Drake University in Des Moines (Iowa). Randić became associate professor in the Department of Mathematics and Computer Science. In 1985 he was elected to full professorship and in 1986 his excellent research record was recognized by the Drake University authorities and Randić became the Centennial Scholar there.

Randić took his first sabbatical leave from Drake University in 1986 and spent a very productive year at Cornell University in Ithaca (New York),

University of Pennsylvania (College Park) and University of Georgia in Athens. In 1987 he was promoted to the Ellis and Nelle Levitt distinguished professorship. There are only three such professorships at Drake University. He almost died in the autumn of 1992. Randić spent the summer of 1992 at the Rugjer Bošković Institute in Zagreb and the National Institute of Chemistry in Ljubljana (Slovenia). After returning to Ames (his home town is Ames while his workplace is 60 miles away in Des Moines), he had a severe heart problem and was operated on open heart in October. Randić recovered extremely quickly and was back at Drake University in December.

Randić took the second sabbatical leave from Drake University in 1993. This sabbatical year was again very productive and was spent at the Ochanomizu University in Tokyo (Japan), Texas A&M University in Galveston and National Institute of Chemistry in Ljubljana. At the end of 1999, Randić retired from his teaching position at Drake University. However, he continues to do highly creative and very productive research commuting between the Natural Resources Research Institute in Duluth (Minnesota) and the National Institute of Chemistry in Ljubljana, with visits to Drake University and Iowa State University.

Randić left Zagreb thirty years ago but continued to visit the Rugjer Bošković Institute on a yearly basis and to collaborate with the members of the Theoretical Chemistry Group. Randić's efforts in organizing research in theoretical chemistry in Croatia and his continuous collaboration with Croatian theoretical chemists was recognized by the Croatian Academy of Sciences and Arts and in 1997 he was elected a corresponding member of the Academy.

During his journeys, Randić has met and collaborated with many distinguished quantum chemists, computational chemists and mathematical chemists such as David M. Bishop, Robert G. Parr, Haruo Hosoya, Roald Hoffmann (who shared the Nobel Prize in Chemistry with Kenichi Fukui in 1981), Marko Razinger, E. Bright Wilson, Frank E. Harris, Per-Olov Löwdin, Lemont B. Kier, Lowell H. Hall, R. Bruce King, Douglas J. Klein, Dennis H. Rouvray, Peter C. Jurs, Subhash C. Basak, Jure Zupan, Alexandru T. Balaban, *etc.*

## RESEARCH WORK

As already mentioned, Randić's research may be split into three main areas. In Cambridge (England) he did research in molecular spectroscopy, in Zagreb in quantum chemistry, and in the USA in mathematical chemistry.

### *Molecular Spectroscopy Research*

Randić published only nine papers in the field of molecular spectroscopy. His first published paper (with Ian M. Mills, who became a distinguished spectroscopist and a Fellow of the Royal Society) was in molecular spectroscopy.<sup>3</sup> His papers in molecular spectroscopy are interesting but not exceptional works. Randić was not born to become a spectroscopist.

### *The Zagreb Period (1960–1971)*

In his Zagreb period Randić published 43 papers, mostly in the field of quantum chemistry. His most interesting papers in this period deal with the model of maximum overlap hybridization for strained systems. In the first two papers of the series, the model was introduced and applied to cyclopropane and related systems.<sup>4,5</sup> This approach was strongly influenced by an earlier work of Coulson and his doctoral student William E. Moffitt (C. A. Coulson and W. E. Moffitt, I. Properties of certain Strained Hydrocarbons, *Phil. Mag.* **40** (ser. 7) (1949) 1–35). Although some of the papers in this period are innovative, such as the study on the difference between the bond orders calculated by the SCF MO method,<sup>6</sup> early *ab initio* computations,<sup>7</sup> or prediction of bond lengths in cyclopentadienyl ligands,<sup>8</sup> Randić would be hardly remembered for his work in quantum chemistry on a global scale. These were the results of just another quantum chemist. Better things were awaiting Randić in the future. However, these works are important for Croatian chemistry because they established research in theoretical chemistry in Croatia and Randić was able to attract to theoretical chemistry a number of young and gifted physicists and chemists. Since these early days, theoretical chemistry in Croatia has become one of the internationally best-recognized research activities in our country. Randić has entered the history of Croatian chemistry as the person who started quantum chemistry research in Croatia.

### *The USA Period (1971–present)*

The USA period is the most important part of Randić's career. In this period he has published more than 350 papers so far and many more are expected to appear. He was the only author of most of these papers. In the USA he started doing research in mathematical chemistry and has produced several seminal papers that are widely cited. The central theme of his research in mathematical chemistry is the nature of the chemical structure.<sup>9</sup> Probably his most important paper is the one in which he introduced the concept of the connectivity index.<sup>10</sup> This paper has been cited more than 1600 times to date. It is used more than any other topological index in the

QSPR (quantitative structure-property relationship) and QSAR (quantitative structure-activity relationship) modeling (e.g., L. B. Kier and L. H. Hall, *Molecular Connectivity in Structure-Activity Analysis*, Wiley, Chichester, 1986). Recognizing the importance of this paper, the Division of Computers in Chemistry of the American Chemical Society (ACS) organized in Randić's honor a two-day symposium *Molecular Connectivity – The First Quarter Century* (Washington, DC, August 22–23, 2000) during the Semiannual Meeting of the ACS (Washington, DC, August 20–24, 2000). The connectivity index has also attracted attention of mathematicians. For example, Béla Bollobás wrote a paper with Paul Erdős (1913–1996) (*Graphs of Extremal Weights*, *Ars Combinatoria* **50** (1998) 225–233) in which they called the connectivity index the Randić weight of a connected graph.

Another important paper, which is also widely cited, is the paper in which Randić developed the conjugated-circuit model<sup>11</sup> for studying and classifying aromatic systems. This paper has been cited more than 1200 times so far. The conjugated-circuit model has been shown to be especially useful for studying the fullerene systems. He has also introduced many new concepts, important in QSPR and QSAR modeling, such as the concepts of orthogonal descriptors,<sup>12</sup> molecular profiles,<sup>2</sup> topographic descriptors,<sup>13</sup> variable descriptors,<sup>14</sup> etc. Judging by his contributions, Randić appears to be one of the most influential researchers in mathematical chemistry in the last three decades. This is well-illustrated by what follows. Because of his seminal contributions to the theory of molecular descriptors, Randić was invited to contribute an article on this topic to *The Encyclopedia of Computational Chemistry*.<sup>15</sup> In the *Handbook of Molecular Descriptors* (Wiley – VCH, New York, 2000) prepared by Roberto Todeschini and Viviana Consonni, Randić is the most cited author.

Randić has published about 400 research papers so far and is still a very prolific researcher. He is the only author of more than 150 papers while in more than 200 papers he has co-authors. One of us (NT) has been his most frequent co-author, in 75 papers so far. The other author of this essay (SN) has published 18 papers with Randić. Randić's most productive year was 1988 when he published 21 papers. He has also published about 10 technical papers and five literary works under the pen name Antonin Medanić.

## MISCELLANEOUS

Milan Randić has served and is still serving on editorial boards of several journals. He was or is a member of the editorial board or the advisory board of the following journals: *Croatica Chemica Acta* (1963–present), *Chemical Physics Letters* (1965–1975), *Journal of Mathematical Chemistry* (1987–



present), *Journal of Chemical Information and Computer Sciences* (1991–2001), and *SAR & QSAR in Environmental Research* (1992–present). Since 1992 Randić has also served with Douglas J. Klein as the co-editor of Mathematical Chemistry Series, published by CRC Press, Boca Raton, Florida.

His excellence in research has been recognized by several awards. Randić was awarded The City of Zagreb Award in 1967, in 1987 The Boris Kidrič Award (Slovenian national science award; Randić shared this award with Irena Fabič and Borka Jerman-Blažič who got her Ph.D. under NT), in 1989 the Governor's Medal for Research (the science achievement award of the state of Iowa) and in 1996 the Herman Skolnik Award given by the American Chemical Society.

Randić is a member of the Croatian Chemical Society, American Chemical Society, Mathematical Association of America, International Society of Mathematical Chemistry (president), Society for Industrial and Applied Mathematics, International Society of Mathematical Modeling, World Association of Theoretical Organic Chemists, and International Society for Quantum Biology.

### CONCLUDING REMARKS

Milan Randić's remarkable research achievements in mathematical chemistry can be best summarized by quoting the words accompanying the Governor's Science medal: »Randić, a world-class research scientist in the application of mathematical graph theory to chemical problems, was cited for outstanding accomplishments in science...« Randić also serves as a bridge between the American and Croatian scientific communities and can be looked at as an eloquent Croatian science ambassador to the USA.<sup>16</sup>

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