

Chapter 2

Electroneutrality: When and Where?

Contents

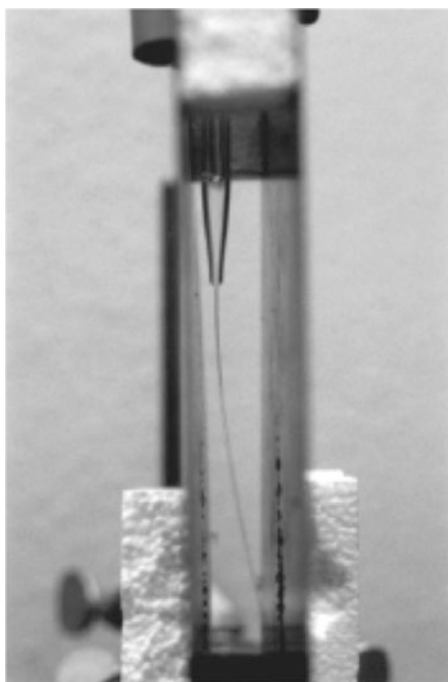
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2.1 A Widespread Belief

Many persons educated in science and engineering tend to view their surroundings as being electrically neutral, except for the electricity used to power equipment and lighting ware in any anthropic environment. When asked about electricity in a quiescent environment, scientists and engineers often invoke electroneutrality, including an “Electroneutrality Principle” that will be discussed ahead.

This is not surprising, since unbalanced electrostatic charge is hardly conspicuous: it is not perceived by our senses, it does not provoke frequent sound emission or color change and some more frequent visual effects are too subtle to be noticed,

Fig. 2.1 Electrical deflection of a distilled water droplet stream within the electric field between two aluminum deflector plates separated by 2 cm Styrofoam spacers. The plate on left was grounded and a potential was induced on the right-hand plate by a statically electrified balloon that was outside the field of view to the right. Reprinted with permission from [1]



as for instance in the case of water drop deformation due to excess charge. This will be treated in detail in Chap. 6, but a short description follows.

Water drops are deformed in the presence of an electrified body and the trajectory of falling water is modified, in the presence of an electrified object, as shown in Fig. 2.1. This has been known for more than two centuries and it is used as a demonstration of molecular polarity in textbooks. Unfortunately, this is eventually accompanied by incorrect explanations, as discussed by Ziaei-Moayyed and Goodman [1]. Some related movies that can be seen in the Internet are supplemented by comments and explanations that are examples of misconceptions on electrostatic phenomena. A microfluidic version of this experiment accompanied by a quantitative analysis was recently published [2], see Fig. 2.2.

2.2 Charge Accumulation, Electrostatic Discharge

Electrostatic charge accumulation usually goes unnoticed, until a critical value is reached for the electric field and a discharge takes place, often leading to fire, explosions, and personal injuries. This kind of behavior is quite different from many other types of physical phenomena. For instance, an object undergoing mechanical stress, torsion, or compression normally shows some deformation, prior to breaking or acquiring a new shape due to plastic deformation. When water is heated, its

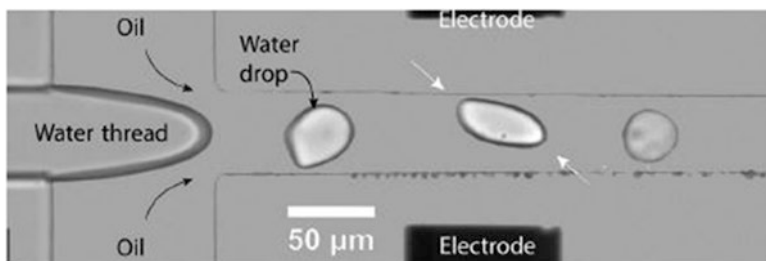


Fig. 2.2 Water droplets in microchannels as they pass between two electrodes. Droplet shape becomes elliptical and they tend to emit daughter droplets, as in the classical Rayleigh explosions. Reprinted with permission from [2]

temperature increases before it starts to boil. On the other hand, unwanted electrostatic discharges take place without any warning: most systems will build-up electric charge that goes unnoticed, until the limit is reached.

A popular class demonstration experiment is the “Kelvin dropper”, an ingenious but simple experimental set-up where falling water drops produce sufficient electricity to provoke sparks. Most reports of this experiment are rather qualitative since voltage build-up is not usually measured, but it has recently been used in microfluidic devices [3] with attractive features, including the possibility of energy scavenging.

Rather dangerous, electrical discharge may be triggered just by making some geometrical change in the arrangement of solids and liquids, for instance by bringing a grounded conductor closer to a pile of charged dielectric powder. The electric field may increase to the point of provoking dielectric rupture of the air and thus provoking a discharge. So, performing what may look like a safety procedure may indeed trigger an explosion. The situation may become still more complex if this is coupled to motion relative to a magnet that will also provoke charge displacement [4].

2.3 Electric Potential, Electric Field, Electrochemical Potential

A complicating factor in the understanding of electrostatic phenomena is the mixed use of electric field or electric potential to describe electric effects on various phenomena. Electric field fits well within mechanical reasoning, since it is easily associated with forces acting on charges and dipoles and thus it is easily related to the motion of charge carriers or the orientation of objects containing separated charges, as dipoles or multipoles.

On the other hand, electric potential fits easier within thermodynamics and its use is convenient, as an intensive factor coupled to charge, which is extensive. It was used by Tolman, in his outstanding work, e.g. on the determination of electrochemical transference numbers of ions by centrifugation. In his writing,

“The method consists in the measurement of the electromotive force produced between electrodes placed at the central and peripheral ends of a rotating tube containing the electrolyte. An equation can be derived, connecting this electromotive force and the transference number of the salt with the speed of rotation, the density of the solution, and the molecular weight and the “partial” specific volume of the substances involved...” [5].

The existence of sedimentation potentials, by itself, is a powerful demonstration that electroneutrality is not expected along any solution or dispersion contained within a tube that underwent centrifugation. Simple extension of Tolman’s equations to larger solutes or particles dispersed in a liquid shows that it should not also be expected along the depth of any water body where charged particles (clays, sand, humic matter) are dispersed, settling under gravity. So, electric potential gradients are expected even under sedimentation equilibrium conditions, in aqueous electrolytes.

Electrochemical potential is currently widely used by two groups of researchers: electrochemical kineticists and electrophysiologists. Both groups are well aware of the importance of the electrochemical potential in the treatment and understanding of the phenomena they are interested in: electrochemical kinetics makes explicit use of the effect of local electric potential on the activation energy of an electrode reaction, while the importance of electric phenomena in signal transmission in living cells and tissue is now perceived even by the layman using medication related to sodium pumps and other nanobiological gear. However, they are a minority. This is verified by comparing the number of entries in the Web of Science, over all years recorded, at a given time: entries for “chemical potential” number 14,180 vs. 2852 for “electrochemical potential”.

Instrumentation researchers have been familiar with non-uniformity of electric potential in insulator surfaces. For instance, Lion stated that “The (usually irregularly distributed) electrostatic charges on the surface of an insulator can be detected and measured by the use of a moving electrode” [6]. His recognition of charge non-uniformity was not widespread and it is usually ignored by experimenters using plastic labware.

2.4 Taking Electroneutrality for Granted

Examples of researchers treating excess charge in matter as a non-issue can be taken from any scientific or technological area: for instance, polymer textbooks only refer to electric charge in the chapters explicitly related to ionic species, either on polyelectrolytes or living polymerization initiated or involving ionic species, notwithstanding the obvious and frequent occurrence of excess charge in polymer materials exiting mills and processing equipment.

Charge build-up during operations widely used in the chemical, pharmaceutical, food, and related industries has been reported. Particles are often electrostatically charged by frictional contact during powder-handling operations [7]. More generally,

during the operation of multiphase systems such as fluidized beds, electrostatic charges are generated when the materials involved are dielectric in nature [8]. The same is observed during a melt agglomeration process [9] and in many other cases.

On the other hand, recent work from the author's laboratory shows that water is often non-electroneutral and that it contributes in many ways to impart non-electroneutrality to other substances [9, 10] that will be dealt in a separate chapter.

2.5 The Electroneutrality Principle

This principle is not often mentioned in the literature. A search in Web of Science using “electroneutrality principle” as the entry returned only 58 documents and in many cases electroneutrality is taken as a rule or principle, without further analysis. For instance, the statements “The main physicochemical principle which must be accomplished in body fluids, is the rule of electroneutrality” [11] and “The mathematical model is based on principles of mass conservation, acid-base equilibria, and electroneutrality” [12] are read in the recent literature.

This principle is an entry in IUPAC Gold Book [13] where we can read, under “electroneutrality principle”: “The principle expresses the fact that all pure substances carry a net charge of zero.” This is correct for “pure substances” but strictly under some assumptions, e.g. zero electric potential [14]. Under non-zero potential, satisfying the condition of minimum electrochemical potential requires that any system contains excess cations under negative potential and excess anions under a positive potential.

The assumption of electroneutrality appears explicitly in recent scholar texts, e.g.: “Ions are not independent in ionic solids, where we take for granted the fact that there are exactly equal numbers of Na^+ and Cl^- ions (or we would be electrocuted each time we salt our food)...” and “One *always without exception* dissolves a strictly neutral salt in water to make an ionic solution...no violation of electrical neutrality is significant in chemical units...” [15]. The electrocution argument is certainly not correct, because this implies the onset of electric current but this is not to be expected from the immobile crystal ions. Moreover, given the size of Avogadro's number, having exactly equal numbers of Na^+ and Cl^- ions implies that no imbalance takes place within a population in the range of 10^{23} individuals, what is not to be expected.

A related issue is the widespread assumption of non-volatility of ionic species, from room-temperature liquids. This matter was discussed in detail by Rockwood [16], connecting it to thermionic emission, and recalling that ions may undergo thermionic emission [17]. The same physics developed by Fermi for the treatment of an electron gas in equilibrium with a metal must apply to ions as well as to electrons, which implies that ions must also have a vapor pressure that is usually neglected. Evidence in favor of ion transfer across solid- and liquid-gas interfaces will be presented, in various chapters of this book, especially Chaps. 4 and 6, on Charge Mobility and Hygroelectricity, respectively.

2.6 Pauling's Principle of Electroneutrality

Pauling's principle of electroneutrality played an important role in the study of molecular structure. The original statement by Pauling in 1948 says that each atom in a stable substance has a charge close to zero and it appears in the Dover reprinting of the 3rd edition of his "General Chemistry" book as follows: "Stable molecules and crystals have electronic structures such that the electric charge of each atom is close to zero. Close to zero means between -1 and $+1$ " [18]. This principle has been used to predict relative stability of molecular [resonance](#) structures, to explain the stability of [inorganic complexes](#), [\$\pi\$ -bonding](#) in compounds and polyatomic anions. However, modern computational techniques may indicate greater ionic character than predicted by this principle [19].

To the present authors, it is not clear how and why this statement may lead anyone to believe that macroscopic matter is also electroneutral. It is well known that the collective behavior of molecules is often not predictable just considering the properties of individual molecules. Moreover, the advent of nanotechnology made clear that size matters and many properties of a given substance may change with size. For these reasons, Pauling's principle of electroneutrality is not a sound basis for assuming that matter is electroneutral, notwithstanding its importance in the determination of molecular structure and properties.

2.7 Factors of Non-Electroneutrality

2.7.1 *Dangling Bonds*

Many scientists and engineers were taught to view common matter as if it were a more or less complex assembly of great numbers of well-behaved ions and molecules, in the Avogadro range. Some situations that do not fit within this picture are usually overlooked. For instance, dangling bonds are structures not contained within the usual descriptions of the molecules and crystalline solids [20] and they have been reported in many cases: at the ends of graphene molecules [21]; in silicon [22], and germanium [23]. OH dangling bonds were detected in the hydration shells around dissolved nonpolar (hydrocarbon) groups [24]; in polymers, e.g. PTFE [25] where it accounts for ferromagnetism; in GaN [26].

Recognizing the existence of dangling bonds in rather simple systems that are among the most intensively studied by chemists, physicists, materials scientists, and engineers should prompt researchers to accept other departures from standard descriptions, including that there are not "exactly equal numbers of Na^+ and Cl^- ions" in NaCl crystals.

2.7.2 *Are Ionic Crystals Electroneutral?*

Crystalline solids themselves have a large number of possibilities for developing excess charge, positive or negative. A fundamental reason is entropy. It is well established that defects develop spontaneously on crystals, since they contribute to increase entropy and to decrease free energy. This often goes at the cost of adding energy to the crystal to create the defect.

A very simple case is a salt crystallizing from aqueous solution. If electrostatics were the only factor for ion deposition on the growing crystals, then perhaps each growing crystal would be exactly neutral at each point in time. However, a neutral salt crystal within a solution of its ions is a rare situation. This is abundantly evidenced by considering surface properties of salt crystals in the presence of dissolved ions. In this area, Lyklema and his collaborators in Wageningen and elsewhere produced a huge amount of rigorous information on excess charge on silver iodide crystals, determined either by potentiometric titration or more indirectly by zeta potential measurements. They examined adsorption and double layer capacitance [27] in the presence of various other ions, polymers [28] under variable pH, co-solvent and variable temperature [29] during five decades, creating solid ground for understanding the behavior of real ionic crystals in water.

Other important contributions come from different groups. For instance, “in the vicinity of the point of zero charge the Nernst equation accurately gives the surface charge” [30].

Indeed, the interaction of water with various ions at crystal surfaces cannot be expected to be non-specific. Silver iodide has also been studied in this respect due to its important role nucleating ice formation, in the atmosphere [31]. A recent finding is that “Water molecules strongly adsorb onto the Ag^+ terminated face to give a well-ordered hexagonal ice-like bilayer that then acts as a template for further ice growth”, but water does not adsorb at I^- terminated basal face or the prism and normal faces [32]. A study of the nucleating activity of silver iodide in super-cooled water showed its dependence upon the potential-determining silver and iodide ions [33].

Apparently, all the knowledge referred to in previous paragraphs is left aside when students are taught to use electroneutrality in their calculations, indiscriminately. For instance, they learn that in equilibrium reactions like $\text{AgI} = \text{Ag}^+ + \text{I}^-$ the two concentrations are equal, $[\text{Ag}^+] = [\text{I}^-]$ and $K_w = [\text{Ag}^+]^2 = [\text{I}^-]^2$. However, under most conditions the AgI particles contain non-zero charge, evidenced by electrophoresis, titration data, and other data. Consequently, $[\text{Ag}^+] \neq [\text{I}^-]$.

2.7.3 *New Ion Sources for Mass Spectrometry*

Mass spectrometry is currently a large family of analytical techniques with a great diversity of equipment configurations, procedures, and areas of interest. The initial step in the analysis of neutral molecules by mass spectrometry is the production of

molecular ions that formerly was done under high vacuum and this was an important limitation for mass spectrometry. In 1994, Hirabayashi and collaborators [34] revolutionized mass spectrometry by introducing an atmospheric pressure chemical ionization (APCI) technique termed “sonic spray ionization” (SSI) that “was unique and revolutionary because it introduced a new concept of ionization to mass spectrometry” producing ions without the assistance of voltage, radiation, or heating. The charged droplets were produced simply by spraying an acidified solution of the analyte in methanol at sonic speed and charge separation was assigned to a statistically unbalanced distribution of cations and anions. This opened the way to ambient mass spectrometry techniques like EASI that brought mass spectrometers into the “real world” for the following reasons: (1) its great simplicity, because only compressed nitrogen or air is required; (2) its ability to simultaneously produce both negatively and positively charged droplets, hence no need to switch high potentials in changing from EASI(+) to EASI(-); (3) the low charge concentration on the droplets, which seems to improve signal-to-noise ratios; (4) the extreme softness of the ionization process; (5) no thermal degradation. One limitation of EASI is the ultra-high-velocity spray stream, which can easily blow samples away, but the formation of bipolar aerosol using only low-pressure systems was recently disclosed [35] and it may find applications in MS, in the near future.

2.7.4 Contact Charging, Mechanochemistry, Tribochemistry

Contact charging is known to almost every person, it has been known since the ancient Greeks but its description in terms of the atomic-molecular theory as it was established nearly one century ago is still a big challenge [36]. Nevertheless, it shows that each one of two solids acquire excess charge upon contact, meaning that neither becomes electroneutral.

Beyond contacting other solids and liquids, any substances and materials in the real world are subject to handling, contact with other substances including gas and liquid flow over their surfaces, cutting and milling, tension and compression and a number of other kinds of mechanical actions. Persons experienced in different kinds of operations, in any environment, are familiar with the appearance of charged particles, liquids and solids, in many situations, from cement to pharmaceutical and food processing [37, 38].

Following IUPAC, the term “mechano-chemical” is used for a “Chemical reaction that is induced by the direct absorption of mechanical energy.” The Gold Book also notes that “shearing, stretching, and grinding are typical methods for the mechano-chemical generation of reactive sites, usually macroradicals, in polymer chains that undergo mechano-chemical reactions” [13]. Judging by the frequent observation of electrostatic charging in mechanically processed materials, many mechano-chemical reaction products carry excess charge that provoke various effects in the making of various types of solids and powders [39].

However, electrostatic separation based on different tribo-electric charging behaviors of components has emerged as a novel, sustainable dry fractionation process [40].

Milling solids provide many good examples, since the resulting powders are often sufficiently charged to adhere to equipment surfaces, blocking its chambers and provoking the interruption of the operation [7]. Since industrial milling operations use up to 5% of the electricity produced in the world, and it dissipates hitherto unknown amounts of charge in the environment, perhaps this would be a good area for introducing energy harvesting equipment.

Unfortunately, mechanochemistry, tribochemistry, and even tribology at large are not popular areas among basic scientists, even though there are excellent reviews and books on this topic [41–46], bringing information, showing opportunities in chemical synthesis [47, 48] while aiming to create a unifying framework that enables predictions of force-induced reactivity [49, 50]. This topic will be treated in a separate chapter in this book and the message left at this point is this: common mechanical action on solids and complex liquids may cause significant chemical change, including the formation of high-energy species like free-radicals and ions that may soon react forming other high-energy species with half-lives reaching many months, in some cases. This is well represented by the words “triboplasma”, coined by Heinicke and “magma-plasma”, by Thiessen, to describe the unique chemical and physical environments created by mechanical action on different solids.

2.7.5 Liquid Junction Potential and Membrane Potential

Liquid junction potential (LJP) is a concept familiar to electrochemists, electrophysiologists [51], and to any person that uses a combined glass-reference electrode for pH measurement. It appears when a liquid–liquid junction is made, contacting two liquids with different concentration and chemical composition of electrolyte solutes [52], or even two different liquids [53].

It is easily understood considering that the diffusion coefficients of the cation and anion in an electrolytic solution are usually different, as well as their electrophoretic mobilities and transport numbers as shown in Fig. 2.3. The magnitude of junction potentials cannot be overlooked and it has been demonstrated that LJPs formed in microchannels can induce appreciable electrophoretic transport of charged species without the use of electrodes or an external power supply [54].

Membrane potential is observed whenever a membrane separates two electrolytic solutions. This is important in various natural and technological processes and it has been widely used in chemical analysis, in the glass electrodes [55] for pH and other ions measurement and various membrane electrodes that were developed for a large number of analytes [56]. Moreover, it is a basic concept in cell biology [57], ions may flow across the membrane by active and passive mechanisms and the changes in the membrane potential trigger important physiological phenomena.

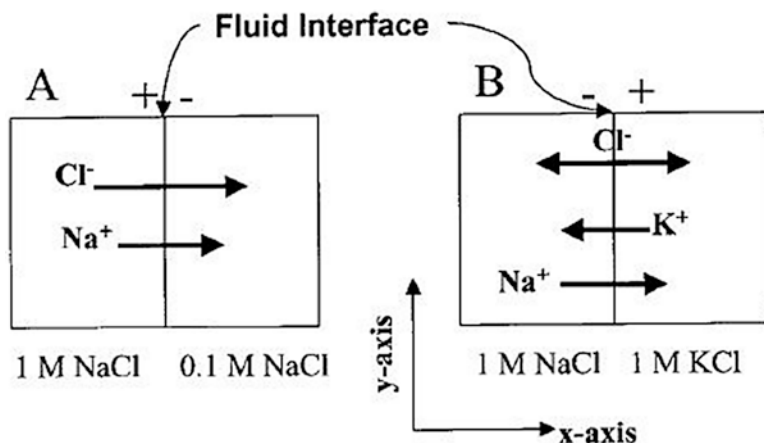


Fig. 2.3 The origin of the liquid junction potential (LJP). *Left:* chloride ions diffuse faster than sodium ions creating a potential gradient (negative at right) *Right:* there is no net flux of chloride ions but sodium diffuses faster than potassium, producing positive potential at right. Reprinted with permission from [53]

Liquid junction and membrane potential are components of the “spontaneous potential” that appears during drilling operations in the oil industry. It was one of the first logging measurements ever made and it was discovered by accident, since it caused perturbations on the electric logging systems. It records the naturally occurring voltage produced by the interaction of connate water, drilling fluid, and shale. Its usefulness was soon realized, and it remains as a useful logging measurement, after many years [58]. In this case, the “membrane” is a layer of shale or sand.

2.7.6 Electrostatics in Chemical Processing

Electrostatic phenomena are well known to chemical engineers involved with gas–solid–liquid flow systems [7], as they cause various types of operational problems and are a source of hazard. Electrostatic charging has been observed during spray-drying [59] and fluidized bed operations [60] where charge and hydrodynamics are mutually affected and excess accumulation of electrostatic charges has a severe impact on hydrodynamics [61].

2.7.7 Electrostatics in Soft Matter

Soft matter provides a large number of examples of systems whose properties and related processes largely depend on charge partition and segregation, this means they show large domains that are non-electroneutral. They include high polymers

and elastomers, polyelectrolyte gels, food processing and storage, waste management, new tailored materials that easily acquire and store excess charge.

Even more important, electrostatic interactions are essential for biopolymer structure and they are thus at the heart of all the biotechnologies that currently have a great impact on human life [62]. Their relevance is now sufficient to attract the attention of theoreticians going deeply into systems that were previously often considered too complex for a proper scientific analysis. As a result, counter-intuitive phenomena like charge inversion of particles and polymers are now understood as induced by correlation [63].

2.8 Conclusions

The question in the title of this chapter can now be answered: electroneutrality is not to be often expected and if so, in very few places. It is not the rule and its conceptual use in most material systems neglects a large amount of sound scientific information.

The evolution of ionization techniques in mass spectrometry shows how great progress was achieved just by recognizing that electric charge separation takes place during simple phenomena under many conditions, in the laboratory and environment. This perception has been obstructed in many cases by the presumed validity of an “electroneutrality principle”, even in the absence of significant experimental or theoretical support.

The examples given in this chapter show how charge partition takes place in macroscopic, microscopic and mesoscopic systems, under equilibrium or non-equilibrium conditions. Consequently, any time a material system is approached, we should ask ourselves which electrostatic patterns it contains and how these change with time.

Electrostatic patterns are all over, they are not rare and irrelevant oddities that are not even mentioned to most science and engineering students. Given their pervasiveness, electrostatic patterns and their consequences can and should be clearly mentioned by lecturers teaching many different disciplines and they should be considered by researchers and engineers, in every area of scientific endeavor.

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