SCIENCE

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THE AMERICAN ASSOCIATION FOR THE ADVANCEMENT OF SCIENCE.

*THE ACHIEVEMENTS OF PHYSICAL CHEMISTRY.**

'PHYSICAL chemistry is the chemistry of the future.' These words, quoted from an address by Prof. Du Bois-Reymond, were used by Ostwald ten years ago in the introduction to the first number of the *Zeitschrift für physikalische Chemie.* In using these words Du Bois-Reymond looked forward to a time when it shall be possible to give a mathematical expression to all forms of chemical knowledge. The picture in his mind seems to have been that of a sort of astronomy of the atoms, in which the motions and forces within the molecules shall be known very much as are the motions and forces within the planetary system.

So far as any practical realization is concerned, the thought is still only a poetic fancy, and whatever progress, if any, may have been made, comes to us from organic rather than from physical chemistry. Indeed, it seems to have become the fashion on the part of several leaders in physical chemistry to speak slightingly of the atomic and molecular theories. Their thought appears to be that it would be better to confine ourselves to the purely empirical and mathematical concept of the atom and molecule and leave the idea of particles

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* Address by the Vice-President before Section C—Chemistry.

which have an actual independent existence entirely in abeyance. It would, doubtless, be possible to give such definitions of atom and molecule as shall include only the results of our actual empirical knowledge and mathematical deductions therefrom. Such definitions would stand on a basis which is incontrovertible, and it is inconceivable that even the overthrow of the atomic theory, or any knowledge which may be gained in the future as to the nature of matter, could change them.

That such definitions possess great value is certain; and that it is very important to distinguish sharply between our positive knowledge and speculations and theories based on that knowledge, every one will admit; but, after all, unless we join that school of philosophy which teaches that there is no real existence outside of our own minds, there is some reality at the basis of and behind all the phenomena which we investigate. And it is the province of science to find out the truth about every real existence of which we can secure any tangible evidence. Our knowledge of atoms and molecules as actually existing particles is, doubtless, a purely speculative deduction from a multitude of diverse phenomena, and yet the mental picture connected with the concept has been, and still continues to be, of very great value in the development of our science. These mental pictures are vague and in many respects incomplete, it is true, and they doubtless do not correspond closely to the real existences for which they are, at present, our best expression; but, to one familiar with the very practical results which have been obtained in the domain of structural chemistry, it is difficult to conceive how such results would have been possible without their use.

While physical chemistry has made little or no apparent progress toward the goal marked out by Du Bois-Reymond, its practical achievements during the last ten or fifteen years have been very considerable, and it is to these practical achievements that I wish to turn our attention.

Whenever the subject of physical chemistry is mentioned our thoughts naturally turn to the subject of solutions. Not that physical chemistry has to do only or chiefly with solutions, for, as I understand it, physical chemistry has to do with all of those physical properties of matter which can only be understood by taking into consideration at the same time its composition; but rather because some of the most valuable results of physico-chemical researches have been made in this field, and because diversity of views has made this the chief recent battle-ground in chemistry.

Up to about eleven years ago our knowledge of solutions was almost entirely of an empirical character. No great generalization, similar to those which had long been known for gases, had been discovered. In 1885 J. H. van't Hoff[1] proposed his theory of osmotic pressure. The empirical basis for this theory lay in the experiments of many different workers, some of them made many years before. Studies in osmose date from the early years of this century and, indeed, some experiments were made more than a century ago. It was not, however, till 1867 that the discovery of true semi-permeable membranes was made. In that year M. Traube[2] showed that membranes may be prepared which will readily allow the passage of water, but which are totally impervious to certain substances in solution. Ten years later W. Pfeffer[3] conceived the idea of preparing such membranes in the interior of a solid partition. By exposing a cup of porous porcelain to the action of a solution of copper sulphate on one side and of potassium ferrocyanide on the other, a precipitate is formed within the mass of the porcelain which is permeable to water, but which is impervious to the passage of sugar and of many other substances. With such cells the osmotic pressure was measured and was found to be surprisingly great. For a one and a-half per cent. solution of saltpetre it is more than three atmospheres. For sea water it would be about twenty atmospheres. Pfeffer's experiments were made with reference to their bearing on the action of organic cells and on other physiological questions, and it was eight years later before their extraordinary theoretical importance was pointed out by van't Hoff.

A careful study of the experimental data given by Pfeffer and others leads to the following conclusions:

First, the osmotic pressure is directly proportional to the concentration of the solution.

Second, the osmotic pressure is directly proportional to the absolute temperature. In establishing this law the experiments of Soret[4] are of especial interest. He subjected a solution of copper sulphate, contained in a vertical tube, to a temperature of 80° near the top and of 20° at the bottom. Under these circumstances the concentration increases below and diminishes above. After equilibrium was established it was found that the per cent. of copper sulphate in the two parts of the solution was inversely as the absolute temperature. The analogy with what would take place in a gas under the same conditions is clear.

Third, solutions which are isotonic at a given temperature contain in unit volume the same number of molecules of the dissolved substance. Another statement of the same law, which gives it also a quantitative expression, is that the osmotic pressure of a solution is the same as though the dissolved substance existed as a gas within the same space. The osmotic pressure of a one per cent. solution of sugar may be calculated by the same formula* which we should use to calculate the pressure exerted by one gram of a gaseous body having a molecular weight of 342 and contained in a volume of 100.6 cubic centimeters.

Every one recognizes, of course, that the laws which have been given for osmotic pressure are identical with the laws of Boyle, of Charles and of Avogadro for gases. Van't Hoff pointed out this analogy very clearly, but he did not give any clear explanation of what he considered as the real cause of the phenomena of osmose. He spoke, from the purely empirical side, of of the attraction which the solution exerts for pure water.[5] Ostwald in his Lehrbuch[6] is even more careful. He speaks of the cell as conducting itself as though there is within it a partial vacuum for water. These expressions are very similar to those of the older text-books, which speak of the expansion of gases as due to the repulsion of their particles for each other, and appear to me equally misleading and unsatisfactory. In a later paper,[7] in reply to a criticism by Lothar Meyer,[8] van't Hoff gives a clearer explanation in terms of the kinetic theory.

If we have a gas in a confined space and introduce into it a small amount of some volatile liquid the vapor of the liquid will rise and fill the space very nearly as though the gas were not present, and when equilibrium is reached the pressure will equal the original pressure of the gas plus the vapor pressure of the liquid. The explanation is that the pressure exerted on the surface of the liquid by the gas is not that of continuous matter, but is due to the

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* $P = \dfrac{760 \times T.}{342 \times 0.045 \times 0.1006 \times 273}$

In this formula,

T = Absolute temperature.

342 = Molecular weight of cane sugar.

0.045 = One-half the weight of a liter of hydrogen.

0.1006 = Volume in liters of 100 grams of the solution.

bombardment of its surface by particles of discontinuous matter. The particles of the liquid find ample opportunity, therefore, to rise between the particles of the gas.

Let us take a second case, which has, however, as far as I am aware, never been realized. Suppose a vessel having a wall impervious to the molecules of one gas but previous to those of a second. If such a vessel containing the first gas is placed in an atmosphere of the second the molecules of the latter will pass the walls and enter the space occupied by the first, exactly as the molecules of the volatile liquid rise among the molecules of the gas above, and equilibrium will be established only when the pressure exerted by the second gas is equal within and without. The pressure within the vessel will then exceed that on the outside by exactly the pressure exerted by the gas whose molecules cannot pass the wall.

The case with osmotic pressure is very similar to that last mentioned. Here we have a semi-permeable wall actually realized. For instance, we may have a wall which will allow water to pass freely but which is impervious to the molecules of sugar. If pure water be on one side of such a wall, and a solution of sugar on the other, equilibrium can exist only when the pressure due to the water alone is equal on both sides; for the molecules of sugar, because of their discontinuous character, can exert no influence to cause the molecules of water to pass one way or the other, exactly as a gas can exert no permanent effect to prevent the vapor of a liquid from passing upward into it. In the end, therefore, the pressure on the side of the solution must exceed that on the side of the pure solvent by the amount of pressure due to the kinetic energy of the molecules of the dissolved substance. If we further suppose that this energy is the same in the liquid as in the gaseous state, and the laws of osmotic pressure give us every reason to believe that it is, the explanation is complete.

This explanation gives us a conception of liquids as very closely related to gases in many of their properties, the main difference being that in the liquid the molecule does not possess enough kinetic energy to separate it from the mass of neighboring molecules, although its motion within the confined space is very similar to that of the molecule of a gas.

But it is not only, nor indeed mainly, in his study of the phenomena of osmose that van't Hoff has rendered the greatest service. Very few perfect semi-permeable walls are known, and osmotic pressures are very difficult to measure directly, so that, if we were dependent on direct measurements, the theory would be of scarcely more than theoretical interest. Van't Hoff pointed out, however, that the concentration of a solution by the removal of the solvent, whether effected by a piston composed of a semi-permeable wall, by the evaporation of the solvent, or by the separation of crystals of the pure solvent by freezing, is in each case a reversible process analogous to the compression of a gas, and that, as with all other reversible processes, it is subject to the second law of thermo-dynamics. This made it possible to connect the lowering of the vapor pressure and the depression of the freezing point of solutions directly with their osmotic pressure. This has given an indirect determination of the osmotic pressure in thousands of different cases. As a practical result we have now at our disposal a large number of methods for the determination of the molecular weights of solid and liquid bodies.

The work of Raoult[9] in this field deserves especial mention, because he developed several methods of determining molecular weights from an empirical standpoint, before the theoretical development of the subject had been given by van't Hoff. Raoult's[7] work attracted the attention of Victor Meyer, who made use of his methods in the study of certain stereomeric bodies upon which he was at work. And it is in connection with stereoisomerism that the new methods of determining molecular weights have, perhaps, been of the greatest practical value in the development of chemical science; for, without the positive proof that the bodies studied are metameric and not polymeric, the foundation for the belief that they are stereomeric would be comparatively weak.

It is probably through articles published by Victor Meyer[10] and Auwers[11] that cryoscopic methods for the determination of molecular weights were first brought to the attention of a wide circle of chemists. Since then a large number of workers have busied themselves with the subject, partly in the development of suitable forms of apparatus and methods of manipulation, partly in the study of the scope and degree of accuracy of the laws and of exceptions to them. The most important of the methods developed are those dependent on the lowering of the freezing point of solutions,[12] on the raising of the boiling point,[13] on the lowering of the vapor pressure,[14] on the determination of isotonic solutions by vegetable membranes[15] and by blood corpuscles, and on the lessening of the solubility of ether in water or of phenol in water by the addition of substances soluble in ether or phenol but not in water. In the last case the determination is either direct in the case of phenol, or by the rise of the freezing point of the water[16] owing to the withdrawal of ether from it.

As was to be expected, the laws of osmotic pressure are subject to numerous exceptions, or rather modifications, for, strictly speaking, no true law of nature is ever subject to an exception. That which, by a figure of speech, we call an exception is really a modification due to the simultaneous application of some other law. The modifications in this case are very similar to the modifications of Avogadro's law, which retarded its acceptance for nearly a half century. Vapor densities are abnormally high on account of the associative tendency of molecules, as in the case of acetic acid, or when too near the boiling point of the liquid, or low on account of dissociation, as in the case of ammonium chloride or of phosphorus pentachloride. In a similar manner the molecular weights of most acids when determined in solution in benzene are twice their normal value, while the molecular weights of electrolytes dissolved in water, and sometimes when dissolved in other solvents, are less than we should expect. In addition to the modifications of the law due to association and dissociation are other modifications similar to the modification of the laws of Boyle and Charles for gases which are highly compressed. These cases have been studied and formulæ for the deviation, based on the formulæ of van der Waals for compressed gases, have been given by Ostwald, Bredig and A. A. Noyes.[17] These formulæ give a satisfactory expression for the deviation in many cases of concentrated solutions. When we consider that strong solutions often give osmotic pressures of many atmospheres, and that the molecules of the bodies in solution are often much more complex than the molecules of most gases, it is readily seen that deviations of considerable amount may be expected.

In 1884 Arrheneus[18] published the results of researches on the electrical conductivity of solutions, on which he had been engaged for two years. In the course of his studies he was led to the conclusion that only a part of the molecules of an electrolyte are concerned in conveying the electrical current, and that it is necessary to distinguish between 'active' and 'inactive' molecules in this regard. The conductivity is greater, in proportion to the amount of the electrolyte present, for dilute than for concentrated solutions, and for an infinite dilution the molecules would, presumably, become all 'active.' Arrheneus pointed out, also, that there is a close connection between the number of 'active' molecules as determined by the electrical conductivity of solutions and the 'avidity' of acids as determined by the thermo-chemical researches of Thomsen. His first explanation of the cause of the difference between the 'active' and 'inactive' molecules, was, however, unsatisfactory and was not well received.

Shortly after, in his first development of his theory of solutions, van't Hoff was compelled to admit that many substances in aqueous solutions cause a depression of the freezing point much greater than they should in proportion to their molecular weights. He expressed the deviation by use of a factor, 'i,' which is, for electrolytes, always greater than unity and expresses the number of times the depression exceeds the theoretical depression as calculated from the molecular weight. This factor was at first considered to be a constant, but it is now known that it is variable and that it increases with the dilution. The obvious meaning of this factor is that the molecules of electrolytes are separated into two or more parts when dissolved in water, or other liquids which have a similar effect in causing electrical conductivity. But, just as chemists were very slow to see that the abnormal densities of ammonium chloride and of many other substances are due to dissociation, so van't Hoff did not draw a conclusion which seemed to be so contradictory to all preconceived notions about the bodies in question. Arrhenius, however, saw the logical conclusion, and his studies had prepared him for its acceptance. As a result, he proposed, in 1887, his theory of electrolytic dissociation.[19]

This theory, which seemed at first very improbable, has shown itself capable of coordinating the facts of many diverse fields of work and has proved more valuable in the incentive which it has given to research and more prolific of results than any other theory proposed during the last decade. According to the theory, an electrolyte when dissolved in water, and sometimes when dissolved in other solvents, is separated more or less completely into its ions.

The empirical basis for the theory lies in the correspondence between electrolytic conductivity and the divergence from the normal depression of the freezing point and lowering of the vapor pressure; in the correspondence of both with the 'avidity' of acids which has already been referred to; in the quantitative connection between each of these and the chemical effect of acids as shown in the inversion of cane sugar and saponification of methyl acetate; in the satisfactory explanation which it gives for the independent migration of ions during electrolyses as established by the work of Hittorf, Kohlrauch and others; in the fact that an electrolyte obeys the same law for dissociation with increasing dilution as a gas under diminishing pressure, first pointed out theoretically by Ostwald and Planck,[20] and then experimentally established by Ostwald, Wildermann,[21] Loomis,[22] and others; and in general by the fact that the properties of a dilute solution of an electrolyte are dependent on the sum of the properties of the ions present rather than on the properties of the chemical compound which those ions may combine to produce. It would take me too far to illustrate this last statement as shown to be true of the density, color and other properties of solution.

The theory has thrown light upon many chemical riddles and has placed the chemist in a position to predict phenomena which could formerly be known only as the result of experiment. It suggests at once the distinction between reactions of ions and reactions of bodies which do not undergo ionic dissociation. The former take place in solutions at ordinary temperatures and so instantaneously that the time factor cannot be measured; the latter frequently require an elevated temperature and are sometimes very slow. The distinction is, perhaps, a practical, rather than a strictly logical one, for theoretical considerations lead us inevitably to the conclusion that only additive reactions, and in many cases not even those, can take place without a previous dissociation of some sort. In this view the distinction between ionic reactions and others is that in solutions of electrolytes a considerable portion of the compounds have undergone dissociation; and as any ion is removed by precipitation, or otherwise, the remainder of the compound of which it is a part undergoes rapid dissociation, owing to the resulting dilution of the solution. In such cases the dissociation appears to take place almost exclusively at one point in the compound, and the reactions are clean and practically quantitative. In what may be called non-ionic reactions, on the other hand, the initial dissociation appears to be trifling and, notably with organic compounds, may take place at several points; the reactions between the resulting parts must be slow and may give rise to a variety of compounds.

In accordance with the theory, only those elements or groups which exist as independent ions in a solution enter readily into combination with other ions. Hence an atom which forms a part of a complex ion as the iron of ferro- or ferri-cyanides and the chlorine of chloro-platinic acid and of potassium chlorate cannot be detected by the ordinary reagents for these elements. This principle is of fundamental importance for analytical chemistry and has, of course, in its empirical form, been long recognized.

In the case of analytical chemistry, especially the new theories of physical chemistry seem destined to transform what has been, hitherto, an almost exclusively empirical science and raise it to a higher plane. Two illustrations of practical applications of the theory in this field may be of interest.

The first is with regard to the indicators used in acidimetry. It has long been known that the same indicator is not equally satisfactory in all cases, but the reason has never been clearly stated till recently. The principles on which the discussion depends are these: an acid solution is characterized by the presence of free hydrogen ions, a basic solution by the presence of free hydroxyl and free metallic ions; in the case of a strong acid or base the number of hydrogen or hydroxyl ions is large in proportion to the quantity of the acid or base present, while in the case of a weak acid or base the number of ions is small; in other words the difference between strong and weak acids and bases is that the dissociation factor of the former is very much the larger. The indicators in use are relatively weak acids or bases for which the free ions possess a different color from that of the pure acid or base. Thus phenol phthaleïn is colorless, while its ion is red; litmus is red, while its ion is blue. In the presence of hydrogen ions the dissociation of each of these substances is diminished in accordance with the well known law of dissociation that the presence of one of the products of dissociation decreases the dissociation of the compound. Hence in acid solutions these bodies are so little dissociated that the color of the compound, and not that of the ion, appears. In alkaline solutions, however, the color of the ions is developed, since the potassium and sodium salts, even of very weak acids, are largely dissociated in dilute solutions.

There is, however, a very considerable difference in the dissociation factors for the different indicators. The dissociation factor is much higher for methyl orange and for cochineal than for litmus and phenol phthaleïn, and while the dissociation factor of hydrochloric and similar acids is so high that a very small excess will cause the change in color, even of methyl orange, the dissociation factor for many acids, and especially for most organic acids, is so low that a quite appreciable excess is required, and the change in color will be slow and uncertain. Hence methyl orange and cochineal are entirely unsuited for the titration of weak acids, and litmus or phenol phthaleïn must be used. For weak bases, and notably for ammonia, the conditions are reversed. The salts of such bases with phenol phthaleïn, or with litmus, undergo hydrolysis in dilute solutions, and a considerable excess of the base will be required before the ions characteristic of the indicator will appear. The salts of the same bases with methyl orange or cochineal are not so readily hydrolyzed, and these indicators are more suitable.

A practical complication arises from the presence of carbonic acid in most of the solutions which we titrate. I will not take the time here to discuss the details of the theory which points out very clearly that, for accurate results, carbonic acid must be excluded from solutions in which litmus or phenol phthaleïn are employed, while, if concentrated, methyl orange or cochineal may be used satisfactorily for strong acids.

The other illustration of the application of the principles of physical chemistry to an analytical problem is one recently given by Stefan Bugarsky.[23] A great many methods for the separation of bromine and chlorine have been developed, but nearly or quite all of them rest on a purely empirical basis. Bugarsky has studied the subject from an entirely different point of view. Sometime since Bancroft[24] determined the electromotive forces developed between oxidizing and oxidizable solutions connected by an indifferent electrolyte, and with a platinum electrode immersed in each. The results may be considered as giving a quantitative expression for the relative oxidizing and reducing power of the various substances studied. Among other things it was found that, no matter what substance was oxidized, iodic acid with sulphuric acid develops a greater electromotive force than bromine with potassium bromide and less than chlorine with potassium chloride. It appears, therefore, that iodic and sulphuric acids together should liberate bromine, but not chlorine, from a solution containing bromides and chlorides. The practical application of this theoretical conclusion appears to have been entirely successful.

It is not alone in chemistry that the theories of osmotic pressure and of electrolytic dissociation have proved of practical value. Nernst has developed from these theories a theory for the cause of the electromotive force in batteries, which, while it may not, as yet, have received general acceptance, is a more useful expression for our present knowledge than any previously proposed. The most important conception at the basis of this theory is that of what may be called a solution pressure for metals, corresponding in some sense to the vapor pressure of liquids. When zinc, for instance, is in contact with water, or an aqueous solution, this solution pressure is a force impelling the atoms of zinc to pass into solution. In order that they may do so, however, each atom must pass over into the state of an ion; that is, it must receive a charge of positive electricity which is carried with it into the solution. But only a very few atoms can pass into solution before the negative charge left in the mass of the zinc in proportion as the positive ions separate from it will cause such an accumulation of zinc ions in proximity with the zinc as to balance the solution pressure. If, however, an opportunity is given for the escape of the negative charge from the zinc, and at the same time positive ions are allowed to escape from the solution at some other point, the zinc will continue to dissolve and currents of electricity will be set up. Thus, in the Daniell, or gravity cell, zinc ions pass into solution and a corresponding number of copper ions are deposited. The force which causes the movement of the ions, and with them the transference of electrical energy within the cell, is mainly the very high solution pressure of the zinc as compared with that of copper. Other factors, such as the osmotic pressure of zinc ions already in solution, which tends to conteract the solution pressure of the zinc, the osmotic pressure of copper ions which aids in the separation of the copper, and the different velocity of translation for various ions which may cause differences of potential when the fluids of the cell are not homogeneous, are most of them comparatively small in their effect.

No means has been found for the direct determination of the solution pressure of metals, but it may be calculated from the difference in potential between a metal and a solution of one of its salts. Methods for the determination of the latter have been devised by Ostwald,[25] and improved by Paschen.[26] By the use of these and other constants which the researches of physical chemistry have placed in his hands, the physicist can now calculate the electromotive force which can be obtained by various combinations of metals and solutions. On this side the theory has rendered essentially the same service for the galvanic cell which the atomic theory rendered for chemical compounds when it furnished the means for calculating their percentage composition. As in the early days of the atomic theory, many of the constants in question are imperfectly known, but since the theory has shown clearly their interdependence, new means for their determination and for the control of their accuracy are constantly being discovered.

Every one who is familiar with the extremely wasteful character of all processes now at our disposal for the transformation of chemical into mechanical energy must have had the thought that there is surely some means of saving a part of the enormous loss. At present the attention of the scientific world is turned toward the transformation of the chemical energy of coal into electrical energy as the probable solution of this problem. It seems to be almost certain that physical chemistry has already made clear the principles by means of which such a transformation may be accomplished. Indeed, Dr. W. Borchers,[27] by the use of a solution of cuprous chloride with producer gas, or carbon monoxide on one side and air on the other, has already obtained an electrical current which corresponds to a transformation of thirty per cent. of the chemical energy into electrical. This is an efficiency three times that of the best steam engines. There is no probability that this method can ever be practically useful, but that a practical method will soon be discovered is, at least, possible.

I have thus far spoken of the achievements of physical chemistry mainly in the direction of the development of the theories of osmotic pressure and of electrolytic dissociation. It is in this field that the most valuable practical results have been secured, because it is here that a new, far-reaching, and extremely useful theory has been developed. But work in physical chemistry has been extremely active in many other directions as well.

The most brilliant chemical discovery of the last decade was a result of the careful study of a single physical property of nitrogen. And, owing to the peculiar character of argon and helium, their further study has been almost exclusively on the physical side.

Ramsey and Shields,[28] by their work on the surface energy of homogeneous liquids, have developed a method for the determination of the molecular weights of this class of bodies.

Traube's exhaustive study of the specific gravity of solutions, promises, if all that he claims be true, and much of it seems to be, to bring order out of an almost interminable chaos of empirical data. Among other things his work has given a new and very rapid method for the determination of molecular weights.

I will not take the time to refer in detail to the work of Brühl and others on the refraction and dispersion of light as dependent on the composition and structure of bodies; to the work of Thomsen, of Stohman and of Berthelot upon thermo-chemistry; to the work of Guye, Walden and others on specific and molecular rotation, and of Perkin on electro-magnetic rotation of polarized light; and to the work of Rowland on spectrum analysis.

In all of these fields and in many others a vast accumulation of empirical data has been secured. This wealth of experimental material has been accompanied and supplemented by theoretical discussions, and many interesting relations have been discovered. Physical chemistry has proved one of the most enticing and profitable fields for work in recent years and claims many enthusiastic investigators in our own country as well as abroad. In the development of the subject perhaps no one has contributed more than Ostwald by his Lehrbuch and by his ably edited Zeitschrift für physikalische Chemie. We may congratulate ourselves that our workers in America are now to have a journal of their own, and we may confidently hope that the new *Journal of Physical Chemistry* will contribute much toward 'the chemistry of the future.'

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SECTION C.—CHEMISTRY.

Section C. of the American Association for the Advancement of Science was called together for organization on Monday, August