Physiology. — *"On Spray-Electricity and Waterfall-Electricity"*. By Prof. H. ZWAARDEMAKER and Dr. F. HOGEWIND.

(Communicated in the meeting of October 25, 1919).

The generation of Spray-electricity and that of Waterfall-electricity are no doubt cognate processes; still they are by no means identical.

It may perhaps be useful, therefore, that we should here enlarge upon their congruency and their difference.

Spray electricity is generated when the air causes waterdrops to break up and diffuse; waterfall electricity is evolved when existing waterdrops strike against a boundary plane of air-liquid or air-solid substance. This induces electrification of the spray-nebula at the very spot where the cloud arises, whereas the electrical charge of the waterfall does not take its beginning before the water reaches the bottom. In either case small and large drops are formed with opposite charges. Both with spray-electricity and with waterfall-electricity the surrounding air is laden to a large distance with those diminutive droplets, driven off in all directions.

With spraying the large drops follow their primitive course till they strike on some impediment or other. These drops have become electrified long before they encounter this impediment. In the case of waterfall-electricity, however, large drops as well as small ones form at the very moment when the electric charge begins, i.e. the moment when the jet collides with the impediment.

In either case the conditions of

"pressure"
and "temperature"

largely reinforce the electrical effect. An overpressure of two atmospheres yields notably more electricity than one atmosphere. To obtain a considerable reinforcement of spray-electricity it is only necessary to store up the nebula in a space, whose temperature is 10° higher. Likewise waterfall-electricity will be considerably increased by heating the reservoir from which the waterfall proceeds. The presence of an electric field will augment either in a marked degree.

The distance at which the disc, doing duty for an impediment, is arranged, has an influence upon either.

In the case of spray-electricity there is an optimal and a critical distance. According to LENARD [1]) waterfall-electricity augments with the distance of the disc.

However, the liquid medium and the small chemical additions are of consequence. In most experiments water was the medium, but paraffinum liquidum will also generate spray-electricity, while waterfall-electricity can be obtained also with mercury. We have chiefly noted the influence of small additions of known chemical nature.

Spray-electricity is markedly raised by the addition of substances that lower the surface-tension and are moreover volatile (odorous matter, antipyretica, narcotica, alkaloids). It may thereby rise to an amount which waterfall-electricity cannot approach by far.

Perfectly pure water does not yield spray-electricity that is distinguishable with an ordinary electroscope. [2])

Traces of odorous matter are capable of rendering it excessive.

Spray-electricity, therefore is a means to detect the presence of small amounts of odorous matter otherwise than by the sense of smell.

Pure water of itself generates waterfall-electricity, as the water becomes positively electric at the moment of its collision.

Minute additions of odorous matter, gustatory substances, colloidal substances, modify the charge of the water considerably, now in a positive, now in a negative sense.

LENARD was the first who studied this problem in 1892, and immediately put forward an interpretation [3]), which he altered a little and extended in a subsequent publication. [4])

In our experiments we made use of LENARD's apparatus, with slight alterations.

A strong metal cylinder of ± 2½ liter capacity, with the lid attached to it hermetically by three screws, is provided at the bottom with a metal pipe with a tap. The pipe terminates in a glass tube with a fine outlet (1 mm. in diameter). In the lid there are two apertures, one of which is connected with the supply-pipe of the compressor with a gas-chamber of 2 $m.^3$ capacity, while the other which is closed with a screw-stopper, subserves the filling of the cylinder.

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1) P. LENARD: "Ueber die Elektricität der Wasserfälle". Wied. Ann. 46 p. 584. 1892

2) Unless in the presence of an electric field.

3) P. LENARD: "Ueber die Elektricität der Wasserfälle". Wied. Ann. 46—1892.

4) Id. id. "Ueber Wasserfallelektricität". Ann. der Physik. Bd. 47—1915.

Inside the supply-pipe of the gas-chamber an amber tube of ± $2^1/_2$ cm. is fitted for isolation, besides two taps: the one close to the cylinder, the other near the main-pipe leading to the gas-chamber.

The whole cylinder is suspended in a trivet from which it is insulated by amber pins.

The outlet is placed at about 1 m. over a large glass receiving-reservoir, in which a zinc plate rests on two wooden blocks. This tank is connected by a conducting-wire with the metal cylinder.

In its turn the reservoir is isolated from the environment by a paraffin-plate supported on four amber feet.

The whole apparatus is connected by a conducting-wire to an earthed electroscope.

Thus the cylinder, the receiving-reservoir and the electroscope are connected inter se by an electric circuit; they are at the same time insulated from the environment.

The pressure in the air-pump and the gas-chamber, registered by a manometer, is brought up to two atmospheres, the cylinder is filled with 1 Liter of the liquid to be examined, and the two taps in the supply-pipe are opened, so that the liquid in the cylinder is subjected to a pressure of 2 atmospheres. Now when the lower tap is turned on, the fluid flows under a high pressure out of the glass tube and strikes at an angle of 90° against the zinc plate below it. This produces positive or negative electricity according to the nature of the liquid and causes a deflection of the electroscope.

The deflection, registered by the electroscope after 1 minute's perfusion, is taken as the index for waterfall-electricity.

The entire apparatus being of a rather large capacity the electroscope takes some time before deflecting, which does not occur before the whole capacity is electrified. This takes more time with some liquids than with others.

For this reason the stopwatch is not put in operation before the electroscope begins to deflect and the liquid then continues flowing for a full minute after this.

During the experiment the room is well aired, because the air in the room is also charged and that in a sense opposite to the charge of the liquid. It is obvious that this would greatly interfere with the electrification of the liquid in the subsequent experiments.

Moreover, the receiving reservoir is covered with a close-mesh iron gauze, provided with a circular opening in the middle, through which the jet passes. This gauze serves to keep back the migrating droplets and possible foam, and to allow the extremely hazy nebula to spread in the surrounding air.

Our standard-liquid was pure tapwater, which yielded a mean deflection of 50 scale-marks. The Utrecht tapwater is very pure and contains few salts. In addition the temperature is pretty constant, which is of great importance, since temperature very much influences water-electricity.

A control-test with water was inserted between every set of two experiments, in order to ascertain the accuracy of the apparatus.

Furthermore the cylinder was washed out with tapwater after each experiment to remove small quanta of lingering electrifying substances, which might render the results of the following experiments less reliable.

In the case of water-electricity we found that odorous substances did not all act in the same way. Most of them reinforced the positive charge of the water, others hardly modified it or did not do so at all; a few again even weakened it so as to excite a negative charge. All this occurred seemingly without any special method. It is true, stronger concentrations (which are insignificant with the almost always slightly soluble odorous substances) generally give a greater increase or decrease. Besides, in the homologous series we found an augmentation of the deflection according as we passed from the lower to the higher terms.

For the present it seems utterly impossible to draw a hard-and-fast line, separating the reinforcing from the weakening odorous substances.

That waterfall-electricity is not identical with spray-electricity, may appear e.g. from the behaviour of indol, which markedly increases the charge in the former, but is almost inoperative with the second.

Conversely thymol e.g. gives a strong nebula-charge and hardly any waterfall-electricity.

Another instance is that of *fresh*-distilled water, which very distinctly intensified waterfall-electricity, whereas it remains inactive in spraying.

We subjoin a list of some odorous substances with their charges in the numerator of the fraction and the deflection of tapwater in the denominator, as we noted them down directly after the reading. The sign of the charge is also given:

Phenol $^{1}/_{1000}$ N. sol.: $+\ ^{100}/_{80}$
Cressol $^{1}/_{1000}$ N. sol.: $+\ ^{110}/_{80}$
Xylenol $^{1}/_{1000}$ N. sol.: $+\ ^{120}/_{80}$

Amylacetate $^{1}/_{1000}$ N. sol.:	$+ {}^{130}/_{35}$
Thymol sol. (sat.).	$+ {}^{40}/_{35}$
Toluol (dil. sol.)	$+ {}^{100}/_{50}$
Artificial Moschus (dil.)	$+ {}^{65}/_{50}$
1 % Amylalcohol	$+ {}^{100}/_{50}$
Indol. sol. (dil.)	$+ {}^{100}/_{50}$
Encalyptol (sat.)	$+ {}^{120}/_{50}$
Safrol (sat.)	$+ {}^{100}/_{50}$
Citrol (sat.)	$+ {}^{90}/_{50}$
1 % aethylalcohol.	$+ {}^{100}/_{50}$
5 % ,,	$+ {}^{90}/_{50}$
10 % ,,	$+ {}^{20}/_{50}$
20 % ,,	$- {}^{20}/_{50}$
Bornylacetate (sat.)	$+ {}^{40}/_{50}$
Camphor sol. (,,)	$- {}^{30}/_{50}$
Capronic acid sol.	$+ {}^{10}/_{50}$
Acetic acid $^{1}/_{8}$ % sol.	$+ {}^{30}/_{50}$
,, 1 % ,,	$- {}^{10}/_{50}$

From this it appears that most odorous substances increase the positive charge of the water. Only bornylacetate and camphor two substances closely allied as to smell, lower, resp. reverse, the charge.

The odorous substances that belong to the acids; lessen the charge of the water, just as all other acids do.

Finally very strong concentrations of odorous substances, such as we can procure with aethylalcohol, will lessen the charge or will even produce a negative charge, just as is the case with spray-electricity.

When passing on to *gustatory substances* we found, that on the one hand all sweet substances are more or less reinforcing. though the concentration must be stronger than in the case of odorous substances, and that, on the other hand, all salts and acids lessen water-electricity, while such bitter substances as belong to the electrolytes, also lessened it.

Bitter substances, however, that must be classed under the colloidal substances again raised the positive charge of water, just as all the other colloidal substances examined.

Sweet substances.

Saccharose	1 %:	$+ {}^{70}/_{35}$
Laevulose	1 %:	$+ {}^{100}/_{50}$
Glucose	$^{1}/_{4}$ %:	$+ {}^{85}/_{50}$
,,	1 %:	$+ {}^{100}/_{50}$
Sacch. lactis	1 %:	$+ {}^{120}/_{50}$
Glucocoll	$^{1}/_{4}$ %:	$+ {}^{120}/_{50}$
Glycerin	1 %:	$+ {}^{100}/_{50}$

Bitter substances.

Chinin pur (sat. sol.).		$- {}^{30}/_{50}$
Bisulj. chin.	1 %	$- {}^{30}/_{65}$
Chloret. magnes	1 %	$- {}^{15}/_{50}$
Sulfas natricus	1 %	$- {}^{15}/_{50}$
Chloret. plumb.	$^{1}/_{2}$ %	$- {}^{5}/_{50}$

Colloidal Bitter substances.

Fel Tauri	$^{1}/_{1000}$:	$+ {}^{100}/_{50}$
Extr. quassiae sicc.	$^{1}/_{800}$:	$+ {}^{100}/_{75}$
Glucochol. acid natr.	$^{1}/_{1000}$:	$+ {}^{60}/_{50}$.

Salts.

Sulfas natric.	1 %:	— $^{15}/_{50}$
Sulf. kalic.	1 %:	— $^{25}/_{50}$
Sulf. ammon.	1 %:	— $^{30}/_{50}$
Chloret magn.	1 %:	— $^{15}/_{50}$
„ natr.	1 %:	— $^{13}/_{50}$
„ kal.	1 %:	— $^{30}/_{50}$
„ ammon.	1 %:	— $^{11}/_{50}$
„ plumbic.	1 %:	— $^{60}/_{50}$
„ calcic.	1 %:	— $^{40}/_{50}$
Nitras kalic.	1 %:	— $^{60}/_{50}$

Acids.

Ac. lactic	$^{1}/_{8}$ %:	+ $^{20}/_{50}$
„ acet.	$^{1}/_{8}$ %:	+ $^{50}/_{50}$
„ „	1 %:	— $^{10}/_{50}$
„ hydrochl.	$^{1}/_{8}$ %:	— $^{30}/_{50}$
„ citric.	1 %:	— $^{20}/_{50}$
„ amygdal.	$^{1}/_{2}$ %:	— $^{40}/_{65}$
„ tartaric.	$^{1}/_{2}$ %:	— $^{70}/_{100}$

Bases.

Sol. NaOH	1 % =	— $^{30}/_{50}$
„ KOH	1 %:	— $^{40}/_{50}$
„ NH_4OH	$^{1}/_{4}$ %:	+ $^{25}/_{50}$

Here again we notice the lyotrope series, as with spray-electricity, at all events for the anions series:

Sulfas	Kalic.	$^{1}/_{2}$ N.:	—30
Phosph.	„	$^{1}/_{2}$ N.:	—30
Citras	„	$^{1}/_{2}$ N.:	—25
Chloret.	„	$^{1}/_{2}$ N.:	—22
Nitras	„	$^{1}/_{2}$ N.:	—15
Acetas	„	$^{1}/_{2}$ N.:	—13
Rhodan.	„	$^{1}/_{2}$ N.:	—12
Iodet.	„	$^{1}/_{2}$ N.:	—10

In this list only acetate occupies too low a place, as compared with the lyotrope series in the case of spray-electricity. Of the other substances that proved to be active in spraying, we investigated some glucosides and saponins, antipyretica and alkaloids, which, if not examined as a salt, invariably heightened the charge.

Glucosides and Saponins.

Aesculin	$^{1}/_{800}$:	+ $^{75}/_{50}$
Saponin	$^{1}/_{600}$:	+ $^{70}/_{50}$
Digitalin	sol.:	+ $^{65}/_{50}$

Alkaloids.

Caffein (dil.)	+ $^{100}/_{50}$
Theophylin (dil.)	+ $^{60}/_{50}$
Hydrochl. morphini	+ $^{50}/_{20}$

Antipyretica.

Pyramidon	+ $^{90}/_{50}$
Antipyrin	+ $^{100}/_{50}$

Lastly, all colloidal substances examined, appeared to increase waterfall-electricity, even an albumin-solution, which always contains salts, provided the solution be dialysed.

Colloidal substances.

Gelatin $^1/_6$ % sol.:	+ $^{70}/_{50}$	Extr. quassica $^1/_{800}$:	+ $^{100}/_{75}$
Tragacanth (very dil.)·	+ $^{65}/_{50}$	Glucocholz. natr. $^1/_{1000}$	+ $^{80}/_{60}$
Traganth (dil. sol.)·	+ $^{70}/_{50}$	Albumin ov. sicc. $^1/_4$ %	+ $^{20}/_{30}$
Amyl. oryzae $^1/_8$ %:	+ $^{75}/_{50}$	,, ,, dialysed	+ $^{35}/_{30}$
Dextrin $^1/_5$ %:	+ $^{75}/_{60}$		
Gummi arabic $^1/_{600}$·	+ $^{40}/_{35}$		
Fel Tauri $^1/_{1000}$:	+ $^{100}/_{40}$		

It goes without saying that with stronger concentrations the deflection diminishes in consequence of an increased viscosity.

As said, the influence of the temperature on waterfall-electricity is great.

Tapwater of 8° : 40
,, ,, 35° 100.

Rise of temperature, therefore, increases the positive charge of water exceedingly.

The influence of addition of a saltsolution is also a fact that cannot be denied. Alcohol, which, without salt, increases the positive charge in water considerably, partly loses this faculty at first in the presence of common salt, and even loses it altogether with a concentrated salt-solution. Then the negative charge of the salt predominates. Salt added to the negatively electrifying camphor heightens this negative charge. Camphor and salt co-operate. It should seem then that in the case of waterfall-electricity a simple summation takes place of the effects of water, salt and the volatile addition [1]).

In endeavouring to account for these phenomena we might look upon spray-electricity as well as upon waterfall-electricity as a form of frictional electricity. In either case the friction, between the liquid and the air in the outflow of the spray, between liquid and zinc plate in the waterfall, would set free electrons that are scattered about in the surrounding air. But then the liquid were invariably to be charged positively, which turns out differently in a majority of cases, as shown by the experiments. We presume, therefore, that a more intricate process is at work, in which larger corpuscles arise as carriers of the electric charge. Such formations might perhaps arise from the so-called ions, an equal number [2]) of positively- and negatively-charged ions, round

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1) In the case of spray- electricity the process is a much more complicate one. (See E. L. BACKMAN, Researches Physiol. Lab. Utrecht (5) XIX p. 210.

2) H. ZWAARDEMAKER. "Le phénomène de la charge des brouillards de substances odorantes. Arch. Neerl. Physiol. de l'homme et des animaux" Tome I 1917 p. 347.

which the water vapour may condense into droplets and with which salt-droplets may combine afterwards.

LENARD [1]) believes that *in* the superficial layers of every di-electric liquid there is not only an electric double-layer, generated by the molecular forces of the liquid itself, the negative layer being situated on the outside, but also that these layers differ as to material.

These differences, which vary with the substances dissolved in the liquid (electrolytes, volatile substances, complex molecules) affect the thickness and the strength of the electric double-layer.

It appears then that LENARD reduces the problem of the origin of waterfall-electricity and of spray-electricity to his hypothesis regarding the specific condition of the surface of every di-electric liquid.

Strictly the origin of the electrification would then be, not an emission of electrons, but a discharge of extremely fine droplets, the so-called "carriers", which, varying with the surface condition of the liquid, are either very small and charged negatively, because they take their origin entirely from the outer negatively charged layer of the liquid, or they are somewhat larger and may be positively charged, since the majority of them arise from the interior positive layer of the liquid.

For further particulars we refer to LENARD's article itself.

Suffice it to state that most of our results with waterfall-electricity are sufficiently explained by this theory.

Not however the intensifying influence of rise of temperature (LENARD's private opinion, founded on theoretical considerations, was that a lessening influence was to be looked for).

Neither does this theory explain why camphor and bornylacetate diminish waterfall-electricity; no more is the question of the intensifying action of the sweet substances and the colloidal substances settled by it.

The results obtained before in the Utrecht Physiological Laboratory in experiments on spray-electricity [2]) are much less easy to explain with the aid of this theory.

First of all pure tapwater (Utrecht Water Company) and *fresh*-distilled water (old-distilled water *is* active) give no or an inappreciable charge in spraying.

Secondly the intensification of the charge in consequence of addition

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1) P. LENARD. Ueber Wasserfallelektricität. Ann. der Physik. Bd. 47—1915.

2) H. ZWAARDEMAKER. Het in overmaat geladen zijn van reukstofhoudende nevels. Verslagen K. A. v. Wetensch. Deel XIX N°. 1.

H. ZWAARDEMAKER. Specifieke reukkracht en odoroscopisch ladingsverschijnsel in homologe reeksen. Id. Deel XIX N°. 2.

of an active substance, is generally much greater with spraying than with the waterfall.

Thirdly, according to LENARD's theory, salt-solutions yield a distinct negative waterfall electricity; on the other hand they did not give a charge with spraying [1]).

Finally LENARD's theory proves the bigger positive carriers of a common salt solution to contain sodium; the smaller negative carriers on the contrary consist of pure water molecules. This he demonstrated by bringing the carriers between the plates of a condenser, in which process the bigger positive Na-containing carriers went over to the negatively charged condenser-plate where the presence of sodium could be proved. The smaller negatively charged carriers went over to the positive plate, where no sodium could be found.

A similar experiment with *sprayed* salt-solution [2]) shows that the big, positive carriers as well as the small negative ones contain sodium.

We conclude, therefore, that though there is a correlation between waterfall-electricity and spray-electricity, they are obviously not quite identical.

H. ZWAARDEMAKER. Reukstofmengsels en hun laadvermogen door nevelelectriciteit Id. Deel XXV 30 Sept. 1916.

H. ZWAARDEMAKER. Le sens de l'adsorption des Subst. Volatiles Acta Oto-laryagologica.

H. ZWAARDEMAKER en H. ZEEHUISEN. Over het teeken v. h. ladingverschijnsel en de bij dit verschijnsel waargenomen invloed op de lyotrope reeksen Verslagen K. A. v. Wetensch. deel XXVII 1918.

E. L. BACKMAN. De olfactologie der methylbenzolreeks. Id. Deel XXV 27 Jan. '17.

E. L. BACKMAN. Ueber die Verstäubungselektricität der Riechstoffe. Arch. f. d. ges. Phys. Bd. 168 S. 351.

C. HUYER. De olfactologie v. aniline en homologen. Diss. "Onderz." (5) Deel XVIII, p. 1, 89.

1) Afterwards we succeeded in demonstrating a slight negative charge also with spraying a 1 % common salt solution by lessening the capacity of the receiving disc. This charge, however, is not nearly so great as the one evoked in the waterfall and is far inferior to the spray-electricity generated by additions of volatile substances and of substances that activate the surface.

2) A. STEFANINI and G. GRADENIGO. Inhalazione di Nebbie Salina Secche. Lucca 1914, p. 22.