to examine preparations of our products under the microscope, and were repeatedly surprised to observe that solutions containing apparently but a very small amount of raffinose, would completely crystallize under the microscope into forms which could scarcely be distinguished from pure raffinose.

The accompanying illustrations demonstrate this tendency far better than any mere description.

No. 1 shows the normal form of the crystals of pure sucrose seen under the microscope.

No. 2. The crystals of pure raffinose.

No. 3. The crystal form of a mixture of the two with a specific rotation of 76°.

It is to be noted that the influence of the raffinose is manifested not in the appearance of isolated raffinose crystals but in a modified and abnormal appearance of all of the crystals, which are all similar to each other but entirely distinct from those of sucrose.

This tendency on the part of raffinose to so distinctly modify the crystals of sucrose, would seem to afford a means of readily identifying the presence of small amounts of the former in mixture with sucrose.

OUR PRESENT KNOWLEDGE OF ARGON, WITH A PARTIAL BIBLIOGRAPHY.

By C. LeRoy Parker.

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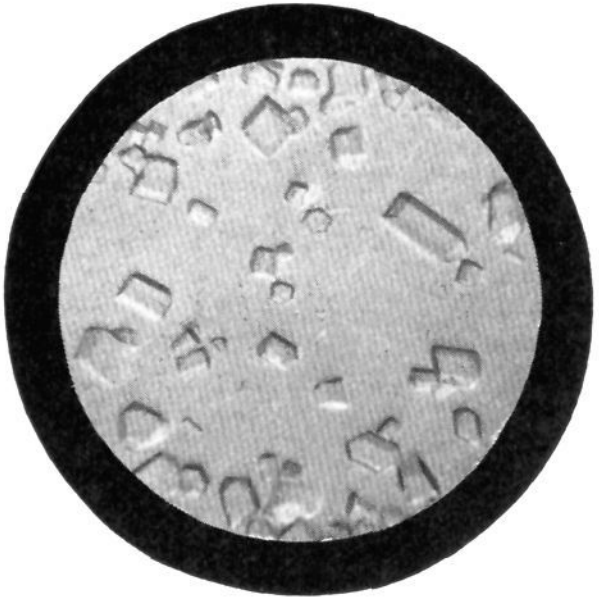
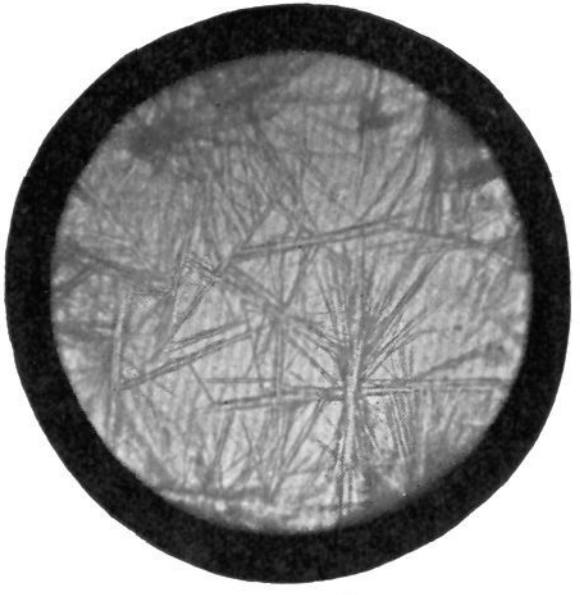
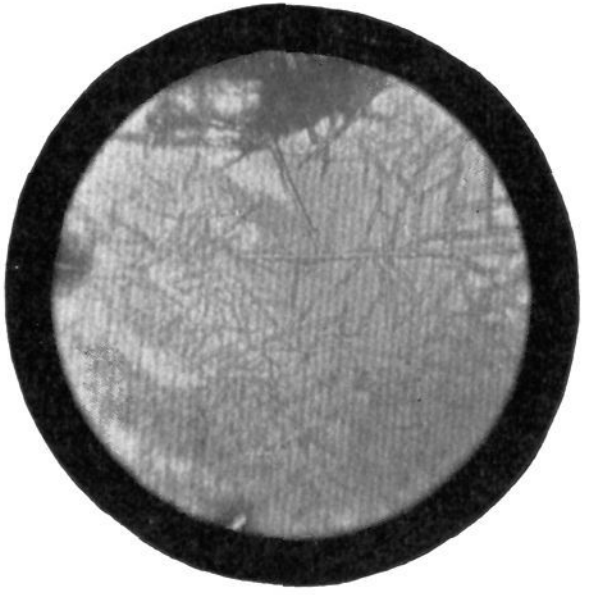
SINCE the appearance of the account by Lord Rayleigh and Prof. Ramsay, of their joint discovery, there has been evident, in this country and abroad, a general air of expectation, which even the remarkable results that have been obtained have not been sufficient to remove. In fact, instead of allaying this expectancy it has rather been increased, by the accounts of researches thus far made, so that now, even more than at first, the scientific world is looking forward to the solution of the remaining problems concerning argon, with an interest, rendered particularly intense by the commonly accepted conviction that, upon the nature of the solution will depend, in a great measure, our views of certain chemical theories which we have heretofore regarded as firmly established. While the results of the inves-



No. 1. Pure Sucrose.
Specific Rotation 66.5°.



No. 2. Pure Raffinose.
Specific Rotation 103.5°.



No. 3. A Mixture of Sucrose and Raffinose.
Specific Rotation 76°.

tigations of the properties of argon fall far short of clearing up many uncertain points concerning the character of the gas, they are not without direct and important bearings on them and it is with a view of presenting and discussing the essential features of these results that this paper has been prepared.

The account of the discovery and early researches on argon, by Lord Rayleigh and Professor Ramsay, has been given such wide circulation and these have now become matters of such familiar knowledge to chemists that I will take up the time of the Society, in connection with them, only so far as is necessary to sum up their results and consider them briefly in connection with those subsequently obtained.

In order to successfully study the character of argon it was necessary, first of all, to obtain it in considerable quantities in a pure state. The first method, sometimes and very appropriately called the Cavendish method, since it is the one by means of which Cavendish isolated and measured an impurity in atmospheric nitrogen, consisted in passing electric sparks through a mixture of air and oxygen—mixed in about equal quantities—confined in a eudiometer over dilute alkali. When all of the nitrogen had combined with the oxygen and been absorbed by the alkali, the excess of oxygen was absorbed by alkaline pyrogallate, leaving the required gas free. A second method, by means of which argon could be obtained more satisfactorily and in larger quantities, consisted in passing pure dry air over red hot copper, which removed the oxygen, and then over heated magnesium, which removed the nitrogen, the inert gas being left. This process, which is here merely outlined, required a circulation of not far from two days through the system of tubes containing the absorbents before the argon could be obtained pure. The argon used in many of the most recent investigations was obtained by keeping up an electric arc in a mixture of air and oxygen for about two days, removing the nitrogen and oxygen as already described. Other methods for obtaining argon have been suggested, but their utility has yet to be demonstrated.

The proportions in which argon exists in the atmosphere was ascertained by the experiments which led to the discovery of the gas, to be about $\frac{1}{125}$ by volume and $\frac{1}{120}$ by weight. The most

recent investigations on this point are those of Th. Schloesing,[88] who has found that the proportion of argon in atmospheric nitrogen is 0.0119, a result which agrees to a remarkable degree with those obtained by Rayleigh and Leduc. Schloesing has further shown[88] that in the case of argon, as with oxygen and nitrogen, its proportion in the atmosphere varies to a degree scarcely perceptible on analysis. Kellas, who has made researches along somewhat the same line as those of Schloesing, has found[60] that argon exists in a greater proportion in respired air than in normal air. In the researches on the occurrence of argon other sources than the atmosphere have been discovered. It has been found to be given off in small quantities, in connection with other gases, when clevite, samarskite, euxenite, and a few other minerals are treated with acids or heated *in vacuo*,[33] but as yet the condition in which argon exists in these minerals has not been determined.[68] A still more interesting source of argon is that of meteoric iron, which was found in Augusta County, Virginia. The specimen was analyzed by Prof. Ramsay and found to contain the inert gas.[35] Argon has been discovered in the waters of a number of mineral springs. Bouchard has found it in certain springs in the Pyrenees,[49] Kayser in the springs of Wildbad, in the Black Forest,[48] Rayleigh in the Bath Springs,[77] and Ch. Mouren has found that it constitutes a part of the gases which escape abundantly in bubbles from the spring of Maizières[64] (Côte d'Or). Bedson and Shaw have found argon in the gases enclosed in rock salt,[41] and Th. Schloesing has discovered it to be a constituent of fire damp and of the gas of Rochbelle.[83] Argon has been sought for without success in animal and vegetable substances[23] by Geo. MacDonald and A. M. Kellas, but Schloesing and Richard, as a result of an investigation, which shows something of the extent to which researches on the occurrence of argon have been carried, have found that it forms a part of the gases of the natatory bladder of fishes and physaliæ.[74]

Concerning the true character of argon there have been many and varied opinions advanced, but none so widely accepted as the one originally advanced by the discoverers of the gas, which is that it is an element and monatomic.[5] It has been suggested that argon might be an oxide of nitrogen, a view speedily dis-

carded when the methods of isolating argon became better understood. Another view advanced by Dewar[3] and others is that argon might be an allotropic form of nitrogen bearing the same relation to that element that ozone does to oxygen. This would explain the concurrent existence of argon and nitrogen, and the fact that many of the lines of their spectra are near to each other. It would also explain the inactivity of argon and would allow it a proper place in the periodic system. Against this view are the facts that in decomposing compounds of argon no nitrogen is detected and that in no way has any one succeeded in obtaining nitrogen from argon or argon from pure nitrogen, and that, if we adopt N_3 as the formula of argon its density would not at all conform to the density found for argon. Further evidence against this theory is found in the results of researches by Peratoner and Oddo,[55] who, observing that the molecular weight of argon approximates that of a triatomic polymeride of nitrogen, made a number of experiments on a gas obtained by the decomposition of aziomide, but could find no trace of argon.

The view suggested by Crookes in his paper on the spectra of argon,[9] that the discoverers of argon may have added "two members to the family of elements" has been somewhat extensively adopted. This view exhibits the least incompatibility with our present theories of chemistry and, if shown to be the true one, will remove the grave suspicions which since the discovery of argon have rested on the periodic classification of the elements. The fact of the duality and simultaneous appearance of the spectra of argon, which, as Friedländer has pointed out,[55] has been regarded as evidence of the compound nature of the gas, cannot rightfully be so regarded since the same phenomena have been observed by W. N. Hartley in connection with nitrogen, which is of undoubted elementary character.[56] The observation of three different spectra of argon, by Eder and Valentia,[71] appears to have a very important bearing on the theory suggested by Crookes and to deserve careful consideration in forming our opinions concerning the elementary or compound character of argon.

Whatever other light spectroscopic investigations may throw upon this question, this much is certain, as Hartley has pointed

out,[86] that since no compound gaseous substance known can withstand the temperature of the electric spark, without exhibiting the spectra of its elements, and since the spectra of argon are not those of any other known substance it follows that, if the gas is a compound, all of its constituent elements have been hitherto unknown.

The most commonly accepted view concerning argon, *viz.*, that it is an element, is based chiefly upon the evidence of its being monatomic. As pointed out in their original paper,[3] Lord Rayleigh and Prof. Ramsay found the ratio of the specific heats of argon to approximate 1.67, which is the theoretical ratio of the two specific heats of a monatomic gas.

In the absence of contradictory evidence, this fact would have been considered satisfactory proof of the monatomicity of the argon molecule and the consequent elementary character of the gas, but the density of argon, having been found to be approximately twenty, the atomic weight would, if the molecule contains but one atom, be forty, in which case there would be no proper place for it in the periodic system. To accept one as the atomicity of argon means the overthrow of the present classification of the elements. On the other hand, to assume that argon has an atomicity other than one, in which case it finds a proper place in Mendeléeff's system, is in direct opposition to the kinetic theory, which has not hitherto been called into question.

Still other theories have been advanced to explain the nature of argon. Nasini has suggested that it might be a diatomic gas. but with its atoms so nearly spherical as to make it resemble a monatomic gas in certain respects.[40] This view is thought to afford the best explanation of the peculiar character of argon by a great number of the chemists who doubt its monatomicity. Another view is that argon contains both monatomic and diatomic molecules. This view, which was referred to by Prof. Ramsay in his lecture given before the Oxford Junior Scientific Club on a recent date,[78] is regarded by him, as it is by chemists generally, as being improbable in the extreme.

In the investigation of the physical properties of the new gas the ground has been so thoroughly covered that we can hope for no extensive advance in this direction.

The density of argon, which it has thus far been found im-

possible to completely reconcile with its other properties, was found by Lord Rayleigh to be 19.94.[67] The refractivity and viscosity were found by the same physicist to be respectively 0.961 and 120, air being the standard.[53]

As regards the spectra of argon, the early and accurate knowledge of which has been of incalculable value to the investigators of the gas, Mr. Crookes[6] noticed that at the end of the capillary tube containing the argon, near the positive pole, there appeared a reddish, and at the end near the negative pole, a bluish glow, two spectra being produced, twenty-six lines of which appeared to be common.

In comparing the spectra of argon with the sharp line spectrum of nitrogen Mr. Crookes says: "The sharp line spectrum of nitrogen is not nearly so striking in brilliancy, number or sharpness of lines as those of argon, and the most careful scrutiny fails to show any connection between the spectra. I can detect no lines in common." Between the band spectrum of nitrogen and the spectra of argon Mr. Crookes observed two or three close approximations of lines, but no positive coincidences.

Eder and Valenta have observed that in addition to the red and blue spectra there is a third spectrum produced when very large condensers are used with a Ruhmkorff's coil and powerful currents employed in the primary circuit.[71] This third spectrum is distinguished from the other spectra in that many of the lines appear more distinct while others are less so, and by the appearance of entirely different lines as well.

In connection with the red and blue color, which Crookes has observed, it is interesting to note that, during the action of the silent electric discharge in a mixture of benzene vapor and argon, Berthelot observed an intense greenish fluorescence, and that this gave a spectrum showing lines and bands in the yellow, green, and violet, similar to those observed in the spectrum of the aurora borealis.[28] While none of the lines coincided exactly with the principal line of the aurora borealis, 557, the argon line 555.7, is very near the latter and Berthelot is convinced by his comparative study of the two spectra, that there is some relation between the presence of argon in the atmosphere and the production of the aurora borealis.

At the time of the discovery of argon no chemical properties

of the gas, other than its extreme inertness, had been discovered, hence the name argon, from the Greek prefix ἀ- privative meaning without and ἔργον, work. Although subsequent researches have shown the term to be, in a strictly literal sense, a misnomer, it is not to be overlooked that the name argon was only given after repeated attempts to induce chemical combination of the gas with other elements had been made.

The following is a list of the more important experiments made at that time and subsequently, with a view of studying the chemical properties of argon. It was mixed successively with oxygen, hydrogen, and with moist and dry chlorine and subjected to the action of the silent electric discharge without undergoing change. It was brought in contact with sulphur and phosphorus at a red heat and no change took place. Tellurium was distilled in the gas, as were sodium and potassium, without losing their luster. It was not absorbed by red hot caustic soda nor soda lime. It passed unaffected over red hot potassium nitrate and sodium peroxide. Persulphide of sodium and calcium were without action on it at a red heat. Platinum black did not absorb it nor did platinum sponge. Wet oxidizing and chlorinating agents, as potassium permanganate, aqua regia, bromine water, bromine and alkali, and hydrochloric acid were entirely without action upon the gas. It resisted the action of nascent silicon and boron, as proven by contact with a mixture of sodium and silica, and one of sodium and boric anhydride. It was subjected to the influence of the electric spark in contact with fluorine, boron, titanium, lithium, and uranium without inducing chemical action.

An apparent combination of argon and carbon was observed by Prof. Ramsay by making an arc between two rods of purified carbon in an atmosphere of argon.[43] Further investigation along the same line,[76] however, here led to the conclusion that argon does not combine with carbon even at the temperature of the electric arc and that the expansion which led to the inference that a chemical union had taken place was due to the carbon dioxide gas occluded on the surface of the carbon not being entirely removed, before the rods were introduced into the argon receiver. As further evidence bearing upon the possible union of argon and carbon, a mixture of carbon tetrachloride and argon was exposed for several hours to a silent electric dis-

charge. The argon did not enter into combination but was recovered without loss.

Other attempts to induce argon to enter into combination have been singularly successful. Berthelot, by means of the silent electric discharge, has effected the union of argon with benzene,[20] carbon disulphide,[36] and apparently with mercury. Berthelot describes the product formed by the union of argon with the elements of benzene, the first compound of argon produced in the laboratory, as consisting of "yellow resinous matter which, under the action of heat, decomposes, forming volatile products and leaving a bulky carbonaceous residue."

Troost and Ouvard appear to have been able to effect the combination of argon with the vapor of magnesium by means of a powerful electric discharge.[50] Argon contained in a tube, furnished with magnesium wires connected with a Ruhmkorff's coil, was found to gradually diminish in volume under the influence of the discharge and finally to disappear, seemingly becoming united with the vapor of the magnesium. Under the same conditions platinum appeared to volatilize and combine with the argon in the same manner as the magnesium.

P. Villard has found that upon being compressed to 150 atmospheres, in presence of water, argon combines with the water to form a crystalline and dissociable hydrate, which is analogous to the hydrates of gases already known.[84] Almost as remarkable as the discovery itself is the failure on the part of chemists to detect argon previous to 1894. Cavendish isolated the gas, apparently obtained it in a pure state, and the account of his research has been in the hands of chemists for a hundred years. Simply because Cavendish inferred the relative freedom from impurities of his "phlogisticated air," from the smallness of the residuum he obtained, chemists were willing to leave the critical investigation of this residuum to the future until its explanation became necessary in clearing up other difficulties. It would be hard to find an example of a fallacy, arising from undue devotion to authority, more instructive than that of our mistaken views regarding the composition of the atmosphere previous to Lord Rayleigh's and Prof. Ramsay's brilliant discovery.

It is further interesting, in connection with the long delay in

detecting argon, to note the researches of Mallet, who, in 1870, published an account of an industrial method for preparing oxygen,[1] in which he unknowingly describes what could not fail to be the production of, if not pure, at least highly accumulated argon in connection with oxygen. Mallet's method consists in repeatedly pumping air into water, under high pressure, liberating the absorbed gases and again pumping them into water, repeating the process until the oxygen is sufficiently freed from other gases, for the use to which it is to be applied.

The isolation of the argon from the nitrogen would result, as Brauner has pointed out,[17] from the superior solubility of the former gas, the solubility of argon in water being to that of nitrogen in the ratio of 2.5 to 1. Repeating the process eight times produces a gas which, according to Mallet, consists of 97.3 per cent. of oxygen and 2.7 per cent. of nitrogen. That this so-called nitrogen is really nearly pure argon there can be little doubt; in fact, that the action of the process is, as here described, is proven by the fact that the curve representing graphically the decrease in the percentage of nitrogen after each operation shows that after the first few repetitions of the process the absorption coefficient of the so-called nitrogen which is gradually becoming richer in argon, approaches the coefficient of absorption of oxygen which is approximately the same as that of argon.

The failure to detect argon previous to 1894 is the more remarkable from the fact that the records of the progress of chemistry abound with accounts of atmospheric research. As Brunner has said "L'analyse chemique de l'atmosphère a été de tout temps une des parties de la chimie qui est attiré le plus l'attention des savants." A single periodical, the *Chemical News*, *1860-1896*, describes about 200 investigations on air, and yet, notwithstanding the abundance of such researches, a careful survey of the literature on the subject since the time of Cavendish's classic experiments to those of Rayleigh fails to reveal even so much as a surmise that the air contained any gas which had not been isolated and studied.

It is not the author's intention to infer from this that the discovery of a gas, of the character of argon, had never been pre-

[1] *Ding. poly. J.*, 199, 112; *J. für Gasbereitung*, *1870*, 538.

dicted, but merely to show that no such predictions has been made as a result of the study of the atmosphere.

There are two very remarkable instances of the anticipation of the discovery of argon. The first was that of Lieut. Col. Sedgwick, in 1890, and the second that of Boisbaudran shortly before the discovery of argon was announced.

Sedgwick, as a result of his study of the forms and relations of atoms, reached the conclusion that there must be an inactive element of the atomic weight commonly assigned to argon. He conceives atoms to be spheres with one or more flat places, the number of which correspond to the valence of the element. Sedgwick, on page 60 of his book "Force as an Entity," published in 1890, points out that an element whose atoms are perfect spheres, would be inactive and have an atomic weight of 20.

In his remarks on atomic weights[1] Boisbaudran gave a classification of the elements which led him to assume the existence of an unknown family of elements of atomic weights 20.0945, 36.49 ± 0.08, 84.01 ± 0.02, and 132.71 ± 0.85. These elements, according to Boisbaudran, should be non-metallic, of even atomicity, the first two abundant in nature, and the member having the atomic weight 20.0945, which he now identifies with argon, should be more volatile than oxygen.

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A RAPID METHOD FOR THE DETERMINATION OF SILICON IN SILICO-SPIEGEL AND FERRO-SILICON.

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IN 1894 Furnace "A" at the Edgar Thomson Steel Works made the first silico-spiegel ever manufactured in this country. A determination of silicon and manganese was required in each cast, the silicon especially, as soon as possible.

The "aqua regia" method was the one tried. Various strengths of acid were used, but it was found almost impossible to get the metal into solution, even after passing the crushed "shot" through bolting-cloth. As high as 400 cc. aqua regia was used on one-half gram, the aqua regia being four parts nitric acid and three parts hydrochloric acid.

The fusion method, as recommended by Williams,[1] was also tried, but we were never able to make a determination in less than six to eight hours. The furnace made only a short run on silico-spiegel and for the time the matter was dropped. Last year we again made some silico-spiegel and were successful in finding a method for determining the silicon in a rapid and accurate manner and give the method in detail below.

Preparation of the Sample.—For the success of the method it is essential that the sample should be in a very fine state of subdivision. Grind in an agate or crush in a diamond mortar so that the sample will pass through a sieve made of bolting-cloth.

[1] *Trans. Am. Inst. Min. Eng.*, 17, 542.