

## ELECTRON CHARGE CLUSTER SPARKING IN AQUEOUS SOLUTIONS

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### ABSTRACT

**Valve metals** (metals whose oxides provide electron emission) can produce luminescence in an aqueous solution. Luminescence increases with the applied electric potential up to a threshold value where sparks are visibly produced. This phenomena is explained by the controlled voltage gradient provided across the oxide layer of the valve metals. Some oxides can support an electric field gradient of one million volts per meter or more. When the gradient exceeds a critical value a **charge cluster** (believed to consist of about one hundred billion electrons) is emitted from the electrode. This charge cluster is believed to be the active mechanism in the **Neal-Gleeson Process** that can be used to fission heavy metal ions and reduce radioactivity.

### INTRODUCTION

The discovery (patent pending) of the Neal-Gleeson Process has shown that naturally radioactive thorium and uranium compounds, dissolved in water to make an electrolyte can be stabilized by the use of an appropriate electrochemical reactor [1]. Kenneth Shoulders has shown that high-density charge clusters, under appropriate conditions, can cause nuclear reactions [2]. Sam Faile and Nicholas Reiter have shown that exploding bridge wires under atmospheric pressure and short high-potential electric discharges appear to produce some nuclear reactions [3]. Shoulders has further shown that the fracto-emission of charge clusters is a probable cause of the nuclear events and excess heat in cold fusion [2]. The combination of these experiments have been extended to explain a variety of current and projected experiments [4]. The basic premise proposed was that the creation of high-density charge clusters is much more prevalent than previously considered.

The authors have used the above premise to search the literature for some historic experimental and/or theoretical evidence of charge clusters. This paper reviews some of the literature dating from as early as 1957 (which cites a reference as early as 1906) to 1995. The few papers collected and cited in this article represent an exemplary but not exhaustive review of the historic experimental history of one version of the development of charge clusters.

### THE VALVE METALS

In the early days of the growing electron-tube industry, the first electronic tube used as a diode was termed a **Fleming Valve**. The term **valve** for an electron tube was more prevalent in England than in the United States. The early electron emitters were the hot filaments. Later, the cathode was added. To make an effective cathode, a combination of a conducting metal and a substance that was a good emitter of electrons (thermionic emission) was required. It was found that certain metal and metal oxides were suitable. Thus, the term **valve metals**.

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As the solid-state devices (diodes and transistors) became popular, there was also a studied search for material that would provide the basis for controllable displays. Therefore, considerable experimental work was done with luminescence of certain metal oxides. To quote van Geel et al. [5] in their introductory remarks: "It has been known for a long time (cites A. Günthershulze, *Ann. Phys.* vol 21, pp 929-954, c1906) that during the anodic oxidation of several metals, such as Al, Zr, Ta, Zn, W, Mg, a light-effect occurs. In this process the metal is placed as the anode in a suitable electrolyte; during the passage of current oxygen is developed at the anode, an oxide film is formed, whilst at the same time luminescence can be observed." These same oxide films (at least some of them) can serve as electron emitters when suitably heated. The name for this series of metals has been **valve metals**, although it is hard to find literature that defines what precisely is meant by a **valve metal**.

## BEYOND LUMINESCENCE

Some of the earliest papers we found [5] reported on experiments with oxide layers on aluminum. Later papers, such as Waring and Benjamini [6], Alwitt and Vijh [7], and Wood and Pearson [8], reported more generally on the development of luminescence and with the magnitude of the breakdown voltage on silicon and valve metals in general.

Some of the interesting observations include the following: van Geel et al., [5] report on the anode and cathode flashes when the aluminum oxide electrodes are powered with 200 cps alternating current at 25 volts. Waring and Benjamini [6] report on the phenomena of **breakdown** with the following comments: "Since the thickness of the oxide is proportional to the voltage ... there seems to be no reason to anticipate a limiting voltage or breakdown. We have, however, noted the increase in bubbling just before breakdown." Further, these authors note that: "Since the field during growth is about  $1.6 \times 10^7$  v/cm [sic] and therefore near the breakdown limit anyway, this increase is enough to cause the weaker spots in the oxide to give way, thus **showing the sparks**." In their conclusions, the authors state, "**Spark** discharges penetrate into the solution," and, "When **sparking** begins, the general glow decreases; the current and luminescence are concentrated in these limited, intense, breakdown locations."

Alwitt and Vijh report on the breakdown voltage (at which visible sparking was observed) for a variety of valve metals including high-purity aluminum, niobium, tantalum, and zirconium. With the electrolytes used, the breakdown voltages varied from 100 to 350 volts. The authors suggest that the breakdown voltage is related to the thermal and electrical stability of the oxides; that the sparks were associated with a strong field extending into the electrolyte; and that sparking may be the electrical discharge through gas generated at the anode surface.

Wood and Pearson [8] reported that no sparking is observed with their use of molybdenum and vanadium. Tantalum, niobium, zirconium, and hafnium all displayed the spark-producing phenomena. With the electrolytes used, the authors reported no sparking with aluminum, titanium, and tungsten. The breakdown voltages ranged from 118 to 300 volts. Alloying of pure metals appears to reduce the breakdown voltages. A table of values is given for various niobium alloys, showing breakdown voltages ranging from 40 to 270 volts. These experiments were performed on anodes.

S. Ikonopisov provides a ten-page critical review that covers galvanoluminescence, anodic glow, cathode flash, anodic flash, alternating current glow, and continuous cathode glow [9]. Ikonopisov cites the earliest work on galvanoluminescence as being reported in 1887. However, the phenomena of most interest to this review, the breakdown of the oxide layers, are considered "beyond the scope of the present consideration." However, the author does provide a rather complete list of metals that show some degree of galvanoluminescence: Ag, Al, Bi, Cd, Ce, Ga, Hg, In, Mg, Nb, Sb, Si, Sn, Ta, Ti, W, Y, Zn, and Zr with references for each element. Here are a few of the interesting comments: "...even minute traces of halide contamination are known to create punctures or conducting corrosion spots in the anodic film on Al." "The

anode glow brightness increases markedly on simultaneous UV excitation." "...the anode glow-spectrum is continuous while the scintillation spectrum at the sparking voltage consists of lines due to aluminum and oxygen." "The threshold voltage (of anode glow) appeared to be almost independent of the frequency in the range from 16 Hz to 5 kHz but the output decreases at audio frequencies." "UV emission has been observed during anodization of Al, Ta, Si, and Mg...." In aluminum oxide: "The photoluminescent brightness increases with the temperature of previous heating of the films, but heating above 700°C produces an irreversible loss of the photoluminescence." There is evidence that "...the presence of water is indispensable for the anode glow."

S. Tajima [10] reports on luminescence breakdown of anodic oxide films on aluminum. This author states, "Thus the hitherto unsolved problems which had caused confusion in complete understanding of the art are almost solved." This 1977 summary article with 94 references has been followed by about twenty more years and we still have some unresolved mysteries in aluminum luminescence. Tajima does sort out and provide tables of some of the various forms of luminescence which contributes to a better appreciation of the phenomena. The author writes about adding impurities to aluminum to modify the luminescence and reports, "The intensity increases up to 20-30 times by addition of Mn 1.5%." This observation suggests that experimenters may want to try some degree of doping in their investigation of the sparking phenomena. Tajima also observes, "It is well known that in the case of breakdown by electron avalanche, the increase of dielectric film thickness and its temperature rise lower the electric field strength needed for breakdown. ... Therefore, higher formation voltage and thicker barrier layer make easier the breakdown by electron avalanche." Further, the author cites others, "...electrolytic breakdown is apparently controlled at the oxide-electrolyte interface rather than by the bulk oxide." Tajima, in Table 2, lists breakdown voltages for different electrolytes ranging from 20 to 500 volts.

Szklarczyk, Kainthla, and Bockris [11] report on the use of highly pure water and high voltages to explore the breakdown of the water between platinum electrodes. In some cases, the electrodes were coated with wax. Data included voltages as high as 30,000 volts. The following conclusions were reported by the authors: "The breakdown potential is the potential at which the Fermi level of the cathode becomes equal to the energy of the bottom of the conduction band of liquid water. Under such conditions electrons can be readily transferred from the cathode to the anode without interaction with water molecules. At the anode, holes would enter the valency band for water.. The head of the so-called streamers is occupied by electrons moving in the conductivity band of water at a rate too high for chemical interaction to occur."

Xu, Tzeng, and Latham [12] report that substantial emission of diamond-coated molybdenum electrodes can be achieved at potentials as low as 5,000 volts per meter. A most important finding was that points were not necessary. With the diamond coating, it appeared that the emission occurred with graphite impurities embedded in the diamond film. A clever experimental arrangement used a transparent anode so that video camera pictures could be taken of the emissions.

A short paper by Bonifaci, Denai, and Atrazhev [13] reports on experimental findings with various electrolytes on the onset of photoemission as a function of voltage. By using a needle-pointed tungsten electrode, it was found that a field strength of 10,000 volts per centimeter occurred at the tip of the electrode. Schwirzke [14] reports on the emission of electrons from spots on a cathode when the voltage is about  $10^7$  volts per meter. The author provides important information concerning the formation of a unipolar arc where positive ions are generated on or near the cathode surface and emitted to return to the cathode surface. The author states, "Since the emitting spot cannot deliver  $j_{CL}$  [maximum current density] without turning itself off, it must be that  $j_{FE} < j_{CL}$ ." The symbol  $j_{FE}$  is the field-emitted current density. The author makes a strong point of the mechanism by which positive ions are produced at the cathode. Experiments were performed. The author indicated that, "The sound velocity of air at room temperature represents a lower limit for the expansion velocity of the desorbed particles." This statement suggests that the experiments were performed at atmospheric pressures. No data is provided to show variations as a function of the pressure of the gases

surrounding the arcing electrodes. However, the description of the formation of the unipolar arc suggests a method by which positive ions could be produced and entrained in a charge cluster and that this plasma sheath could be a part of the mechanism by which short pulses can produce toroid-shaped charge clusters.

Rousar et al. [15], reports on sparking at the cathode in electrochemical machining. The onset of sparking makes for a rougher surface and is to be avoided. Kirkici et al. [16], reports on the erosion of various films due to surface breakdown of the dielectric. It is important to note that the several different dielectrics (diamond on silicon oxide, diamond on silicon, diamond-like carbon on silicon oxide, diamond-like carbon on silicon, and silicon oxide on silicon) all suffer severe degradation after multiple repeated "shots" or breakdowns.

Hurlen and Gulbrandsen [17] show that the growth of the oxide layer is determined by both the growth next to the electrolyte surface and next to the metal surface. They note, "At the metal-oxide interface of a growing anodic film, the whole current is by metal ion transfer and possibly by electron transfer." Later in discussing equations the authors state, "...one should expect the anion transport to gain predominance at low current densities, and the cation transport to do so at high current densities." The authors also define a **passive film**: "...a steady-state passive film is considered to be a compact oxide film which has reached a steady thickness over which metal ions are transmitted from metal to solution at a steady rate, the potential independent steady-state passive current."

Gomer [18], citing 69 references, discusses the essential features and theory of field emission, field ionization, and field desorption. The concept of field emission is considered to be one of the key concepts in the creation of charge clusters. In the section on field emission in liquids, Gomer makes the following observation, "Although not exploited, to my knowledge, field emission can provide a simple method of electron injection into dielectric liquids without the use of either vacuum or high energy accelerators." His 1994 observation was certainly correct. Another observation, "It was also noticed that ion energy distributions were fairly narrow at low fields but broadened on the low energy side as the field increased. **This indicated very clearly that ionization then took place farther and farther in front of the emitter...**" Although these measurements were made in low-pressure gases, they may also be true in liquids, but at smaller dimensions. Gomer also notes a resonance effect that should be carefully studied by any who are working in low-pressure gases. See the potential energy diagram for electrons in an atom near a metal surface, as depicted in Fig. 1 (after figure 10 in Gomer's paper). In his section on field ionization, Gomer notes, "Field emission and field ionization are almost certainly the mechanism of dielectric breakdown in liquids."

One of the models for the onset of field emission is the presence of points, corners, whiskers, or other **projections** on the surface of the material providing the field emission. Jimenez et al. [19], provides evidence for the **projection** model on large-area cathodes. This careful study, using Nb and Au surfaces, supports the concept of **projections** being a useful source for field emission. However, the paper does not cover the high fields necessary for sparking. In contrast, Xu [12] found that with diamond coatings, the spots for field emission were often valleys. This difference was explained by the possible embedded graphite particles in the diamond layer.

Dumin et al. [20], discuss the breakdown of thin oxide layers by high field emission. This paper presents a model of the breakdown of an oxide layer that includes thickness, field, polarity, time, and temperature attributes. However, the article is mainly directed at the semiconductor industry. Metikos-Hukovic discusses the behavior of tin as a **valve**

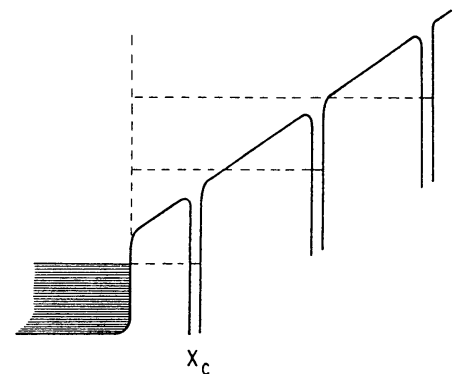


Fig. 1 Potential energy diagram for electrons in an atom near a metal surface in the presence of an applied field, illustrating the resonance effect in field ionization.

**metal** [21]. Some of the tin oxides are transparent. This feature was used by Xu et al. [12], in a clever method for viewing a cell's operation through a transparent anode window. Plastic deformation of cadmium affects the electroluminescence according to Dragon et al., in their 1995 paper [22].

More recently (1995) Ord and De Stet [23] describe some typical properties of **valve metal oxides** in their study of an ideal transparent anodic oxide film on zirconium. The oxide film grows by a high field conduction mechanism and the potential and film thickness increase linearly with time under constant-current growth. **Valve metal oxides** have a high relative permittivity, are strongly affected by electrostriction, and are optically anisotropic. The oxides exhibit the property of electrochromism, i.e., during cathodic reduction hydrogen is inserted into the oxide forming an optically-absorbing outer layer and the film is returned to its initial transparent state by subsequent anodic oxidation. The film is formed in a carbonate buffered aqueous electrolyte.

Hummel et al. [24], discuss the effect that spark-processing has on later cathode luminescence. The basic effect of spark-processing is to shift the wavelength of the cathode luminescence toward the blue end of the spectrum. Using a stream of compressed air during the spark processing also increases the brightness of the luminescence by as much as an order of magnitude.

A variety of papers have addressed the acceleration that can be provided to electrons by the **breaking of relativistic plasma waves**. Modena et al. [25], report on their efforts to provide improved electron acceleration by making use of the phase velocity of a plasma wave. Up to 44 MeV of accelerated plasma electrons has been achieved by this method. However, in the judgement of the authors of this paper, the creation and acceleration of electron charge clusters which carry positive ions is a more splendid method of creating high impact energies with relatively low input energies.

Jones and Kunhardt (Polytechnic University, Farmingdale, N.Y.) report on the development of pulsed dielectric breakdown in liquids [26]. This paper presents a thoughtful model of the four stages of the breakdown of liquids: 1. Formation of a low-density site near the electrode. 2. Growth and expansion of the local density so that ionization can take place. 3. Electron avalanche and formation of an ionizing front. 4. Propagation of the ionization front. The authors derive mathematical expressions for each of these stages of dielectric breakdown.

The authors would like to suggest a rather novel approach to the formation of bubbles and the **breakdown** phenomena. Julian Schwinger, a Nobel prize winner, suggested that the Casimir energy in filling a dielectric hole is the source of coherent sonoluminescence [27]. The concept is that a bubble of vapor in a dielectric fluid begins to diminish in size as the vapor gradually returns to a liquid state. At some radius of the bubble the Casimir forces become highly predominant and an very strong local blast of energy is produced (such as blue light) as the bubble accelerates to a collapse. The authors of this review paper suggest that the inverse procedure may be the source of some of the observed phenomena associated with the generation of an electron charge cluster.

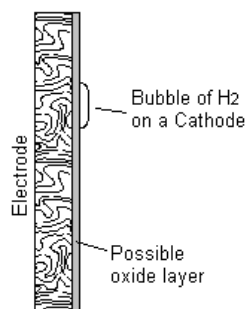


Fig. 2

As shown in Fig. 2, electrical energy is fed into a bubble at some local spot on the surface of an electrode. Because of the Casimir force (which is proportional to the fourth power of the radius), the formation of the bubble requires a considerable amount of local energy to be produced and fed into the bubble formation to overcome the energy of the Casimir force. As the bubble increases in size the expansion forces generated inside and stored in the bubble will overcome the Casimir force and explosively inject the energy into the local region near the electrode. This release of energy may help form the electron charge cluster. As shown by Shoulders [28], an electron charge cluster can ionize the local medium. In fact, it appears that the charge cluster can both provide energy to its

environment, especially by electron emission, and also maintain its own energy. The source of that energy may well be the space energy that produces the Casimir force itself. In this proposed method of breakdown of a dielectric liquid, the electron charge cluster is produced and the charge cluster itself becomes the means for ionizing, at least locally, the dielectric fluid. Apparently, the charge cluster will be expelled from the electrode (especially electrodes having a high-resistance layer where a strong electric field can be generated) into the fluid through the formation of a bubble, often at a surface projection or whisker.

Those who experiment with the creation of electron charge clusters in a dielectric liquid will, hopefully, be guided by reading some of the extensive literature, such as listed in the references to this article. When creating charge clusters to be used for the promotion of nuclear events in an electrolyte, the following protocols should be considered:

1. Sparking at the electrode is necessary but not sufficient for the production of nuclear events. Apparently a charge cluster can produce an observed spark but fails to have sufficient energy to promote a nuclear reaction.

2. It appears necessary to maintain (or periodically renew) the oxide layer on a valve metal to produce nuclear-active charge clusters. It is, of course, the concept that the charge cluster must carry piggy-back positive charges and must achieve a critical energy level to promote nuclear reactions [2].

3. The molarity (and the resulting conductivity of the electrolyte) may be an important operational parameter. The concept is that the charge cluster must be able to persist for some short time period and energetically impact a nucleus in the electrolyte to be able to produce a nuclear reaction. It is believed that the potential gradient between electrodes must be maintained at some critical value for nuclear reactions to be favored. A lower field gradient (higher conductivity) in the electrolyte may lead only to Joule heating and not to the desired level of nuclear reactions.

4. Experimental evidence suggests that the hydrogen and oxygen nuclei are involved in the sometime multiple or sequential nuclear impacts that result in the nuclear reactions.

5. The Coulomb barrier may be much less than the field strength of a charge cluster. The charge cluster must have sufficient electrons so that this field strength can aid in overcoming the Coulomb barrier before nuclear reactions can be expected.

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