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TRIBUTE

The Dielectric Barrier Discharge and the Start of a Beautiful Friendship: Personal Remembrance of Dr. Ulrich Kogelschatz

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Abstract

One can safely say that the pioneering work of Dr. Ulrich Kogelschatz and his team on the physics and chemistry of the dielectric barrier discharge was foundational and remain of great relevance to the present day. Kogelschatz and his team made some of the early high-resolution measurements of the current pulses associated with filaments and conducted simulations to elucidate the chemical processes taking place in DBD plasmas. On the engineering/technology side, he contributed to the development of DBD-based excimer lamps, to the optimization of DBD-based ozone generators and to many other advancements. His numerous papers on the physics and applications of DBD have educated a generation of researchers involved with such devices. I consider myself one of those students who learned a lot from him, both from his writings and from personal interaction. In this perspective, I first briefly summarize the working of barrier discharges and some of Kogelschatz's contributions to this field. Then, I highlight some of the most valuable interactions I had the privilege to have with him, who I considered to be a mentor and a dear friend for many years. Please note that I use the formal title "Dr. Kogelschatz" in the title and subtitles but in the text, I use Ulrich (or Uli) as that is how I addressed him in person.

Keywords Dielectric barrier discharge · Low temperature plasma · Ozone · Excimer lamp

Brief Overview of Barrier Discharges and on Dr. Kogelschatz's Contributions

The first report on barrier discharges was made by Theodose du Moncel in 1855 [1, 2]. Du Moncel used planar metal electrodes covered by glass plates and separated by a small gas gap. As a power source he used a Ruhmkorff coil, an induction coil that allowed for the generation of high AC voltages from a low voltage DC source. In 1857 Werner von Siemens reported on the design of a DBD apparatus which he used to generate ozone [3]. The DBD apparatus used by Siemens had a cylindrical geometry with tin foils as electrodes and glass as dielectric.

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The mechanisms and modes of operation of the dielectric barrier discharge have been studied in depth by Kogelschatz and other investigators resulting in a relatively well understood device [4–10]. DBDs use a dielectric material to cover at least one of two electrodes. Typical operation of the DBD involves the use of high voltages at frequencies in the kHz-MHz range, applied between dielectric covered electrodes which are separated by gaps ranging from the millimeter to several centimeters. As an illustration, Fig. 1 shows a photograph of a DBD used by my research group for surface modification. Investigators were able to extend the frequency of operation all the way down to the line frequency (50/60 Hz) and even to DC by replacing the dielectric by a high resistivity layer [11]. Such device is known as the resistive barrier discharge (RBD) [11]. In this device the high resistivity film plays the role of a distributed ballast that limits the discharge current and prevents it from transitioning into a spark. Using a dielectric wire mesh as an electrode, other investigators were able to generate a stable discharge in various gases (air, argon, nitrogen, etc.) at the line frequency of 50 Hz [8]. In addition, in the early 2000s, investigators discovered that by using nanosecond high voltage pulses they could control better the electron energy distribution function (EEDF) and thus enhancing the rate of ionization and excitation in the plasma generated by the DBD [12–14]. This provided a means to control the plasma chemistry, which is beneficial for various applications.

The DBD is one of the most used plasma sources to generate low temperature atmospheric pressure plasmas. Its industrial applications are numerous and many of these benefited greatly from Ulrich's contributions. While a scientist at the Brown Boveri Corporate Research, Ulrich was involved in broad experimental and theoretical studies on ozone synthesis from oxygen in dielectric barrier discharges. He and his team investigated the details of electron kinetics in atmospheric pressure oxygen and their results allowed the field of ozone generation to move from trial and error to science and modeling-based endeavor. Ulrich and his team showed that the filamentary nature of the discharge worked to the benefit of ozone generation and that ozone is generated and destroyed inside the filaments of the discharge. By judiciously tweaking the operating parameter of the discharge they were able to double the ozone concentration while

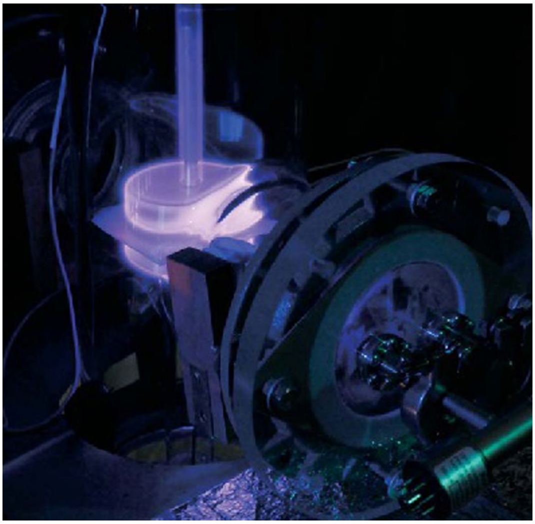

Fig. 1 Dielectric Barrier Discharge for surface treatment

reducing the specific energy by half. Using their new design, Brown Boveri was able to construct large scale ozone generators which were considered a major milestone for that industry.

Based their work on electron kinetics in DBD, Ulrich and his team proposed new designs to make powerful excimer lamps. Excimer lamps use DBD-based discharges in rare gases and rare gas/halogen mixtures to generate narrow band UV and vacuum UV radiation that is used in various industrial applications including in scanners and copiers.

One last application that I would like to mention here and where Ulrich and his team made early contributions is using plasma for the reduction of greenhouse gases and/or the conversion of these to more useful chemicals like liquid methanol. This line of research is still pursued today by several research groups and its relevance to the climate change crisis cannot be overstated.

Finally. and to conclude, although he did not contribute to the biomedical applications of the DBD, Ulrich was aware of this new development from its beginning in 1990s. He followed closely the progress of the research and has on many occasions expressed to me personally that he expected that this emerging application would open a new and exciting area of research for low temperature plasma. In fact, he mentions this in the first paragraph of page 26 of the paper that is the subject of this special issue (see Ref. [6]).

For more information on Ulrich Kogelschatz and his scientific achievements the reader can consult reference [15].

My Scientific Interactions with Dr. Ulrich Kogelschatz

I first met Ulrich Kogelschatz in person in 1998. He came to Virginia to visit old colleagues from NASA Langley (Hampton, Virginia) where he once spent nearly two years as a Post-doctoral Associate. He heard that I was giving a lecture at Old Dominion University on my recent work on the biological applications of the DBD and asked the organizers if he could attend. At the conference room I was surprised to see him in the audience. I, of course, knew of him from his publications and was aware of his scientific reputation. His presence made me a little nervous, but I was also deeply honored to see him there. After my talk I had a little chat with him. He was very kind and told me that he read my 1996 TPS paper on the topic (see ref [16]) and was interested to see if I made more progress. He then expressed to me that this novel application of the DBD (biological application) sounded very exciting and asked me to keep him informed about the progress of this research. I was extremely encouraged by his interest, approval, and support.

The next time I saw Ulrich was during a second visit to Old Dominion University in the early 2000s. There was research work going on at the ODU Applied Research Center on excimer UV generation and he was invited to give a lecture on excimer lamps. After his lecture he came to my lab and asked if I could show him the Resistive Barrier Discharge (RBD) in operation. I have developed the RBD with Prof. Igor Alexeff a few years earlier, and at the time of Ulrich's visit I had a student using it for a decontamination experiment. Ulrich inspected our discharge very carefully and gave us some helpful suggestions. He noticed that we placed an instrument on top of thick manual to adjust for height. He smiled and said, "In academia labs you can do that, but not in an industrial R&D lab, where experimental procedures have to be tightly followed".

In January 2003, Prof. Kurt Becker, who was the chair of the Engineering Physics Department at Stevens Institute of Technology, organized a workshop in Hoboken, New

Jersey. The campus of Stevens Institute of Technology is located right across the Hudson River, facing Manhattan. That winter was very cold (temperatures were hovering around − 10 °C), and the surface of the Hudson was covered by a sheet of ice. So, when Kurt Becker asked us to line up outside for a group picture, we all hurriedly stood up with Manhattan in the background and asked the photographer to be quick, then we rushed back inside, our noses red, ears about to turn blue, and our fingers numb from the cold (see photograph in Fig. 2). This workshop was held to organize an effort to write a book on non-equilibrium air plasmas at atmospheric pressure. All preselected chapter authors were invited to attend and "brainstorm" on what should be included in this book and to come up with a workable timetable and a table of contents. Uli, who was a co-editor of the book, was also the lead author of chapter 2 and chapter 6. Chapter 2 was about the history of non-equilibrium air discharges. Chapter 6 included sections on barrier discharges, microdischarges, corona discharges etc. I was invited by Uli to write the section on homogeneous barrier discharges, which I did. I was also in charge of chapter 9 which covered the applications of atmospheric pressure plasmas. Uli wrote the section on electrostatic precipitation and the section on ozone generation. Eva Stoffels and I wrote sections on the then emerging biomedical applications of atmospheric pressure plasmas. Chapter 9 also covered various other applications ranging from material processing to plasma-aided combustion. The book was published in 2005 by the Institute of Physics Publishing under the title of "non-equilibrium air plasmas at atmospheric pressure" [17] (see Fig. 3).

Fig. 2 Group picture of the book authors with Manhattan in the background (not all the authors were present). Ulrich Kogelschatz is standing third from the right. Kurt Becker (workshop organizer and book lead editor) is 7th from the right and me in the middle (with blue Jacket), 10th from the right. To my immediate left (between me and Kurt Becker) was Osamu Ishihara who oversaw chapter 5 on modeling. Both of us were pupils of Prof. Igor Alexeff at the University of Tennessee. Osamu became the president of Chubu University, Kasugai, Aichi, Japan from 2017 to 2021. Second from right stood Robert (Bob) Barker whose AFOSR directorate sponsored the book workshop. Bob's directorate was the first funding agency unit that supported and provided the early funding (around 1997) for the biomedical applications of cold atmospheric pressure plasma (CAP). The goals of that AFOSR program were to develop CAP devices/sources and use them for biological and chemical decontamination and for wound healing. To the left of Bob stood Erich Kunhardt, one the of leading gas discharge physicists of 1980s and 1990s. Remaining co-authors shown (and not shown) in the picture, were all reputed leading scientists that made equally valuable contributions to the book

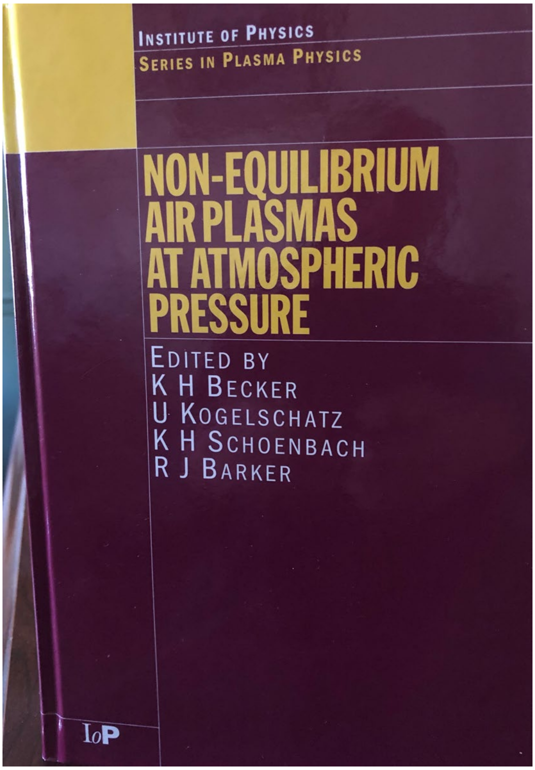

Fig. 3 Book edited by Becker, Kogelschatz, et al. and published by IoP in 2005. Some of the most prominent scientists in discharge physics and chemistry contributed to chapters of this book. In addition to U. Kogelschatz, these included W. Rich, Y. S. Akishev, E. E. Kunhardt, J. P. Boeuf, L. C. Pitchford, R. N. Zare (pioneer of the LIF diagnostic technique), S. Okazaki, and others. A. Garscadden wrote the Foreword. The book also featured an extensive applications chapter which, for the first time in a book, included a coverage of the then emerging field of the biomedical applications of cold plasma. I had the privilege to lead the applications chapter and to co-author with Eva Stoffels the sections on the biomedical applications

Conclusion

It is without a doubt that Ulrich Kogelschatz was a very influential scientific leader in the field of low temperature plasma and specifically the physics and chemistry of the dielectric barrier discharge and its many applications. His papers, including the subject of this special issue (reference [6]), are still widely read and continuously well cited. He was always enthusiastic and shared his knowledge generously. I experienced this first-hand when he visited the ODU Applied Research Center and gave us valuable feedback on our VUV source experiments, DBD and RBD research, and others. It had been my great honor to be his friend and I feel fortunate to be given the opportunity to write my perspective and share some anecdotes related to my friendship with Uli. To conclude, I would like to thank Prof. Annemie Bogaerts, guest editor of this special issue, for inviting me to write this paper.

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