

exist between these two properties may be quite obscured by the different impurities. This is what generally happens when different people study the properties of varying samples.

One of the most recently undertaken of all these investigations is the study of surface tension of these same organic liquids. Surface tension, as you well know, is that tendency (caused doubtless by the cohesion of the molecules of substance) which forces any liquid surface to contract as much as possible, thus making drops and bubbles spherical, and drawing liquids up in capillary tubes. Surface tension is of peculiar interest in relation to the theory of compressible atoms, because it gives another clue to the cohesive forces holding matter together in the liquid and solid condition; but the published data even for such a common substance as water vary widely, often as much as 10 per cent. Hence the more careful study of this important property formed a legitimate part of the scheme of investigation for which the Wolcott Gibbs Memorial Laboratory was planned. In the first year of this research, which is being continued at the present time, we were able to find most of the important causes of the serious divergences in earlier work. Many of the experimenters had immersed their capillary tubes (in which the effect was to be measured) in other tubes much too narrow for the purpose, not realizing that even a tube one inch in diameter causes an appreciable "capillary" rise of the liquid contained within it. Again, they failed to allow for optical imperfections in the glass of the tubes containing the liquid, their methods of measurement were sometimes inadequate, and the mathematical formulæ used for calculating the results were often faulty. Therefore, in our preliminary work, which was reviewed briefly in the

July number of the *Proceedings* of this academy, it is hoped that a distinct advance has been made.

If high quality had not been sought, of course the number of investigations could have been much greater. Some one has wisely said that the output of physico-chemical work is inversely proportional to the square of the grade of accuracy desired. In the brief space of these few minutes it has been impossible to give much more than a mere list. Those of you who are specially interested will find many of the researches already published in full; brief accounts of others are in the Year Books of the Carnegie Institution of Washington, and before long it is hoped that the rest also may be printed.

An investigator for whom much has been done feels gravely the responsibility which rests upon him of doing much in return; and although in this case he feels the necessary human inadequacy and incompleteness of the work just described, nevertheless he hopes that at least a beginning of accomplishment has been made, and that in the future the Wolcott Gibbs Memorial Laboratory, through many years, will yield ever increasingly useful additions to the sum of human knowledge.

THEODORE W. RICHARDS

HARVARD UNIVERSITY

THE LIFE OF RADIUM

THE life of radium, or the length of time required for a given quantity of radium to be transformed and converted into other elements, is a physical magnitude of considerable importance and interest. Its chief significance lies perhaps within the special field of radioactivity where radium occupies a unique position in being the only highly radioactive radio-element which possesses physical and chemical properties, and occurs in a sufficiently high state of

concentration, to permit its being obtained in reasonable quantities in an isolated and purified condition. For this reason radium is considered and accepted as a standard or typical radioactive substance, and its physical and chemical properties, including the value of its atomic weight, are known with a considerable degree of precision. For some time in the future, therefore, radium will occupy this position of relative importance and will serve as the basis for calculation and comparison with other radio-elements possessing less striking chemical individuality.

An accurate knowledge of the life of radium is also important in the field of geology, because of a method which is available for estimating the geological antiquity of some of the older rocks and minerals. This method is dependent on the determination of the progress of the radioactive disintegration which has taken place in those minerals containing appreciable proportions of uranium. For the accurate calculation of these important magnitudes an exact knowledge of the rate of disintegration of radium is essential.

It is possible, moreover, to obtain an estimate of the probable life of radium by a calculation involving as its basis a number of other important physical constants. These constants will be referred to more specifically later. If a knowledge of the life of radium can be arrived at by experimental methods not directly involving these constants, then, if the results given by the different methods are in good agreement, there is good reason for assuming that the accepted values for these constants are not very different from the true values.

The disintegration of radioactive substances is of such a character that the transformation of the substance into other elements can be expressed by a law in which the rate of transformation is an exponential

function of the time. The rate of transformation is independent of the amount of material undergoing disintegration and is independent of the temperature, the pressure or of any other external condition to which we can subject the radioactive substance. It proceeds in such a manner that if half of the material present is transformed in a given period of time, half the remaining quantity will be transformed in a subsequent time of equal duration, and half the amount still left will undergo change in the third equal interval. This will continue indefinitely until the amount remaining will be too small to merit consideration. Since under these conditions some of the atoms of the radio-element will have an inappreciably short life, while others will have an inconceivably long one, it is impossible to attach any special significance to the term "life of" a radio-element except under certain definite restrictions. The life of a radio-element may therefore be somewhat dogmatically expressed in terms of the time required for exactly one half of it to be transformed into other substances. This constant is known as its "half-value period," and it is the half-value period of radium which particularly concerns us at the present moment.

The first estimate of the probable life of radium was published by Sir Ernest Rutherford in the first edition of his textbook "Radio-activity (Cambridge, 1904). In the disintegration theory proposed by Rutherford and Soddy the assumption was made that the expulsion of α -particles by radium and other radio-active substances was coincident with the changes taking place on the disruption of the atoms, namely, that the appearance of the α -particles was indicative of the simultaneous breaking up of the atoms of the radio-element. Rutherford further postulated the theory that each α -particle had its

origin in the disintegration of a single atom, or in other words, that each changing atom gave rise to a single α -particle. If this assumption were correct, then an estimate of the total number of α -particles emitted by any radioactive substance would afford a basis for judging as to the number of atoms which underwent transformation in any given period. From the results of experiments by Wien on the number of β -particles projected from one gram of radium bromide, and from considerations based on the ionization produced in a gas by the α -particles emitted by a known quantity of radium, Rutherford reached the conclusion that one gram of pure radium element expelled 2.5×10^{10} α -particles per second. From data based on experimental evidence it was assumed that the number of molecules in one cubic centimeter of hydrogen at standard pressure and temperature was 3.6×10^{19} . Taking the atomic weight of radium as 225 it was then calculated that there were 1.8×10^{21} atoms in 1 gram of radium.¹

If the total number of atoms present was 1.8×10^{21} and the number transformed per second was 2.5×10^{10} , then the fraction of the whole undergoing change per second would be 1.4×10^{-11} , and per year 4.4×10^{-4} . This indicated that the half-value period of radium was about 1,500 years.²

Another estimate of the life of radium was made by Rutherford in the Bakerian lecture delivered before the Royal Society in May, 1904. Assuming that the heating effect, which had been observed and measured by P. Curie in radium salts, was due to the bombardment of the salt by the α -particles emitted from the radium which it contained and concluding that heat

energy which appeared was derived from the kinetic energy of the moving α -particles, Rutherford calculated the kinetic energy of a single α -particle on the basis of the data then available. This he found to be 6×10^{-8} erg per second. The heating effect of about 100 gram calories per hour observed for one gram of radium corresponded to 1.2×10^6 erg per second. Considering the radium salt as containing four α -ray products (Ra, Ra Em, Ra A and Ra C) and assuming an equal distribution of the heating effect between these, it therefore appeared that the number of α -particles expelled per second per gram of radium itself (and therefore the number of atoms of radium breaking up per second) was 5×10^{10} . Applying the same line of reasoning as had been used in the first instance for deriving the number of atoms in one gram of radium, Rutherford obtained the value of 800 years for the half-value period of radium.³

In the year 1905 Rutherford⁴ performed an experiment in which the electrical charge carried by the α -particles from a known quantity of radium was measured. This was found to be equivalent to 4.07×10^{-9} ampere per second for the particles emitted by one gram of radium. Assuming the charge on each particle to be the same in value but opposite in sign to the charge carried by a single electron; viz., 1.13×10^{-19} coulomb, this gave the number of α -particles per second from one gram of radium as 6.2×10^{10} . Estimating, in this case without the previous error, the number of atoms in one gram of radium as 3.6×10^{21} , the value obtained for the rate of change of radium corresponds to a half-value period of about 1,300 years.

A new and more accurate determination

¹ An error was made in this calculation, and the correct number based on the data used should have been 3.6×10^{22} . This would have given 3,000 years for the half-value period.

² See preceding footnote.

³ The error mentioned previously was repeated here, and the correct value given by this calculation is not 800, but 1,600 years.

⁴ *Phil. Mag.*, 10, p. 193.

of the deflection of the α -particles from radium in a magnetic and an electric field was made in 1906 by Rutherford.⁵ This gave a value of 5.1×10^8 for the ratio of the charge to the mass (e/m) of an α -particle. Since the value of e/m for the hydrogen ion in the electrolysis of water is nearly 10^4 , Rutherford decided that of a number of possible explanations of these two differing values, the most probable one was that the α -particle consisted of an atom of the element helium (atomic weight 4) with a charge twice that of the electron. If this assumption is introduced into the last previously considered calculation of the life of radium, the number for the half-value period comes out 2,600 years instead of 1,300 years.

In 1908 Rutherford and Geiger⁶ devised an experiment in which the actual number of α -particles emitted by a known quantity of radium could be accurately counted. They also accurately measured the charge carried by a known number of these particles, and demonstrated the correctness of Rutherford's earlier assumption that the charge on a single particle was twice that carried by a single electron. From the counting experiments it was evident that the number of α -particles emitted per second from one gram of radium was 3.57×10^{10} . The results of these experiments also gave data from which a more accurate estimate could be made of the number of atoms in one gram of hydrogen, viz., 6.2×10^{23} . Using the numbers thus derived the magnitude of the half-value period of radium was again calculated and found to be 1,690 years.

A direct determination of the rate of disintegration of radium by measurements of the decrease in radioactivity of a given radium salt is not practicable from an ex-

perimental standpoint. The rate of disintegration is so relatively slow and the experimental difficulties of accurately measuring the very small yearly decrease in the amount of radium present are so insurmountable that this method of attacking the problem is practically excluded. There is, however, a way in which a knowledge of the life of radium can be obtained which depends upon very different principles from those involved in calculations employed by Rutherford. This method was first suggested and applied by the writer, and its general principles can be briefly described as follows:

The work of Boltwood, McCoy and others has conclusively demonstrated that radium is a transition product in the radioactive disintegration of the element uranium. The sources of radium consist solely of old minerals containing uranium. In these natural compounds the uranium has been undergoing transformation for long periods of time and the products of its disintegration have been accumulating and have been retained in association with the unchanged uranium in the mineral. Now the nature of the successive changes occurring in radioactive substances is such that, in any system such as that represented by a uranium mineral, after sufficient time has elapsed a comparatively simple relation will exist between the quantities of the different genetically connected elements present. The condition finally attained is known as a state of radioactive equilibrium. In this state a simple expression will define the relative amounts of the different, related radio-elements contained in the mineral, and, what is more important to our immediate interests, a very simple relation will exist between the amounts of the different radio-elements undergoing transformation in equal periods of time.

The rate of change of a radio-element is,

⁵ *Phil. Mag.*, 11, p. 348.

⁶ *Proceedings of the Royal Society*, A, 81, p. 141; *ibid.*, A, 81, p. 162.

so far as our knowledge extends, an unalterable and unvarying factor. It may be defined in terms of the fraction of the whole amount of the element present which undergoes transformation in any convenient unit of time, a year for example. This factor is called the constant of disintegration of the radio-element. Its character is such that if P represents the number of atoms of a radio-element initially present, e is the base of the natural system of logarithms, t is the time expressed in the chosen units, and λ is the disintegration constant; then the number of atoms, P_t , of the element which will remain unchanged after the expiration of an interval t units from the start will be expressed by

$$P_t = Pe^{-\lambda t}.$$

Now, in any radioactive system comprising a parent substance like uranium and a series of disintegration products, including radium, for example, when a state of radioactive equilibrium has been established the conditions will be such that the number of atoms of each of the radio-elements in the series which undergo change in a given interval will be the same and equal. Thus if U be the number of atoms of uranium and λ_1 be its constant of change, and if Ra be the number of atoms of radium with a constant of change λ_2 , then $\lambda_1 U = \lambda_2 Ra$, and this will also equal the product of the *number of atoms* of any other radio-element in the series multiplied by *its* disintegration constant. It should be evident from these considerations that the quantity (number of atoms) of radium formed in any given interval will be equal to the quantity (number of atoms) of radium which is transformed in the same interval, an essential requirement to the postulated condition of equilibrium. If, then, we can determine by experiment the quantity of radium which is formed in such a system, we obtain

through this a direct measure of the quantity of radium which has changed to other elements during the observed period, and if we know the amount of radium present in the system we can determine the ratio of the two amounts which will be the disintegration constant of the radium. If radium were formed directly from uranium it would be easily possible to separate the uranium from a quantity of mineral containing a known amount of radium, purify it from all but traces of radium, allow it to remain until measurable amounts of radium had been produced within it, and then compare the radium formed from the uranium with the radium present initially in the mineral. This was attempted, but it was found that the rate of production of radium was too slow to be determined with any accuracy and was far less than was to be expected from theoretical considerations. This obstacle was overcome when in 1907 the writer was able to separate from uranium minerals a previously unidentified radio-element which was intermediate between uranium and radium in the series of atomic transformations, and which by its own disintegration produced radium in readily measurable quantities. To this element the name "ionium" was given. It thus became possible to separate the ionium from a mineral containing a known amount of radium, and to determine the rate of growth of radium in this ionium. This is a measure of the rate of production of radium in the mineral and therefore a measure of the rate of disintegration of the radium.

The two diagrams (Figs. 1 and 2) will perhaps be useful in making the general conditions and method of procedure more easily understood to those without a technical knowledge of the subject. In the first (Fig. 1) the amount of uranium changing per year relative to the total

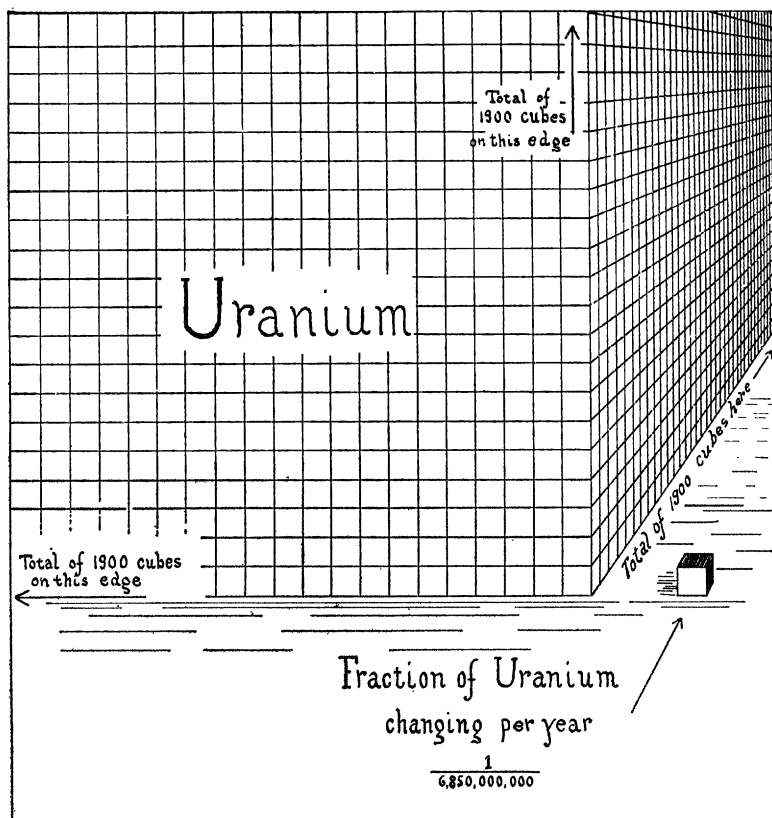


FIG. 1.

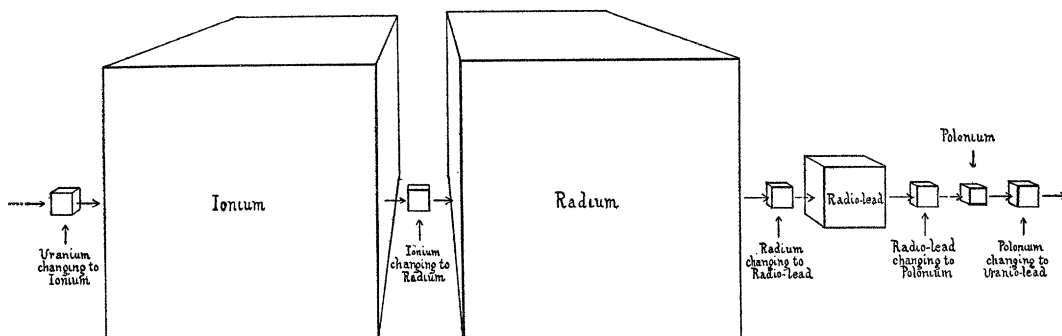


FIG. 2.

amount present is shown by two cubes whose volumes are proportional to the number of atoms involved in the transformation. In the second diagram (Fig. 2) the first cube on the left is supposed to be of the same size as the smaller cube in the first figure. Since the constant of change of

ionium is as yet undetermined, it has been assumed for convenience to be approximately the same as that of radium, and the amount of ionium in the mineral is therefore indicated as of the same order as the amount of radium. With this limitation, and omitting the slight complications in-

volved by the existence of branch products, like actinium, and products of a rapid rate of change, like the emanation and radium-A, the diagrams represent the general conditions and changes to be found in an old⁷ uranium mineral. The chief relation of interest shown by the diagram is that since the radium changing to radio-lead can not be determined experimentally with sufficient exactness, it is equally satisfactory and very much simpler to determine the ionium changing to radium and compare its quantity with the total amount of radium in the mineral. As a matter of fact the actual amounts of radium involved in these two quantities need not be known, it is only their relative values which are required, since the value of the disintegration constant is determined by the ratio of one of these to the other. In this respect the method is independent of any standard of purity of radium preparations, an advantage which is not possessed by other methods which have been used for attacking the problem. Thus, for example, the estimate of the half-value period of radium made by Rutherford and Geiger as a result of their experiments in 1908, had to be altered from 1,760 years to 1,690 years, when in 1912 the present international radium standard was adopted.

The results of a number of experiments conducted by the writer according to the method just outlined were published in 1908. In the most satisfactory of these experiments the mineral taken was a quantity of pure, primary North Carolina uraninite, almost free from secondary alteration products. About 40 grams of this material were used and the ionium was separated (with the thorium, which has identical chemical properties) by the ordinary analytical methods for the separation of thorium.

⁷ There are well-known examples of minerals too young for a state of equilibrium to have yet been reached between their radioactive constituents.

The growth of radium in this preparation of ionium was then measured over a period of 147 days, and a rate of change for radium corresponding to a half-value period of 1,990 years was obtained. The results of the other experiments were in fair agreement with this value, which was assumed to be the most probable one. It is interesting to point out that this estimate was made between the time of Rutherford's estimate of 2,600 years and Rutherford and Geiger's estimate of 1,760 years.

In view of the disagreement of the value obtained by the "growing" experiment with the value as calculated from Rutherford and Geiger's work, it was reasonable to suspect that in the "growing" experiments all of the ionium was not separated from the mineral. Such a suggestion was, in fact, made subsequently by Rutherford. A careful investigation of some of the conditions associated with the usual methods employed for the chemical separation of small proportions of thorium from large amounts of uranium, indicated that a complete separation of the thorium under such conditions was extremely uncertain if not altogether impossible. The chemical behavior of uranium and thorium are strikingly similar: in the case of the uranous (UO_2) salts the chemical analogy of the two elements is such a close one as to make any separation at all almost impossible. Since an incomplete separation of the ionium would lead to *too small* a production of radium in the growing experiments, under the assumption that the separation was complete the calculated half-value period of radium would receive too high a value. It was therefore highly desirable that the experiments should be repeated under conditions which would avoid any uncertainty, and which would give an altogether trustworthy value for the life of radium as determined by this method.

This work was undertaken in the university year 1913-14 in my laboratory by Miss Ellen Gleditsch, who came to this country from Norway for a year of study on a fellowship of the American Scandinavian Foundation. The work has been carried out in a very satisfactory manner and, after encountering a number of difficulties, she has quite recently completed her experiments at the University of Kristiania. A paper by her on the subject will appear in the January number of the *American Journal of Science*.

Miss Gleditsch carried out four separate operations, which may be briefly described as follows:

The *first* was with a specimen of very pure North Carolina uraninite weighing 110 grams and containing 2.46×10^{-5} gram of radium. The growth of radium from the ionium separated from this material gave a value for the constant of change of 3.7×10^{-4} (per year), which corresponds to a half-value period of 1,836 years.

The *second* was with a specimen of Norwegian uraninite known as Cleveite, weighing 180 grams and containing 3.2×10^{-5} gram of radium. The ionium separated from this material grew radium at a rate corresponding to a value for the constant of 3.9×10^{-4} and a half-value period of 1,780 years.

The *third* was with a specimen of Norwegian uraninite of the variety known as Bröggerite, weighing 200 grams and containing 4.1×10^{-4} gram of radium. In this experiment the radium grew at a rate corresponding to 4.2×10^{-4} for the value of the constant and indicated a half-value period of 1,640 years.

In the *fourth* experiment a specimen of very pure Bröggerite was used, weighing 100 grams and containing 2.1×10^{-5} gram of radium. The value obtained for the constant of change of radium was 4.1×10^{-4}

and the half-value period corresponds to approximately 1,670 years.

In this series of experiments the methods for effecting a complete separation of the ionium were progressively developed and improved. In the last two, the ones in which bröggerite was used, there were definite indications that a complete separation had been accomplished. Moreover, Miss Gleditsch also measured the amount of radium in one of my original ionium solutions in which the radium had been growing for a period of nearly seven years and found that the rate of growth had been constant throughout the entire interval. This fact disposes of the possible objection that the life of ionium is too short to give an accurate value for the constant of radium as determined by this method.

It is therefore apparent that the different methods which have been used for estimating the life of radium give results which are in excellent agreement with one another. This agreement increases the assurance with which the estimated values of certain important physical constants involved in the calculation can be accepted as approximating closely to the true values. As a matter of interest these constants will be mentioned.

Number of α -particles emitted per second by one gram of radium (element) $= 3.57 \times 10^{10}$.

The charge carried by a hydrogen ion in electrolysis $= 4.65 \times 10^{-10}$ E.S. units.

The number of atoms in one gram of hydrogen $= 6.2 \times 10^{23}$.

The mass of the hydrogen atom $= 1.61 \times 10^{-24}$ gram.

The number of molecules in one cubic centimeter of any gas at standard pressure and temperature $= 2.72 \times 10^{19}$.

The volume of the radium emanation in equilibrium with one gram of radium

=0.62 cu. mm. calculated, =0.63 cu. mm. found.

The rate of production of helium per year per gram of radium =163 cu. mm. calculated, =164 cu. mm. found.

B. B. BOLTWOOD

A SUGGESTED EXPLANATION OF "ORTHO-GENESIS" IN PLANTS

THE purpose of this paper is not to discuss what is called orthogenesis in plants in general, but to cite certain notable illustrations of it, and to suggest a possible explanation. There may be some difference of opinion as to the proper definition of orthogenesis, but it is used in this paper as standing for progressive evolution in a given direction, in contrast with more or less successful variations in several directions, involved in the theories of natural selection and mutation.

My thesis is not to prove that orthogenesis differs in kind from such explanations of evolution as natural selection or mutation, but that the persistent variation which results in what is called orthogenesis is in response to a persistent change in the conditions of living. It is an explanation of orthogenesis which contradicts its original meaning and makes it a physical rather than a vitalistic phenomenon.

Another prefatory statement should be made. The conclusions reached in this paper are not simply inferences from a series of observations, but are based chiefly upon the results of experimental work which indicates that the changes called for can be induced as responses to changed conditions.

The gymnosperms are unique among the great plant groups in the length of their available history, recorded in such a way that our knowledge of the group may be said to be fairly continuous. Other great groups are either relatively short-lived, or

their records, at least so far as our knowledge of them is concerned, are very discontinuous. As a consequence, many lines of advance among gymnosperms can be traced in unbroken series from the Devonian to the present time, involving structures that have been assumed to be beyond the influences of external conditions. I wish to call attention to four such lines of advance, and to draw certain conclusions which have some bearing upon evolutionary theory.

1. *The Egg*.—A remarkable series of progressive changes is recorded as one traces the development of the female sex organ (archegonium) from the most primitive gymnosperms to the most recent. The gradual change consists in the shifting of the time of appearance of the archegonium in the ontogeny of the gametophyte (the sexual individual). In the most primitive gymnosperms the archegonia appear at what may be called the full maturity of the gametophyte, just as they do in the prothallia of ordinary ferns. An unbroken series can be traced, representing an earlier and earlier appearance of archegonia in the ontogeny of the gametophyte, extending from full maturity to very early embryonic stages. In this ontogeny three stages may be roughly distinguished: (1) free nuclear division; (2) primary wall formation; (3) growth of tissue. It is toward the end of the third stage that archegonia appear in the most primitive gymnosperms; and the gymnosperms of to-day, whose archegonia are late in appearing, as the Cycads, are primitive in this feature, though they may be advanced in some others.

As one proceeds with the history of the group, it can be observed that the appearance of archegonia shifts back through the third stage, more and more tissue being developed after their appearance. Next they are observed forming at the second stage, that of primary wall formation. In