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Gaston Planté and his invention of the lead-acid battery—The genesis of the first practical rechargeable battery

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ABSTRACT

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Keywords: Lead-acid battery, history Gaston Planté Accumulator Secondary battery Rechargeable battery In 1860, the Frenchman Gaston Planté (1834–1889) invented the first practical version of a rechargeable battery based on lead–acid chemistry—the most successful secondary battery of all ages. This article outlines Planté's fundamental concepts that were decisive for later development of practical lead–acid batteries. The 'pile secondaire' was indeed ahead its time in that an appropriate appliance for charging the accumulator was not available. The industrial success came after the invention of the Gramme machine. In 1879, Planté obtained acceptance for his work by publishing a book entitled *Recherches sur l'Electricité*. He never protected his inventions by patents, and spent much of his fortune on assisting impoverished scientists.

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1. A portrait of Gaston Planté the man

Raymond Louis Gaston Planté was born in Orthez, Department of Basses-Pyrénées, in the very south-west of France on 22 April 1834, and died in Bellevue, near Paris, on 21 May 1889. A photograph of Planté is shown in Fig. 1.

1.1. Ancestry

The Planté family can be traced back, to the sixteenth century [1]. The Béarn, a province on the northern foot of the Pyrenees, was famous for its love of liberty, and was often keenly defended by its own Parliament and its local administration. The last Count, called 'our good Henric' became King Henry IV of France in 1589. On his death in 1620, the Béarn was attached to the Kingdom of France.

Protestant Jean Planté, Gaston's grandfather, emigrated to Spain in 1759, and earned money as a ship-owner in Bilbao and Santander. He married a Spanish lady of high lineage and was ennobled by the Spanish King Charles IV. In the latter years of his life, Jean settled in Orthez, the picturesque former capital of his native region of Béarn. The large house, which belonged to the family until the end of 1959 following the death of the last Planté proprietor, had a most beautiful door that was adorned with magnificent carvings and pillars. In 1814, when the English and Spanish invaded the South of France, a bullet burst through the handsome portal. Afterwards, the Duke of Wellington and Maréchal Soult, the opponents of the important Battle of Orthez, alternately dwelt in the house.

Raymond Planté, Gaston's uncle, and other descendants stayed in the country and during most of the 19th century they were Deputies of the Basses-Pyrénées and Mayors of Orthez.

Pierre Planté, Gaston's affluent father, was a cultured man with a great love of music. On the first floor of the house, over and a little to the left to the abovementioned door, Pierre's son Léopold was born in 1832, then Gaston (1834), and finally Francis (1839). Respectively, the three brothers became famous in different fields, namely, law, physics, and as one of the finest pianists of France. Léopold would later become an eminent barrister in Paris. The 11year-old Francis gained the first prize in a contest at the Paris Music Conservatory. The famous virtuoso was later known as the 'God of the piano', and gave concert recitals until the age of 91.

1.2. Education and academic career

The gifted, 6-year-old Gaston moved to Paris with his family in 1841, where there were better schools to cultivate his diverse talents, which included drawing and making music. It was said that if Gaston had practiced a little more, he would have become equal to his brother Francis on the piano. Gaston, however, turned his attention to literature, natural sciences, mathematics, and physics. He attended a private school and then the Lycée Charlemagne, where he received his Bachelor of Letters at the age of 16, and a Bachelor of Sciences at 19. Gaston wrote and spoke fluent English, German,

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Jasson Stanty

Fig. 1. Portrait of Gaston Planté and his signature from the English edition of his book Recherches sur l'électricité [3].

Spanish, and Italian. He read both Greek and Latin and studied the literature of several European countries in their native languages. One time, he astonished Professor Bjerknäs by talking to him in Norwegian about his thoughts on hydrodynamic vibrators.

The Sorbonne, the most well-known university of Paris, granted the degree of Master of Science to him in 1855. Gaston's professors noticed his remarkable intellect and his dexterity in practical work. Consequently, in 1854, 34-year-old Alexandre-Edmond Becquerel, the father of the later Nobel laureate Henri Becquerel, invited 20-year-old Gaston Planté to be his laboratory assistant at the Conservatoire des Arts et Métiers.

Planté's paleontological work and his first scientific discovery were completely different from his later career in electrochemistry. Fossils of a prehistoric flightless bird, which he had found in a quarry with 'argile plastique' formation deposits at Meudon near Paris, caused considerable excitement in 1855 because of its singularity. The large species, which had lived roughly 55 million years before, was finally named *Gastornis parisiensis*, after Planté's forename, by the French Académie des Sciences in agreement with the British Museum. In the 1870s, similar species were found in North America. Gaston continued to search and found plant fossils in the Paris basin that he gave freely to the Natural History Museum.

Planté was described as 'a studious man of zeal' by the French geologist Louis-Constant Prévost. Gaston liked to journey abroad and observe in minute detail the foreign customs and different ways of living. His cosmopolitan character was a rarity among his countrymen and also his contemporaries in the neighbouring countries who preferred strong national allegiances. On the advice of Lord Chesterfield, he took notes in the language of the country being visited. Until the 1860s, Planté kept diaries in his fine handwriting. These contain profound insights into the thoughts of the young Gaston, humoristic verses, considerations on ethics and, of course, detailed descriptions of his travels and correspondence in different languages. He was religious and modest and imposed on himself the will and patience to conform his life to following his beliefs. Advanced in years, the helpful and generous Gaston was considered by some people who had known him, as a kind of saint. Due to his skilfulness in practical work, Planté was invited to perform electrical experiments, which included a demonstration of Rhumkorff's electromagnetic machine to Emperor Napoléon III and his wife in the Palais des Tuileries in 1858. To be 'mixed up with royality' did not seem to impress him much, as he recorded in English in his notes. At the end of that year, he quitted his position as Becquerel's assistant and set up his own research laboratory in his apartment, No. 56 rue des Tournelles, close to the place des Vosges and to the rue de la Cerisaie, in the old Parisienne quarter of Le Marais, where the illustrious Benjamin Franklin had already worked on electricity in the *Hospital des Célestins* at the end of the 18th century. Scientists and ordinary people, who visited this laboratory, were filled with admiration looking at the state-of-the-art of science that Planté presented with such simplicity and authority.

1.3. Life-task electrochemistry

In March 1860, Planté presented his seminal lead-acid battery to the Académie des Sciences, the principle of which he had elaborated the year before. Nine elements in a wooden rack, with the terminals connected in parallel, delivered remarkably large currents. Until 1875, Planté attempted many designs of lead-acid cell under various operating conditions such as short circuit, trickle charging, and float charging. He made the first serious analysis of 'polarization' in an oxidising medium (see Section 2).

Planté published the results of his investigations in Comptes Rendus de l'Académie des Sciences and in various periodicals. He then gave an account of his broad experience in his book Recherches sur l'Électricité [2], which was published in 1879, and in a second edition in 1883 (Fig. 2). The English translation of the book [3] was published under the title 'The Storage of Electrical Energy and Researches in the Effects Created by Currents Combining Quantity with High Tension' a few years later and quickly became popular in the British Empire. This work is a model of clear language and elegant demonstration that is still of interest today. The book was dedicated to Don Pedro II d'Alcantara, Emperor of Brasil as a 'feeble witness of my profound thanks. You were the first to encourage my work'. The technology-keen monarch had visited Planté in his laboratory twice in 1877. His Majesty's invitation to demonstrate the rheostatic machine during a dinner in front the marvelling guests resulted in Planté being awarded of the Order of the Rose of Brazil, and in receiving financial patronage.

Modestly the book begins with these words of St. Augustine: 'Quaero, pater, non affirmo': I question, Father, I do not affirm. Remarkably, in 1879 Planté had already a vague idea of the corpuscular nature of electricity before the existence of the electron was proved.

'336. Conclusions relating to the nature of electricity. The analogies which we have just enumerated permit us, we think, to consider electricity as a purely mechanical motion of ponderable matter. This movement consists in the extremely rapid flow, or transport, of a very small quantity of matter, in regard to the electric spark, the voltaic arc or electrical discharge in general.' [3]

George Johnstone Stoney's concept [4] of a fundamental unit of electric charge and the rough estimate of the value of the 'unit of electricity' was due to his work completed in 1874, which was finally presented at the British Association for the Advancement of Science meeting at Belfast in 1881. Hermann von Helmholtz drew attention to the existence of an elementary charge in 1888. The name of the electron may have only first appeared in Stoney's paper of 1891. The discovery of the electron, as a particle, is associated with J.J. Thomson's determination of its charge to mass ratio in October 1897.

From 1875, Planté directed his research toward high voltage discharges, electric phenomena in the atmosphere, and applica-



Fig. 2. Title page of Recherches sur l'Électricité: Planté's book on secondary batteries and phenomena related to high-voltage power sources.

tions in therapeutic treatment. Early Planté accumulators heated the platinum wires of the 'galvanocaustic cutting blade' for dentists and surgeons, and powered laryngoscopic devices. Gustave Trouvé assisted in developing a polyscope with a fine piece of glowing platinum inside a little reflector, instead of a light bulb. Planté's battery was also used for miners' electric lamps, electric bells, signal horns and luminous signals on ships, electric candlesticks and arc lighting, the electric brakes of steam trains, and so forth.

In 1880, Planté's beloved father, with whom he had been living, died, and he never quite got over the loss. Possibly this was the reason why his research in the nine years that followed focused more on fundamental mysteries in nature, e.g., globular lightning, hailstorms, waterspouts, cyclones, polar auroras. One night, he studied the lightning during a storm over his property at Meudon: 'that which was launched from the heavens to the ground describing a curb and which remained visible for an appreciable moment, forming a rosary of brilliant seeds.' Awesome experiments followed to reproduce lightnings in miniature using high-voltage electricity.

Planté investigated the difference between static electricity and the 'dynamic' electricity from batteries. This involved the invention of a mechanical device with a rotating commutator that he called the 'Rheostatic Machine' (machine rhéostatique), which he published in the *Comptes Rendus de l'Académie des Sciences* on the 29th October 1877. By rapidly rotating the shaft, a series of high voltage sparks many centimetres long could be generated. Using this device, Planté explored Lichtenberg figures (branching patterns created by high voltage discharges along the surface of an insulator covered with dust), the electrical breakdown of air, and the behavior of thin wires when subjected to pulses of high electric current. He charged a bank of mica capacitors in parallel from a battery source and then connected the capacitors in series to create a high-voltage power source for his rheostatic spark generator. At the time, he coupled multiples of his accumulators to his rheostatic machine. In the 1870s, Planté realized discharges in series at voltages of up to 200 000 V. The results were discussed in his book *Phénomènes Électriques de l'Atmosphère* in 1888.

1.4. Late industrial success

Gaston Planté was for some years head of the electroplating laboratory of Christofle, a firm that was established in 1830 and had become a renown manufacturer of sterling, silverplate and stainless-steel flatware. At that time, electrochemical methods were also used for the reproduction of figures and sculptures. Planté invented a new, less-expensive process for, among others, the decorative group at the front of the Paris Opéra.

The lead-acid battery came to the world 10 years too early because, at first, it had to be charged with Bunsen and Daniell cells. At the Breguet Company in 1873, Planté met the Belgian engineer Zénobe Théophile Gramme (1826-1901) who built direct-current generators (1869-71) that were based on Pacinotti's ring armature (1860). Planté recognized that his own lead-acid technology could play an important role in advancing the use of electricity as an energy vector. Shortly before the Gramme machine was officially demonstrated by Hippolyte Fontaine at the Vienna Exhibition in 1873, in connection with the transport of electric power, Planté had already investigated the reversibility of this electric generator, which was able to work as an electric engine in conjunction with a lead-acid battery. At the Vienna Exposition, Planté exhibited a combination of a steam engine and a pump at a distance of 2 km, the two electrical machines acting as generator and receiver. Bréguet, the manufacturer of the Gramme machine, then produced the first commercial Planté's storage batteries.

Planté never visited the United States of America, but he exhibited a battery in New York and found only a small loss of capacity on its return. His apparatus appeared at Munich and Vienna and added considerably to the renown of their creator.

Reseachers seized the lead-acid system. The French engineer Camille Alphonse Faure (1880-81) succeeded in achieving high capacities from electrode coatings of sulfur-containing lead powder pastes after a few cycles of 'formation'. Faure covered both sides of smooth lead plates with a thick layer of 'red lead' (Pb₃O₄). At the first charging, lasting about two days, the red lead was changed to PbO₂ at the positive electrode, whereas that on the negative electrode was reduced to spongy lead. Faure's battery was manufactured by S. A. La Force et la Lumière. The metallurgist J. Scudamore Sellon, in 1881, applied a 'spongy lead' paste to a perforated thin metal plate to improve adhesion. He had already replaced the soft lead in the grids with lead-antimony alloys that later became the dominant technology. It is known that Ernest Volckmar, J.W. Swan and others developed a lead grid at the same time [5]. Also in 1881, a lead electrode with a large surface area caused by a ribbed structure was registered as a patent by Charles F. Brush, while S.C. Currie invented the basic form of tubular plate ('ironclad plate'). The Brush plate was nothing less than the predecessor of the later 'Planté plates', which are still in use today. Henri Tudor introduced cast Planté lead plates with a large surface of thin vertical ribs, intersected at intervals by horizontal ribs to give the plates strength. The negative plate was a shallow box of two grids riveted together, and the small holes pierced in the smooth sheets at the outer surfaces were pasted. The Hart cell combined Planté and Faure plates hanging by side lugs on glass slats and separated by glass tubes. The American Gould cell had homogeneous plates of the Planté type, which were formed from sheet lead blanks by gradually raising the surface into a series of ribs and very fine grooves.

Planté and Faure batteries quickly proved to be useful for electric traction on road, rail and water, and even a small balloon was propelled at the speed of 4 m s^{-1} . In May 1880, Gustave Trouvé, patented a small electric motor and described its possible applications (French Patent No. 136,560). In 1881, he powered a three-wheeled electric vehicle by a lead-acid rechargeable battery:

'A motor weighing 5 kg, powered by 6 cells of Planté producing an effective work of 7 kg m per second, was placed, on the 8th April [1881], on a tricycle whose weight, including the rider and the batteries rose to 160 kg and recorded a speed of 12 km/h.'

This was more almost 60 years after the first electric vehicles were built around 1832. Then Camille Jénatzy (Belgium, 1899) set the land speed record of 109 km h^{-1} in his cigar-shaped electric car, powered by two 80-cell 'Fulmen' lead-acid batteries. Claude Goubet (France, 1885), Isaac Peral (Spain, 1886) and Gustave Zédé (France, 1888) launched the first electric-powered submarines. The alkaline cells in Zédé's boat were replaced in 1891 by 'Laurent-Cely' lead–acid cells. The *Plongeur*, the first submarine in the world to be propelled by compressed air (rather than hand-cranking), had already been launched in France on 16 April 1863.

Eventually, it was Morse's electric telegraph and the later telephone companies in the USA that forced the deployment of batteries on an industrial scale. By the end of 19th century, Siemens' dynamo (1867) and Swan's (England, 1878) and Edison's (USA, 1879) versions of incandescent lamp ensured a fast growing need for the storage of electrical energy.

Already in 1824, the Imperial Continental Gas Association in London had been established to set up street lighting with gas lamps in different European cities. The first *electric* street lighting of the Grands Magasins du Louvre in Paris, the 'City of Lights', employed eighty carbon arc lamps. The 'Yablochkov candle' (1875) was powered with alternating current, which ensured that both electrodes were consumed at equal rates. Joseph Swan's incandescent lamp lit Mosley Street, in Newcastle-upon-Tyne in 1879. The first permanent electric street lighting in Germany was commissioned by Sigmund Schuckert in Nuremberg in the summer of 1882. Planté supplied the Imperial Palace of Franz Josef in Vienna with stationary and portable equipment for lighting in 1883. The city of Paris installed combined batteries and dynamos for the distribution of electricity for lighting in 1882.

1.5. Planté's legacy

Without ever ceasing to study his accumulator, Planté directed his research towards the production of high voltages from 1875. His book of 1879 described the accumulator of 1859, but it was too late to claim any patent protection at that time. The term 'accumulator' means a rechargeable battery which works 'by the action of a reverse current' [11] and accumulates the chemical work of an electric current. Outside of Europe, synonyms have been preferred since the 19th century such as secondary battery, storage battery, or reversible battery.

The industrial applications of his battery and his rheostatic machine were permanent a source of income for Planté. He never took financial advantage from his discoveries, and retained from his income not more than was necessary for a modest standard of living. Unconcernedly, Gaston let other people make a fortune from his inventions: 'I am delighted whenever one would like to make use of my ideas, because this shows me that one does not always think them bad ones.' And he never accepted an appointment to a public institution. When some members of the Académie des Sciences suggested his election, he replied that he would lose much time in soliciting their votes, and that he preferred to be at work in his laboratory. In this respect, he resembled physically and morally his beloved brother Francis, the pianist, so much so that Francis's wife allegedly could not distinguish between the two when they were standing on the other side of the street.

Planté received honours from several scientific societies. The Lacaze Prize in 1881, an appreciable sum of 10000 francs, he donated to the Société de Secours des Amis des Sciences (Humanitarian Society of the Friends of Science) for the benefit of needy researchers. In the same year, Planté was awarded France's highest honour: the Chevalier de la Légion d'honneur. His sponsor, Monsieur Hervé-Mangon wrote to the French Finance Minister: 'I do not believe I am exaggerating by confirming that M. Planté is one of the greatest inventors of our time.' The Golden Ampère Medal was awarded to Planté in 1882 by the Society for the Encouragement of National Industry. At the ceremony, Jean-Baptiste Dumas said: 'I am happy to give you this medal bearing the effigy of Ampère Table 1

Key events in the life of Gaston Planté and his posthumous acknowledgment.

1834	Born 22 April in Orthez, France
1841	Movement of the Planté family to Paris. Boarding school of Abbé Poiloup at Vaugirard; thence as a day student to the Lycée Charlemagne
1850	Bachelor of Letters at the Lycée Charlemagne
1853	Bachelor of Sciences at the Lycée Charlemagne
1854	Laboratory assistant in physics at the Conservatoire des Arts et Métiers (Conservatory of Arts and Crafts) in Paris under Edmond Becquerel
1855	Master of Science at the Sorbonne University. Discovery of fossils of the prehistoric flightless bird Gastornis parisiensis or 'Gaston's bird' near Paris.
1859	Invention of the lead-acid 'secondary battery of great power' in his private laboratories: a spiral roll of two sheets of pure lead separated by a
	linen cloth, immersed in a glass jar of sulfuric acid solution
1860	Professor of Physics at the Association Polytechnique (Polytechnic Association) for the Development of Popular Instruction. Presentation of nine
	lead-acid cells connected in parallel to the Académie des Sciences
	Series of studies concerning electrolytic deposits, undertaken in the laboratories of the Maison Christofle
1862	Deputy for the Inspector General in the French section at the London Universal Exhibition
1864	Jury Member at the International Exhibition in Bayonne
1867	Member of the Admission Committee and of the Reunion of Offices of the 10th Group at the Paris Universal Exhibition
1879	Publication of Recherches sur l'Electricité (Research on Electricity)
1881	Lacaze prize, Ampère medal, Chevalier de la Légion d'honneur
	C.A. Faure develops further the lead-acid battery using a paste of lead oxide for the positive plate instead of a solid lead sheet
	C.F. Brush files US patents on a lead-acid secondary battery with electrically deposited spongy lead and oxidising the coating by electrical action
1888	Publication of <i>Phénomènes électriques de l'Atmosphère</i> (Electrical phenomena of the atmosphere)
1889	Died 21 May in Bellevue near Paris
1989	Centenary of his death. The Bulgarian Academy of Sciences establishes the Gaston Planté Medal, which is awarded to scientists for significant
	contributions to the development of lead-acid battery science and technology. The first recipient was Ernst Voss, Varta Batterie AG, Germany

and am sure that in the future our successors will give it with Your effigy.' Planté, however, considered the medal to be unproductive capital, sold it, and gave the proceeds to the poor.

Planté fell ill in 1885. Nervous diseases and pains in the eyes obliged him repeatedly to interrupt his work. In April 1889, he suffered a weakening of his sight when he was working on terrestrial magnetism and brilliant electric discharges. For fear of going blind, he taught himself the Braille alphabet. On 21st May 1889, Planté died from a stroke in his garden at Bellevue sous Meudon, at the early age of 55.

He bequeathed most of his fortune to scientific institutions and to destitute researchers. The Société de Secours des Amis des Sciences received his three properties in Bellevue. His house in Bellevue was converted into a retirement home for impoverished scientists. The Académie des Sciences received an endowment to establish a biannual monetary prize in the field of electricity. One of the first Planté prizes was awarded to Pierre Curie.

The following discourse outlines Planté's basic concepts of the lead–acid battery in an historical context (Table 1).

2. The dawn of the lead-acid accumulator

Planté is not considered to be the inventor of the rechargeable battery, although the lead–acid system was the first practicable secondary battery. In 1802–3, the German physicist Johann Wilhelm Ritter (1776–1859) found that a pile of layered discs of copper and cardboard soaked in a brine of table salt was able to generate a transitory 'secondary' current, after it had been charged electrically. This basic concept of the 'accumulator' fell into oblivion, however, as the only known charging equipment at that time, the Voltaic pile which had appeared in the summer of 1800 [5], was an impractical provider of electrical current.

Nearby six decades later, in 1854, the German physician and physicist Wilhelm Josef Sinsteden (1803–1891) constructed the first promising rechargeable cell by using two large lead plates in a vessel of diluted sulfuric acid [6]. On repeated charging and discharging, the cell eventually reached a measurable capacity.

In 1859, Gaston Planté invented his 'pile secondaire' which was composed of a pair of spiral-wound lead plates, separated by rubber strips, in a vessel of dilute sulfuric acid. This arrangement allowed the generation of much higher currents. The plates were charged for about 24 h by means of two Bunsen cells or three Daniell cells. Oxygen was evolved at the positive plate and combined with lead to form a brown coating of 'lead superoxyde' [sic] (PbO₂), whereas hydrogen at the negative electrode left the pure lead plate unchanged. Early vessels for the lead plates were also made from lead. Successive oxidations in the 'forming' process, gave rise to a spongy 'lead-peroxyde–sulphuric acid' cell that had a high electromotive force (an archaic expression of voltage), a low resistance, a large capacity, and almost no 'polarization'. All the active materials and the formed lead sulfate, fixed in the place where it is formed, were insoluble in the dilute acid–an invaluable practical attribute. Unfortunately, the formation of the spongy PbO₂ masses required much time, and the separating strips of caoutchouc rubber were found to have a short life. Again, the secondary battery was neglected.

Today, the half-cell reactions of the lead–acid battery are written in nearly every chemistry schoolbook. During discharge in sulfuric acid, lead(IV)-oxide is cathodically reduced to lead(II)-sulfate at the positive plate and lead is anodically oxidised to lead(II)-sulfate at the negative plate, as follows.

At positive plate:

$$PbO_2 + 3H^+ + HSO_4^- + 2e^- \rightarrow PbSO_4 + 2H_2O$$
(1)

At negative plate:

$$Pb + HSO_4^- \rightarrow PbSO_4 + H^+ + 2e^-$$
(2)

Cell reaction:

$$PbO_{2} + Pb + 2H_{2}SO_{4} \underset{Charge}{\overset{Discharge}{\rightleftharpoons}} 2PbSO_{4} + 2H_{2}O$$
(3)

Each cell in a lead-acid battery provides about 2 V. At full discharge, both the positive plate and the negative plate are converted to lead sulfate, and the electrolyte solution suffers dilution by the product water. When charged, the reverse reactions occur, and overcharge will lead to the electrolysis of water and consequent production of oxygen at the positive plate and hydrogen at the negative plate.

These aspects were important for Planté's work, who investigated the first serious analysis of 'galvanic polarization' in an oxidising medium. The obsolete expression polarization had been unwittingly introduced earlier to account for overpotential and overvoltage phenomena exhibited by electrodes and electrochemical cells, respectively. Modern electrochemists have since investigated the kinetic inhibitions of the electrode processes. Brockhaus' Encyclopedia of 1894 defined the term electric polarization as:

'During electrolysis, [...] oxygen is accumulating at a positive platinum plate of the voltameter, and [...] hydrogen at the negative plate. If the decomposing Voltaic battery is removed from the electric circuit, and the leads of the voltameter are connected with each another, then an electric current flows though the leads in the opposite direction of that current that originated from the battery through the platinum plates. This results in that those gases generate an electromotive force. [...] Such a reason causing a counter current is called electric polarization, galvanic polarization or Voltaic polarization. In gas batteries generated by polarization the original electric current performs a chemical work and then converts this chemical work back again in electrical current. [...] This principle has be used for the construction of accumulators.' [7] (Translated from the German by the author)

When sulfuric acid was electrolysed for a moment with the aid of platinum electrodes, it was found earlier by William R. Grove (1839) that the electrodes could themselves produce a current when detached from the primary battery. Planté observed at lead electrodes the 'secondary current which arises in a galvanic cell on closing the voltameter, immediately the primary current is cut off', that is the 'development of opposing E.M.F.'. The electromotive force (E.M.F.) is used by Planté synonymously with the modern term voltage across a galvanic cell.

The voltameter or voltascope was an appliance similar to a coulometer, comprising two electrodes in an electrolyte solution, an external power source for charging the device, and a galvanometer or a tangent compass for current measurements during charging or discharging, respectively. Brockhaus' Encyclopedia of 1894 explains its purpose for the determination of currents:

'[The] Voltameter, an instrument causing electrolysis in order to measure the intensity of galvanic currents, for example an apparatus for the decomposition of water acidulated with sulfuric acid, provides a measure for the electric current in a certain time by the quantity of the evolved gaseous components of water, e. g. the liberated hydrogen and oxygen or the oxyhydrogen. [...] With the more precise metal voltameters, the amount of deposited [...] copper or [...] silver [...] on the negative electrode is measured.' [7] (Translated by the author)

Jacobi (1839) connected the unit of electric current with the current that generated 10 cm³ oxyhydrogen per minute at 760 mmHg ambient pressure. The international Ampere or 'silver ampere' in the international system of electrical and magnetic units was valid from 1893 to 1948, and was therefore unknown to Planté. It was defined as the unvarying current that would deposit 1.118 mg of silver per second from a solution of silver nitrate in water.

3. An overview of Planté's original work

Gaston Planté's book *Recherches sur l'Électricité* (1883) is still available in the Bibliothèque Nationale de France [2]. An English translation was published under the title *The Storage of Electrical Energy* [3]. This work includes the main results of the research that Planté presented to the Académie des Sciences between 1859 and 1879. The first part describes electrical equipment such as voltameters, i.e., the abovementioned instrument for measuring a current by means of the amount of metal deposited, or gas liberated, from an electrolyte solution in a given time due to the passage of cur-

Table 2

	(1000 11.1)
Contents of Plante's book. Recherches sur l'Electricité	(1883 edition)
Contents of Flaine S Dook. Recherches sur F Lieunche	(1005 cultion).

First part: The accumulation and transformation of the energy of the voltaic battery by means of secondary currents			
Chapter I	The study of secondary currents		
Chapter II	Storage of the energy of the voltaic battery by		
	means of secondary cells with lead plates		
Chapter III	Transformation of the energy of the Voltaic Battery		
*	by means of lead plate Secondary Batteries.		
Second part: Applications			
<i>Use in alternations</i>			
Uses m: gaivanocaustics and therapeutics in general; exploding mines;			
domestic services; signalling by means of lights			
Third part: Effects produced by electric currents of high tension			
Chapter I	Use of secondary batteries, luminous sheath []		
Chapter II	Engraving on glass by electricity []		
Fourth part			
Chapter I	Analogies with globular lightning []		
Chapter II	Comparisons with the phenomenon of hail		
Chapter III	Comparisons with waterspouts		
Chapter IV	Comparisons with polar aurora		
Chapter W	Comparisons with spiral pobulao		
Chapter V			
Chapter vi	Analogy with solar spots		
Fifth part: Rheostatic machine			

Sixth part: Analogies between electrical phenomena and effects produced by mechanical actions

rent. The fifth paragraph of the first chapter cites the previous work of Sinsteden (1854), who charged voltameters with lead, silver and nickel plates by the use of his magneto-electric apparatus, and obtained secondary currents, sufficiently intense to raise wires to a state of incandescence. The work of Faure and Brush in the years after 1880 is not mentioned. The second part discusses possible applications of the battery, the third and fourth parts relate to electric currents at high voltages, the fifth part addresses the transformation of dynamic electricity to static electricity via his so-called rheostatic machine, the sixth part is devoted to analogies between electrical phenomena and mechanical actions. The full list of contents is given in Table 2.

3.1. Secondary cell with coiled lead plates

In the 37th paragraph of the above book, Planté describes the properties of the lead–acid battery that he had presented in the March 1860 issue of *Comptes Rendus de l'Académie des Sciences* [8]. Illustrations of the different cell/battery constructions (taken from Planté's book) are presented in Fig. 3

'37. [...] It is thus that we were led to construct, in 1860, a secondary element of great power or quantity, by using an arrangement similar to that which Offershaus and Hare had employed for the voltaic cell proper, namely, by rolling two long, wide lead plates into a coil, separated one from the other by a thick cloth, and then immersing them in a glass jar full of water acidulated with a tenth part sulphuric acid' [3].

This arrangement is shown in Fig. 3(a) and (e). The sketch in Fig. 3(b):

'represents a secondary battery of nine cells, having a total surface of ten square metres, the earliest effects of which we exhibited at the Académie des Sciences, on the occasion of the meeting of March 26th, 1860. By passing through this apparatus the current from five small Bunsen cells, we obtained, after a few minutes' action, a very bright spark possessed of great heating power when the two terminal wires of the battery in which the cells were coupled up, either in three parallels of three cells each in series, or all in parallel as shown in [the] figure, were brought into contact for an instant'.



Fig. 3. Drawings from *Recherches sur l'Électricité*. (a) Couple secondaire à lames de plomb en spirale (Secondary cell with coiled lead plates). (b) Batterie secondaire de grande surface (Secondary battery of large surface). (c and d) Secondary cell with parallel lead plates (a, b, c, a', b', c'). (e) Lead plates separated by rubber strips.

Fig. 3(c) and (d) shows secondary cells with parallel lead plates.

3.2. Secondary cell with parallel lead plates

Around 1868 [9], Planté thought about reducing the resistance due to the thick linen cloth separator (toile grossière) between the lead plates, which, moreover, became 'rotten in the acidulated water' (in French: 'cette toile s'altérant à la longue dans l'eau acidulée') and could no longer prevent the lead plates from coming into contact with each other, and thereby short-circuiting the cell.

'39. [...] To overcome these objections, we used another arrangement consisting of two series of parallel lead plates, with the terminals of the even row of plates joined on one side, and those of the odd row joined together on the other side, put into communication with the two poles of a primary battery. These plates, brought very near to each other, and separated in the middle by insulating rods, were arranged vertically in a rectangular gutta percha cell, furnished with interior grooves, for holding the parallel lead plates. [...] The cell having been filled with water acidulated with a tenth part of sulphuric acid, a cover is fused to the upper edges. A very little hole only, is left, in order to allow an escape for gases arising from electrolysis, during the passage of the primary current. [...]

40. To illustrate the heating effects that can be produced with this apparatus, a thick platinum or iron wire is fixed between the terminals. [...]

41. By using six lead plates, 20 cm long by 22 high, and considering that the double surfaces of all the plates are used, with the exception of the two outside surfaces, we have thus a small

secondary battery for quantity, with a surface of about half a square metre, which will redden iron, steel, or platinum wires one millimetre in diameter, after being submitted for a short time to the action of a primary battery of two Bunsen cells' [3].

In 1872, Planté returned to the spiral-wound arrangement [10] shown in Figs. 3(a) and 4(a). His coil of lead plates was also presented in the journal *Les Mondes*. The lead plates were about 60 cm long, 20 cm wide, and 1 mm thick.

'43. [...] We separated these plates no longer with a thick cloth, but by narrow strips of India rubber, presenting the advantage of not being injured in the acidulated water and only covering a very small portion of the surface of the electrodes. Two pairs of India rubber strips about one centimetre wide by half a centimetre thick, are necessary to prevent the plates touching each other. The terminals are shaped at the opposite ends of the plates, in order to avoid as much as possible any contact, and to equalise the distribution of the primary current upon the surfaces of the electrodes, by separating the two points by which the positive and negative electricity flow into the secondary cell, as far as possible. But this is not indispensable if the plates are very uniformly rolled together. The chemical action of the primary current is then distributed equally over the whole surface of the secondary couple, even when the two poles are very close to each other. We then coil the lead plates, thus separated by two or three pairs of India rubber strips, round a wood or metal cylinder, [...]

45. [...] The glass jar containing the lead plates immersed in acidulated water, is covered with a vulcanite disc which carries the metal parts intended to close the secondary circuit, when the cell is charged. The terminals of the two lead plates are con-



Fig. 4. (a) The secondary cell of 1872 with rolled lead plates (60 cm × 20 cm × 1 mm) charged by two Bunsen cells. (b) Charging of Planté cell by Gramme machine. Drawings reproduced from *Recherches sur l'Électrictié*.

nected by means of the binding screws [...], with a primary battery of two small Bunsen elements and with the little copper plates' [3].

3.3. Chemical actions in lead-acid cells

In 1843, August-Arthur de la Rive had obtained a promising galvanic cell, made of powdered lead dioxide packed around platinum or carbon, that delivered a higher current than either Grove or a Bunsen battery. By means of a voltameter see Section 2), Planté investigated, in 1859, the way in which metals such as copper and silver are chemically oxidised and gradually dissolve in different aqueous solutions under the action of electric current. In the case of two lead wires in sulfuric acid, insoluble 'peroxyde' (PbO₂) was formed and coated the positive pole in a permanent manner. Each time, the primary power, delivered by two Bunsen cells was removed from the lead–acid cell, Planté was able to measure a 'secondary' current in the opposite direction by means of a galvanometer or a tangent compass for a short duration. Planté reached the following conclusions.

'17.[...] This adherence and insolubility of the peroxyde of lead, added to its affinity for hydrogen on account of the high degree of its oxydation, contribute to produce, in a voltameter with lead electrodes, a more intense secondary current and longer in duration than that of any of the other metals.

18. Lead covered with peroxyde of lead, in water acidulated by sulfuric acid, acts in fact in a manner exactly the reverse to that of zinc in the same liquid. It tends to decompose the water, by absorbing hydrogen, and to become the positive pole of a cell, if it is connected with lead not oxydised, whilst pure zinc tends to decompose the water, by absorbing oxygen, and becomes the negative pole of a cell in which it is opposed to another metal. [...] The lead plate placed at the negative pole does not undergo, by the action of the primary current, as marked a change as that of the positive pole; nevertheless, as lead is always more or less oxydised by exposure to the air, it is brought to a more perfect metallic state by the hydrogen which is manifestly the means of reducing the cell, and its tint changes from bluish grey to a much lighter grey. When we then close the secondary circuit, water being decomposed in the cell, simultaneously with the appearance of hydrogen upon the peroxydised plate, oxygen is carried to the plate rendered formerly metallic by the primary current, and oxydises it lightly. This oxydation is even visible; as the negative lead plate immediately darkens upon the closing of the secondary circuit. A single plate of lead, or one united with another plate exactly the same, would be thus oxydised in water acidulated by sulphuric acid, and it would not give any E.M.F. [...].

20. We have measured, several times, the E.M.F. of a voltameter with lead electrodes completely polarised by a sufficiently prolonged action of the primary current. We have found, by operating immediately after breaking the circuit of this current, that it was approximately equal to 1'5 [150%], a Bunsen element being taken as a unit' [3].

In the following paragraph, Planté clarified the role of sulfuric acid, and explained why Johann Wilhelm Ritter, who used salt water, had not noticed any marked effect with lead during his studies of secondary batteries.

'22. If we use salt water in the voltameter, instead of water acidulated by sulphuric acid, there is formed round the negative pole chloride of lead, scarcely soluble, and a very bad conductor, so that the primary current is rapidly diminished, and the secondary current itself is much weaker [...]. We have found that the E.M.F. of this current [...] was but '08 [8%] that of a Bunsen element' [2].

Planté explained the chemical actions in a lead–acid cell in Chapter II of the first part of his book. He emphasised that the proper pretreatment of the electrodes is the decisive prerequesite during the first charge of a lead–acid cell or battery.

'49. When a secondary cell of large surface [...] is new, that is, when the lead plates comprising have never served to transmit the current in a voltameter, and it happens to have the current from two Bunsen cells passed through it, oxygen gas appears almost immediately upon the positive plate; some of it at the same time oxydises the surface of the plate, and this becomes quickly covered with a very thin coating of peroxyde of lead. On the other hand, the hydrogen, after having reduced the slight layer of oxyde with which the lead was probably covered by exposure to the air, soon appears, and if, in a few moments, we try the secondary current given by the apparatus, we find it is already very strong by the sharpness of the spark produced when the secondary circuit is closed and opened again instantaneously, with a copper wire of low resistance. But the current thus obtained is of very short duration. [...] This happens from the layer of peroxyde of lead on the surface of the positive plate being very thin, and as it becomes quickly reduced immediately the secondary circuit is closed, it cannot furnish a sufficient quantity of electricity.

50. [...] We have then, after a first experiment, two plates, the surfaces of which present a molecular condition differing from

that in which they were when new. They are covered with thin layers of oxyde and reduced metal respectively [...].

51. [...] If we first consider the lead plate which was negative [...] the consequence is that if the primary current be passed through it again, the first portion of hydrogen will be devoted to reducing this layer of oxyde, instead of the thinner layer resulting only from exposure to the air, [...]. Then a period longer than the first will take place before the appearance of hydrogen upon the surface of this plate; for this gas will not begin to be set free till the oxyde is completely reduced to a state of pulverulent or highly divided lead upon the surface of this plate.

52. If we study what takes place upon the positive plate for the second time, [...] oxygen [...] is more easily absorbed, and we also begin to note a delay in the appearance of oxygen upon this plate, a period which corresponds to the time necessary for re-oxydising the layer of reduced lead upon its surface. [...] When these operations have been renewed a great number of times, the surfaces of lead in the secondary cell will be found in a more favorable condition for oxydation or reduction; the layers of oxyde alternately formed or reduced, will become thicker, and the resulting secondary effects will show a longer duration and greater intensity. This is in fact what is observed: the more a secondary cell is charged and discharged, the greater is the duration of the secondary current' [3].

3.4. The formation treatment

From the 53th paragraph of Planté's book [2] onwards, the electrochemical pretreatment to form the lead–acid secondary battery is outlined in detail. This most important step, which takes a long time, he termed 'formation' of the lead plates. The electrochemically active material of the positive plate is almost completely converted in time to lead dioxide, whereas that of the negative plate is reduced to a spongy form of lead, i.e., the reverse of reactions (1) and (2), respectively.

'53. [...] We have thus attained the extention in duration of the discharge of secondary cells, by charging them successively a great number of times, and discharging them in proportion, so as to develope upon their surface, and even produce to a certain depth in the thickness of the plates, these layers of oxyde and reduced metal, the finely divided state of which favors the development of the secondary current. This result has also been obtained in a still more marked manner, by successively changing, several times, the direction of the primary current acting upon the secondary couple. [...] Thus, the secondary cell must be first discharged, then recharged in the opposite direction. [...]

54. We have seen moreover that it was advantageous, [...], to allow a period of repose of several days between the reversals, in order to give to the deposits of oxyde and reduced metal, time to attain a crystalline nature, and strong adherence to the surface of the plates. [...] The peroxydised plate, and the plate covered with reduced lead, are both of them strewn with shining points, and become changed in their molecular construction, not only at the surface, but some distance within the pores of the metal; we even notice that the peroxydised plate in particular ends by showing considerable fragility.

55. The plates thus changed lose none of their weight, by any number of charges and discharges. [...] The lead is continually oxydised and reduced at the same time that the water is alternately decomposed and vice versa.

56. These combined operations that we have called by the name of *formation* of secondary couples, and which consist, as we have just seen, in *forming* or maturing them, in order to produce deposits of a certain thickness, allow of our obtaining heating effects of considerable duration in the discharge.

57. Prolonged immersion of the lead plates in the acidulated water, before the action of the primary current, hastens materially the formation of secondary cells. [...]

58. The intensity of the primary current also has a great influence over the more or less perfect formation of secondary cells. [...] The resulting oxyde from a sufficiently prolonged feeble current is black; that which a stronger current creates has a clear brown color characterising the true peroxyde of lead. [...]

59. [...] The lead plates should be penetrated little by little, as deeply as possible by the oxydising and reducing actions of the primary current, so as to completely change their molecular formation. [...] Thus a secondary cell the plates of which have been submitted for several hours at once to the action of the primary current, being left to itself for a month, without being discharged, then taken in hand again and charged in the reverse direction, will give a discharge of double the duration which it could give before'. [3]

Once the electrodes are sufficiently formed, one current direction is then adopted so that the secondary cell is always charged. Planté's recipe to prepare a lead-acid secondary cell properly reads as follows.

'59. [...] The secondary cell being filled [...] with water acidulated by a tenth part of pure sulphuric acid, we allow the current from two Bunsen elements to go through it six or eight times, in different directions alternately, on the first day. The secondary cell is discharged between each change of direction [...] by the incandesence of a platinum wire [...]. Gradually we increase the time during which the secondary cell is submitted to the action of the primary current in the same direction. This period is successively extended, after the first day, from a quarter of an hour, to half an hour and an hour. We leave it finally charged in one direction until the next day. The following day it is charged for two hours in the reverse direction, then in the first direction, and so on. We still notice an increase in the time of the discharge, but a point soon arrives beyond, which this duration does not perceptibly increase, [...]. We then leave the secondary cell at rest for eight days, and it is then recharged in the opposite direction for several hours, without making for that day any fresh reversal. Gradually the period of rest is extended to a fortnight, a month, two months, [and so on], and the duration of the discharge continues to increase' [3].

3.5. Maintenance and gas evolution

Obviously, Planté had no knowledge of phenomena such as overpotential and the kinetics of electrode processes. Hence, the reports of his observations of gas evolution during charging the lead-acid system tend to be both lengthy and convoluted, as demonstrated in the following extract.

'59 [...] When secondary cells [...] are charged, we observe that the gases are completely absorbed, for a certain time, to such a point, that, with a secondary couple having a surface of one square metre, submitted to the action of two Bunsen elements, it takes twenty to thirty minutes before any gas appears upon the surface of the plates. [...] When the gases begin to be liberated, it is a sign that the battery is no longer doing useful work towards producing the secondary current. [...], and there is no further advantage gained by prolonging the action of the primary current, [...] [The] liberation of gas may indicate that it is fully charged; [...]

66. [...] We have noticed, when using secondary cells with lead plates, that they often gave off a strong odour of ozone, especially when they were charged in the reverse direction by means of a rather strong current. [...] we found that ozone could be as easily produced with lead electrodes as with platinum, and even in a stronger degree' [3].

Here are Planté's recommendations for the maintenance of his secondary cells.

'61. [...] intervals of rest of several months [...] would tend to increase the resistance of the cells, and to render their charge longer and more difficult. It is consequently preferable to charge them from time to time, or to keep them constantly charged by a weak current' [3].

3.6. Preservation of charge and efficiency

Planté's remarks on the preservation of the charge in a secondary cell consider the activation and passivation of chemical reactions and different types of charging equipment.

'76. [...] It may be explained by taking into consideration that, if the deposit of peroxyde of lead has a certain thickness, the surface in immediate contact with the solution is alone reduced to a state of protoxyde, and then protects the underlying layers from any action.

77. [...] The deposits are sometimes loosened, and fall in flakes to the bottom of the liquid. We then conclude that, when the secondary circuit is closed, the electrical actions called into play cause chemical reactions, such as reduction or oxydation, beneath the non-conducting coatings which act as a protection when an electric circuit is not made.

78. [...] Secondary couples, when discharged, are able to give at the end of a certain time, without having been again charged, a residual charge, similar to that obtained from Leyden jars. [...] This phenomenon arises from the fact of the layer of peroxyde of lead upon the positive plate not being reduced throughout its thickness by the first discharge of the secondary cell. [...]

83. [...] We have found that the resistance of secondary cells of the various dimensions which we have used, varied from 2 to 5 metres of a copper wire 1 millimetre in diameter. We have noticed that the extent of surface, or the size of the secondary couple, which has a great effect upon the duration of the discharge, has far less influence over the resistance of the cells, than the distance the plates are apart, their more or less perfect degree of formation, and their condition. [...]

84. [...] to charge lead plate secondary cells, it is only necessary to employ a primary current of a higher E.M.F. than one and a half times that of a Bunsen element. [...] Three elements would, no doubt, charge the secondary cell more quickly; but this excess of E.M.F. is not necessary and may even prove inconvenient, if the cells have been formed with a weaker current, in loosening the coatings of oxyde and reduced metal upon the plates by a too rapid liberation of gas.

86. [...] Any apparatus giving a continuous current of electricity in the same direction may be used to charge secondary cells, provided it has sufficient E.M.F. [...]. 87. [...] A secondary cell may be equally charged [...] with the help of the Gramme machine, which gives, as we know, a continuous current in the same direction; and being reversible, like magneto-electric machines in general, can serve as an electromagnetic motor.

90. One of the principal advantages presented by these secondary cells is to afford storage for spare electrical work, or [...] a powerful energy that may be used at will, in a longer or shorter time. [...]

91. [...] By comparing, according to the deposits of copper obtained, the total chemical work returned by the secondary cell during its discharge, with the total chemical work expended in charging it, we found that the proportion of return was from 88 to 89 per cent.

92. The loss of work [...] may be accounted for as follows: First, the spontaneous reduction in the acidulated water of a small portion of peroxyde of lead, [...]. Secondly, the incomplete formation of the secondary cell: a portion of the gases being then driven off without producing any useful chemical effect [...]. Thirdly, the polarization or the development of opposing E.M.F. [...] a lead plate secondary cell, well formed affords a very perfect accumulator of the work of the voltaic battery.'

4. Concluding remarks

Gaston Planté declared that the lead-acid cell could retain its charge for a long time, and had the ability to 'd'emmagasiner ainsi le travail chimique de la pile voltaique', i.e., to store electrical energy. Based on the work of Johann Wilhelm Ritter and other researchers, he was the first to recognize the prerequisites for an effective lead-acid secondary battery, namely: (i) the insolubility and conductivity of the lead dioxide formed on the positive electrode, whereas hydrogen is liberated at the negative plate to leave metallic lead in a spongy state; (ii) changes in the chemical nature of the active mass during charge, discharge, and storage.

By repeated reversals of the current direction, Planté increased the active surface and capacity of each plate. Through this procedure, the duration of the successive charging currents was also increased from a few minutes to many hours, followed by a period of 'repose' before reversing—in order to generate: (i) more strongly adherent, more crystalline PbO₂ on the positive plate and (ii) a stable film of spongy lead on the negative plate. The process of 'forming' was continued until a fair proportion of the thickness of the plates was converted. After this, the cell was always charged in a given direction.

The three months required for forming and repose were reduced to 60 h by C.A. Faure (1881), who gave the plates a preliminary coating of red lead (Pb₃O₄). Gladstone and Tribe [11], among others, proved that 'sulfuric lead oxyde' (PbSO₄) and 'lead superoxide' (PbO₂) were formed by electrolytic decomposition of Pb₃O₄ on the positive plate and 'spongy lead' on the negative plate. Later inventors put the paste not on to the lead plates, but into the interstices of a grid.

The competition between the plates designed by Planté and Faure was rather lively till the first decade of the 20th century [12]. Pasted plates were found to give better performance in many applications. Nevertheless, Planté batteries continue to be made to the present day, albeit in much smaller numbers. Countless researchers and engineers have contributed to the success story of the lead–acid battery [13,14].

Gaston Planté the man [15], his life and his efforts to support others less fortunate than himself encourage the thought that it would have been a privilege to have met him.

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