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SIR OLIVER JOSEPH LODGE, D.Sc., LL.D., F.R.S.
SIR JOSEPH JOHN THOMSON, O.M., M.A., Sc.D., LL.D., F.R.S.
JOHN JOLY, M.A., D.Sc., F.R.S., F.G.S.
RICHARD TAUNTON FRANCIS
AND
WILLIAM FRANCIS, F.L.S.

> "Nec aranearum sane textus ideo melior quia ex se fila gignunt, nec noster vilior quia ex alienis libamus ut apes." Just. Lips. *Polit.* lib. i. cap. 1. Not.

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But the character of the relation remains unchanged; hence

$$\left(\frac{n_s}{\mathrm{N}}\right)_1 = \left(\frac{n_s}{\mathrm{N}}\right)_2 :$$

this equation is satisfied if

$$\frac{\mathcal{E}}{\mathrm{T}'} = \frac{\mathcal{E}'}{\mathrm{T}}.$$

Before the compression the average energy $\overline{\mathrm{U}}$ was given by

$$\overline{\mathrm{U}} = \frac{\mathcal{E}}{e^{\frac{\mathcal{E}}{k\mathrm{T}}}-1}. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

After the compression we have

$$\overline{\mathrm{U}}' = \frac{\mathcal{E}}{e^{\frac{\mathcal{E}'}{k\mathrm{T}'}}-1} = \frac{\mathcal{E}'}{e^{\frac{\mathcal{E}}{k\mathrm{T}}}-1},$$

$$\frac{\overline{\mathrm{U}}}{\overline{\mathrm{U}}'} = \frac{\mathcal{E}}{\mathcal{E}'}; \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

but from equation (1) we obtain

$$\frac{\overline{\mathrm{U}}}{\overline{\mathrm{U}}'} = \frac{\nu}{\nu'},$$

Hence

$$\frac{\mathcal{E}}{\mathcal{E}'} = \frac{\nu}{\nu'}, \quad \text{or} \quad \mathcal{E} = h\nu, \quad . \quad . \quad (9)$$

which is the quantum relation of Planck. The average energy $\overline{\mathrm{U}}$ becomes now equal to

$$\overline{\mathrm{U}} = \frac{h\nu}{e^{\frac{h\nu}{k\mathrm{T}}}-1}. \quad . \quad . \quad . \quad . \quad . \quad . \quad (10)$$

Rayleigh and Jeans have shown that the number of oscillations do per unit volume in the frequency interval between ν and $\nu + d\nu$ is equal to

$$do = \frac{8\pi\nu^2\,d\nu}{c^3}. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (11)$$

Hence the energy $d\mathrm{E}$ in the same frequency interval is equal to

$$\left.\begin{aligned} d\mathrm{E} &= do\mathrm{U} = \frac{8\pi h\nu^3\,d\nu}{c^3\left(e^{\frac{h\nu}{k\mathrm{T}}}-1\right)}, \\ \nu &= \frac{c}{\lambda}; \end{aligned}\right\} \quad . \quad . \quad . \quad (12)$$

hence

$$\frac{d\mathrm{E}}{d\lambda} = \frac{8\pi hc}{\lambda^5\left(e^{\frac{hc}{k\lambda\mathrm{T}}}-1\right)}.$$

This is Planck's law of radiation.

In an oscillation taken at random, we find $f(\nu)$ quanta of energy $\mathcal{E}$; therefore

$$d\mathrm{E} = \frac{8\pi\nu^2\,d\nu}{c^3}\,h\nu f(\nu). \quad . \quad . \quad . \quad . \quad . \quad (13)$$

Comparing (13) with (12) we get

$$f(\nu) = \frac{\lambda}{e^{\frac{h\nu}{k\mathrm{T}}}-1}.$$

Laboratory of Physics,
University of Illinois,
October 10, 1922.

XXIX. *Electrolysis with a Dropping Mercury Cathode. Part I. Deposition of Alkali and Alkaline Earth Metals.* By Jaroslav Heyrovský, *D.Sc., Ph.D., Assistant Professor in Physical Chemistry, Charles' University, Prague**.

Introduction.

B. Kučera (Drud. *Ann.* vol. xi. p. 529, 1903) showed that the change of surface-tension of polarized mercury, *i.e.*, the "Electrocapillary parabola," can be very exactly determined by drop-weight. In this method, mercury dropping slowly from a glass capillary in an aqueous solution is polarized, the other electrode being the layer of mercury covering the bottom of the vessel.

In using the dropping mercury as cathode, it has been observed that in neutral or alkaline solutions hydrogen is not evolved even at high polarizations. This arrangement seemed thus convenient to study the cathodic deposition of the most "positive" metals, which are otherwise attacked by water. Besides the high over-voltage the dropping mercury electrode possesses other advantages: it yields in

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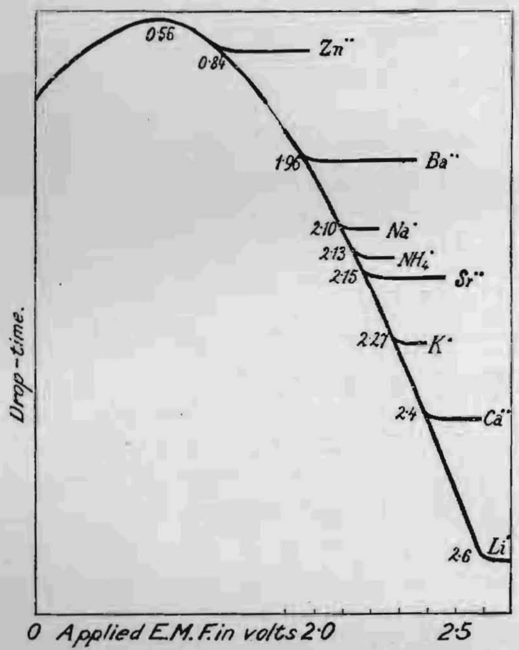
* Communicated by Prof. W. C. McC. Lewis.

the solution always a fresh and pure mercury surface, avoiding thus any "concentration polarization"; further the drops when falling on the bottom mercury used as anode also produce stirring there.

The bottom layer of resting mercury when in a solution of a halide or hydroxide keeps up the well-defined potential of the respective standard electrode, and thus can be relied upon as an exact reference electrode.

With such an arrangement even calcium and magnesium can be deposited in mercury before hydrogen and their amalgam formation can be followed in an exact manner.

Fig. 1.



The ease with which various metals are cathodically deposited in mercury can be determined by observing in each case the shape of the electrocapillary parabola, obtained by the drop-weight method, when readings are continued until considerable amalgam formation is reached. From this point any further increase of polarization does not effect any change in the drop-weight or drop-time. Consequently, when amalgam formation begins, the descending branch of the parabola turns horizontal.

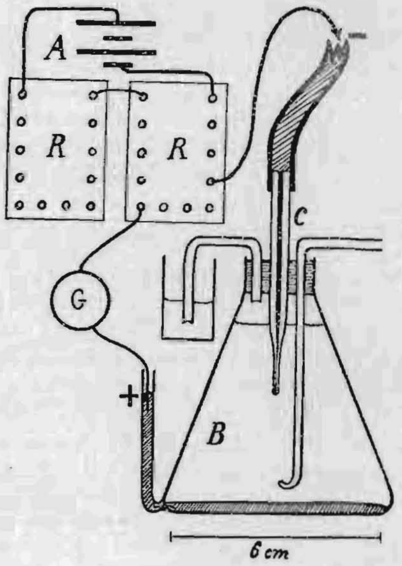
The graph in fig. 1, which refers to decinormal solutions of chlorides of the respective ions, gives us only approximate numbers; the results vary slightly, depending on factors which determine the current, *i.e.*, the size and velocity of drops, strength of the solution, and dimensions of the vessel.

A much more accurate method, however, is to determine in such cases the increase of current caused by the increase of the polarizating E.M.F.; these investigations are described in the following part *.

MEASUREMENT OF THE CURRENT.

The arrangement is shown in fig. 2.

Fig. 2.



The Apparatus. A double lead accumulator A was circuited through two resistance boxes, R. From one of these the current was branched to the electrolysing vessel, which consisted of a conical flask, B, with a platinum contact attached to the bottom; a thick-walled glass capillary passed through the cork stopper C, connected by a rubber tube to a

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* See also J. Heyrovský, *Chemické Listy*, vol. xvi. p. 256 (1922).

mercury reservoir (not shown in the sketch), by the level of which the rate of dropping could be regulated. The glass tubes, which passed through the stopper, served to lead the hydrogen in and out. The negative terminal of the resistance-box was connected to the mercury reservoir above the capillary, the other was connected through a sensitive galvanometer to the bottom mercury layer in the flask.

The capillaries were drawn out of a thick-walled thermometer capillary, with about 1 mm. inner diameter; they were broken off under water when connected with the mercury reservoir, so that the pure and dry mercury rushed into the tip first. It is essential to have the inside of the tube quite free from dust—however, the capillaries must not be washed nor moistened; for this reason capillaries made of fresh glass have to be used. The break of the tip was chosen so as to yield drops of about 4 to 6 seconds time (*i. e.*, 0·02–0·05 mm. inner diameter of the opening).

Since on prolonged use the capillaries get stopped and moistened inside, which might lead to hydrogen evolution in the tip, they have to be frequently exchanged for new ones.

The reflecting galvanometer of the Kelvin type with an astatic needle had 50 ohms resistance; one millimetre deflexion of scale, when two metres apart from the mirror, corresponded to 10^{-8} ampere.

The Method of Procedure. Before starting polarization, the resistances of the boxes were adjusted so as to be one ohm per millivolt of the E.M.F. of the source (about 4 volts); this E.M.F. was determined by compensation with a standard Weston cell with an accuracy to a tenth of a millivolt and was frequently checked. The vessel was then filled with a solution prepared from Kahlbaum's or Merck's purest specimens, and the stopper was sealed with paraffin-wax. In order to prevent oxidation by air at the cathode, it has been found necessary to pass hydrogen for 30 to 40 minutes through the solution.

The room temperature was kept as close as possible to 20° C.; the maximal deviation ±1° had no noticeable effect, which probably lies within the experimental errors of the method.

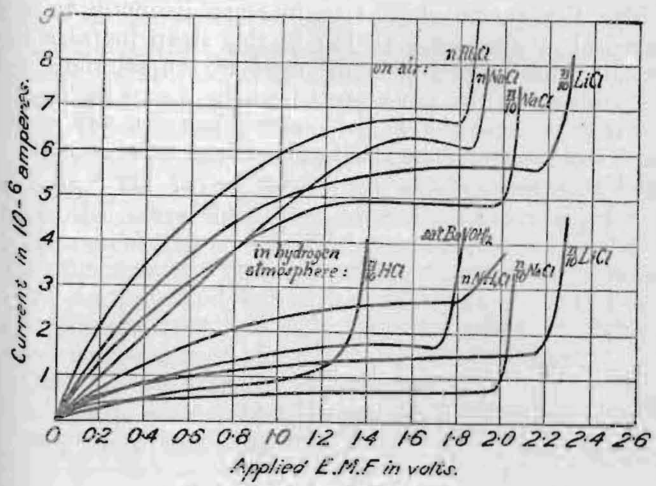
The zero reading of the galvanometer was then taken and the polarization was gradually increased, and for each value of the applied E.M.F. the corresponding galvanometer reading was noted.

OBSERVATIONS.

Some of the typical curves, showing the relation between E.M.F. of polarization and the resulting current, are drawn on the diagram (fig. 3).

When working with solutions in an open beaker in air a small current passes even at small polarizations, which is largely due to oxygen dissolved in solution. The readings in such cases were not steady, but oscillated within sharp limits (1–2 scale divisions) synchronously with the drop-time, this being no doubt caused by the different rates of oxidation of the rhythmically changeable drop-surface. By expelling air with hydrogen, the current was considerably diminished and oscillation stopped entirely.

Fig. 3.

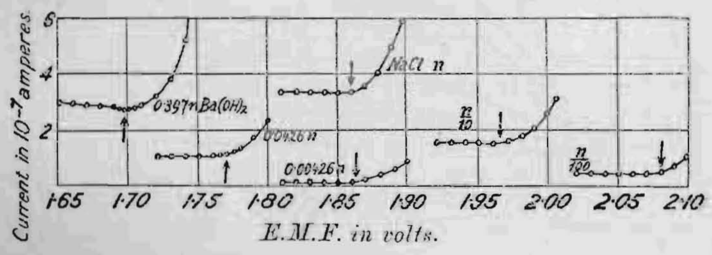


With increasing polarization the current increases at first very slowly until a certain value of the E.M.F. is applied, above which the current increases rapidly, so that a further increase of a few centivolts puts the mirror off the scale. This critical E.M.F. is quite independent of the dimensions of the electrodes of the drop-time and of the amount of air in solution—in fact, it is found to depend only on concentration of the cation.

The independence of the critical polarization, which might be termed the "decomposition E.M.F.," since at this point extensive electrolysis of the salt sets in, has been ascertained in a series of solutions using different capillaries, with different drop-times (from 1 to 5 secs.), with varying distances between the electrodes (0·5 to 6 cms.) and varying size of bottom layer (from 9 to 60 cm.^{2}), further. with varying external resistance in the boxes (from 0·1 to 10 ohms per millivolt). For very quick drops (less than one sec.) a slight shift towards greater polarizations can be observed; for drop-times greater than 1·5 sec. the results coincided. The slower the dropping, the more steady the galvanometer was; therefore, very narrow capillary openings were used, giving about 3 secs. drop-time at the decomposition polarization.

The decomposition E.M.F. was fixed as the value from which the current begins to increase distinctly in a geometrical progression. Owing to this steep increase it was possible to fix this point in most of the solutions within 2–4 millivolts, in some cases even to a greater accuracy. For this purpose the E.M.F. near this value was increased by 5 millivolts and the exact point found from a graph.

Fig. 4.



The more dilute solutions require larger decomposition E.M.F. (see fig. 4), the differences for a ten-fold dilution of a uni-univalent salt amounting to 110 millivolts, for a bi-univalent salt it is about 80 millivolts, which is the well-known difference of "free energies" at these dilutions (*cf.* McInnes and Parker, Journ. Amer. Chem. Soc. 37, p. 1445, 1915). Solutions prepared (by weight or dilution) at different times and from different sources always led to the same decomposition E.M.F.

Traces of noble metals or oxidizing matter in solution cause disturbances in readings; these become troublesome in very concentrated solutions, which therefore had to be prepared in a high degree of purity to give satisfactory results. For much help in this way the author is obliged to Professor B. Brauner, who presented some of his rare specimens and provided for exact standard solutions.

When a cathode of very small dimensions is polarized, the large reference electrode being used as anode, the decomposition E.M.F. must be equal to the difference between the potential of the electrode used as anode and the potential at which decomposition takes place at the cathode. Let us denote the latter as the "deposition potential" of the cation in mercury. To show that this deposition potential is independent of the anion, solutions of chlorides and hydroxides of the same metal in various dilutions were electrolysed, and from the results the deposition potentials referred to the normal calomel electrode were calculated. For this purpose the exact potential difference of the bottom reference electrode in the vessel from that of the normal calomel electrode had to be determined. Therefore, for most of the solutions a calomel or mercuric-oxide-electrode has been set up and compared with the standard calomel electrode. The potential of these single electrodes never differed by more than two millivolts from the polarized bottom mercury layer when both were in the same solution.

To eliminate the liquid junction potential between the normal electrode and the various solutions, a quarter-saturated, half-saturated, and fully saturated potassium chloride solution has been used and the values extrapolated.

The results are given in Tables I. and II.

The results show distinctly that the polarization at which electrolysis starts, E, depends only on the concentration of the cation being equal to the difference of potential of the resting mercury and the deposition potential of the cation $(\pi_0 - \pi')$.

The value π_N, which is the deposition potential of the cation from a solution containing one gram-ion per litre, has been obtained from π' actually observed at different dilutions by subtracting the free energy of dilution of the ions; here the average numbers observed in concentration cells at room-temperature were used, viz.:—

For ten-fold dilution of uni-valent cations 0·056 volt.

For 100-fold dilution 0·114 volt (known from silver- or hydrogen-ion concentration electrodes).

For bivalent 0·026 and 0·054 volt respectively (from $Zn^{\cdot\cdot}$, $Hg^{\cdot\cdot}$, $Ba^{\cdot\cdot}$ concentration cells).

Table I.

Deposition of the Alkali Metals, $t=20°$ C.

Solution.	E the observed decomposition E.M.F.	π_0 observed P.D. of the bottom layer from N. cal.	$\pi'=\pi_0-E$ the deposition potential of the cation.	π_N the calculated normal deposition potential.
n LiCl	2·05	*0·00	−2·05	−2·05
$\frac{n}{10}$,,	2·16	+0·052	−2·11	−2·05
$\frac{n}{20}$,,	2·22	+0·068	−2·15	−2·07
0·0382n ,,	2·26	+0·085	−2·17	−2·07
1·122n LiOH	1·867	−0·1552	−2·022	−2·025
0·108n ,,	1·960	−0·1138	−2·074	−2·021
n NaCl	1·860	+0·0010	−1·860	−1·860
$\frac{n}{10}$,,	1·968	+0·0521	−1·916	−1·860
$\frac{n}{100}$,,	2·080	+0·1072	−1·973	−1·859
1·076 NaOH	1·700	−0·1598	−1·860	−1·862
$\frac{n}{10}$,,	1·810	*−0·104	−1·914	−1·858
$\frac{n}{100}$,,	1·925	*−0·048	−1·973	−1·859
n KCl	1·883	0·000	−1·883	−1·883
$\frac{n}{10}$,,	1·990	0·0532	−1·937	−1·881
$\frac{n}{100}$,,	2·108	0·1080	−2·000	−1·886
n RbCl	1·795	*0·0	−1·795	−1·795
$\frac{n}{10}$,,	1·905	*0·053	−1·852	−1·796
$\frac{n}{100}$,,	2·020	*0·108	−1·912	−1·798
$\frac{n}{10}$ CsCl	1·948	*0·053	−1·895	−1·839
$\frac{n}{100}$,,	2·055	0·1077	−1·947	−1·833
n NH_4Cl	1·795	+0·004	−1·791	−1·787
$\frac{n}{10}$,,	1·900	+0·056	−1·844	−1·788
$\frac{n}{100}$,,	2·010	+0·110	−1·900	−1·786

* Values not actually observed, but taken from analogous cases.

Table II.

Deposition of the Alkaline-earth Metals, $t=20°$ C.

Solution.	E observed.	π_0 observed.	$\pi'=\pi_0-E$.	π_N calculated.
m $MgCl_2$	1·890	*−0·011	−1·901	−1·901
$\frac{m}{10}$,,	1·970	*+0·039	−1·931	−1·905
$\frac{m}{10}$ $CaCl_2$	2·090	*+0·039	−2·051	−2·025
$\frac{m}{100}$,,	2·170	*+0·095	−2·075	−2·021
0·0455n $Ca(OH)_2$	1·985	−0·0880	−2·073	−2·024
m $SrCl_2$	1·850	*−0·011	−1·861	−1·861
$\frac{m}{10}$,,	1·930	*+0·039	−1·891	−1·865
$\frac{m}{100}$,,	2·010	*+0·095	−1·915	−1·861
0·119n $Sr(OH)_2$	1·782	−0·1140	−1·896	−1·862
0·0120n ,,	1·857	−0·0627	−1·920	−1·860
m $BaCl_2$	1·800	−0·011	−1·811	−1·811
n ,,	1·828	+0·004	−1·824	−1·816
$\frac{n}{10}$,,	1·911	+0·0567	−1·854	−1·822
$\frac{n}{100}$,,	1·981	+0·1088	−1·872	−1·810
0·397n $Ba(OH)_2$	1·699	−0·1337	−1·833	−1·815
0·0426n ,,	1·770	−0·0875	−1·858	−1·813
0·00426n ,,	1·862	−0·0363	−1·898	−1·826

The series of the deposition potentials runs as follows:—

Li	−2·023 volts.	NH_4	−1·787 volts.
K	−1·883 ,,	Ca	−2·023 ,,
Na	−1·860 ,,	Mg	−1·903 ,,
Cs	−1·837 ,,	Sr	−1·862 ,,
Rb	−1·796 ,,	Ba	−1·814 ,,

Salts of lithium and calcium, owing to their high cathodic deposition, offered difficulties, since traces of other alkalies or alkaline earths present as impurities are deposited first. Moreover, at such great polarizations in quickly dropping capillaries, especially when long in use, bubbles of hydrogen are evolved. This was still worse with beryllium salt solutions, which, owing to hydrolysis, did not lead to any satisfactory result. Traces of acidity change the electrolysis of salts to that of hydrochloric acid (see in fig. 3) and then hydrogen is evolved before the metal is deposited.

On the other hand, traces of alkalies stabilise greatly the over-voltage and thus remove fluctuations in readings; the alkalinity, however, changes the potential of the resting mercury, making it more negative, and this change has to be measured and taken into account.

The most suitable for these measurements of polarization are alkaline solutions of hydroxides, not only on account of the scarcity of hydrogen ions, but also that the product formed at the anode is mercuric oxide, which is at the same time the depolarizer used in this electrode. Of all solutions, those of baryta are most favourable, as in this instance the traces of all other positive metals would be deposited at much more negative potentials, and impurities of nobler metals are excluded owing to the alkalinity of the solutions.

Electrolysis of dilute ammonium chloride offered no difficulties; it differs from the deposition of metals in that the current curve does not increase so steeply with increasing polarization (see fig. 3). The same was observed with magnesium salts and in concentrated solutions of calcium, strontium, and lithium.

In dilutions greater than hundredth normal the readings became indistinct, in very concentrated solutions impurities mattered much; in concentrated chlorides, moreover, the formation of complexes with mercury brings it in solution, which causes great increase and oscillations of the current. For a similar reason sulphates cannot be used, since mercurous sulphate is considerably soluble and the solution shows increasing conductance during polarization.

DISCUSSION OF THE RESULTS.

There is no doubt that the sudden change in slope of the current-E.M.F. curve (figs. 3 and 4) is due to amalgam formation at the cathode. The formation of amalgams from electrolysis of salts was studied by Coehn and Kettembeil (1901–4), who used aqueous solutions of calcium, strontium, and barium with a mercury cup as cathode. With such an arrangement, however, extensive hydrogen evolution, besides concentration polarization, excludes satisfactory results. Experiments with resting mercury used as cathode repeated by the present author gave the break at much lower and very changeable potentials, due only to the evolution of hydrogen, which depends on catalytically acting impurities present in solution.

According to the theory of electrolysis, even the slightest polarization, say 0·1 volt, deposits some cations on to the surface of the mercury drop, and forms there an extremely dilute amalgam. A very minute current equivalent to this deposition must pass through the solution, but, since the drop-surface possesses now a more negative potential, the current is stopped by the opposing E.M.F. In increasing the polarizing E.M.F. we increase the minute amalgam concentration in the drop surface and diffusion into the inside must result. The amount which diffuses will be proportional to the concentration of the amalgam in the surface, which is given by the formula

$$\pi = -\frac{RT}{nF}\log K_{Me} \, . \, C_{Me},$$

where C_{Me} denotes the amalgam concentration and π the potential of the polarized drop. Thus the current

$$i = k \, . \, C_{Me} = K \, . \, e^{-\frac{\pi n F}{RT}};$$

i. e. the curve should be an exponential.

It will be noted that the increase of current above the decomposition potential proceeds in a geometrical progression, the curve resembling a steep exponential.

The concentration of the amalgam in the drop-surface at the deposition potential can be estimated from the investigations of G. N. Lewis and his co-workers (1912–15) on the alkali-metal amalgam potentials. Assuming that tenfold dilution of the extremely dilute amalgam lowers its potential by 0·06 volt, we obtain from the available data a concentration of the order 10^{-6} gram atoms of alkali metals per litre of mercury, *i. e.*, about one millionth of one per cent.

Mercury Drop Potentials. An attempt was made to measure the potential of polarized mercury by placing under the dropping capillary in the electrolysing flask B (fig. 2) a glass cup with a platinum contact. Normal and tenth-normal sodium chloride solutions were used; the potential of the drops collected in the cup was measured against the bottom mercury, or, which was found always to be identical, against a normal or deci-normal calomel electrode connected with the solution. From the beginning of the polarization and also at the decomposition E.M.F. the drops showed no difference against the pure resting bottom mercury; however, 40 to 60 millivolts above the decomposition potential the drops in the cup showed already 2 millivolts difference from pure mercury, and this difference increased rapidly, so that at a polarization of 2·10 volts the mercury in the cup was already negative by nearly 2 volts more than the reference electrode. Obviously the most dilute amalgams which are formed at the deposition potential are only superficially polarized and lose their negative potential during the fall, being instantly acted upon by water.

Affinity for Mercury. It is assumed that the positive metals combine with mercury and their compounds are rich in mercury. Recently Richards and Conant (J. Amer. Chem. Soc. xliv. (1922) p. 601) pointed out that even at very great dilutions amalgams do not behave as ideal solutions, showing a rather large decrease in potential and a considerable heat of dilution. The explanation is that even at such dilutions combination with mercury still proceeds.

Supposing complete saturation will once be reached, in an enormously dilute amalgam, as is probably the case in the surface of slightly polarized mercury, we cannot expect that this enormously dilute amalgam will have a tendency to diffuse inside the drop as the particles are lighter than mercury and are completely "solvated" with mercury. As soon as the the amalgam concentration increases so far that it will not allow all atoms to be fully solvated with mercury, affinity for mercury as well as the increased concentration will cause diffusion into the interior of the drops; from this point the current curve will proceed much steeper than ordinary diffusion could effect, and this probably happens at the deposition potential.

The atoms of the alkali metals in the surface of mercury are under two opposing forces; the affinity for mercury draws them into the surface, their solution tension, on the other hand, sends them into solution. Thus the deposition potential depends both on the affinity of the metal for mercury and its electrolytic potential. Since at the deposition potential all alkali metals are in an analogous condition, we can use these values to express the relative affinities of the alkali metals for mercury. The deposition potential indicates thermodynamically the very small partial pressure the alkali metal has in the drop-surface; the combination with mercury has lowered the activity of the alkali metal, indicated by its electrolytic potential (E.P.) to the characteristic value π_N.

Thus the affinity, A, of the metal for mercury is equal to $\pi_N - \text{E.P.}$

From the work of G. N. Lewis, mentioned above, and the values π_N we have :—

Metal.	E.P.	π_N.	$A = \pi_N - \text{E.P.}$
Li.........	−3·304	−2·023	1·281
Na	−2·998	−1·860	1·138
K	−3·208	−1·883	1·325
Rb	−3·205	−1·796	1·409
Cs	(−3·3)	−1·837	(1·5)

The affinity is expressed in volt-faradays per gram-atom This series shows that the affinity for mercury increases with increasing atomic weight. Sodium has an anomalous position (*cf.* McPhail Smith, Zeitsch. Anorg. Chem. lviii. (1908) p. 381) and behaves like a nobler metal, its amalgam being very stable—in fact, hydrogen was never observed in these experiments to be formed on mercury drops on electrolysis of sodium solutions. Finally, it may be pointed out that the affinity of cæsium for mercury, which from extrapolation would amount to 1·5 volt, points to the E.P. of cæsium being −3·3 volt (from the calomel electrode).

Besides to Prof. Brauner, the author's thanks are also due to Professors J. Šterba-Böhm and F. Záviška for their help.

The author wishes to express his indebtedness to the prematurely deceased Professor B. Kučera, who suggested to him the study of the polarization of dropping mercury.

Chemical Laboratory of
the Charles' University,
Prague.

XXX. *Indian Drums.*

To the Editors of the Philosophical Magazine.

Gentlemen,—

IN India, like kettledrums, there are many drums which are musical, and which are used in company with stringed instruments. They differ from kettledrums in two respects: (1) they are loaded over a central zone, and (2) they elicit harmonic partials ('Nature,' Jan. 15, 1920, p. 500). It is well known that the partials of an unloaded membrane do not form an harmonic series, and a few of them give a consonant chord. From a look at the Indian instruments, it at once becomes evident that the load per unit area varies as