Flint Fracture and Flint Implements*

Some Experiments Made With a View to Solve an Important Archaeological Problem

By J. Reid Moir

THE correct determination of human flaking upon a flint has always been a vexed question with archaeologists. The difficulty arose apparently, in the first place, with the discovery of the Neolithic axes and arrow-heads, as the late Sir John Evans (*Ancient Stone Implements of Great Britain*, 2d edition, pp. 56 and 362) mentions that in certain old books these specimens are described respectively as "Thunder-bolts" and "Elf Arrows," clearly proving that a supernatural origin was accorded to them. Though we have no evidence that advanced archaeologists were in existence in those somewhat remote times, yet it seems feasible to suppose that even then there were some unable to accept the supernatural theory and who claimed that the flint axes and arrow-heads represented the work of human brains and hands. Be that as it may, the fact remains that no one has for many long years disputed the human origin of these particular flaked flints, which are now universally accepted as affording conclusive evidence of human intention.

But the condition of quiescence so long prevailing in prehistoric circles was rudely shattered by the discoveries of Boucher de Perthes in the valley of the Somme. About the year 1841 it became noised abroad that definite flint implements of man had been found in undisturbed river-gravels of this district, associated with the bones of extinct animals, and that these discoveries afforded evidence of the existence of man at the remote period when these animals existed, and prior to the time when the ancient gravels containing their remains were laid down. Immediately these discoveries were made known to the scientific world, archaeologists were plunged into a most violent and bitter controversy regarding the flaked flints discovered. The majority of authorities made haste to repudiate the claim of Boucher de Perthes, and asserted that the flints had been flaked by natural causes, and if not, then they had been made by the workmen at the pits, who had surreptitiously inserted them in the gravel from whence they were removed by the enthusiastic but misled archaeologist. The various and extraordinary arguments put forward against the human origin of the palæolithic implements discovered by Boucher de Perthes are fortunately preserved and can be studied in a book entitled *The Antiquity of Man*, consisting of papers selected chiefly from the *Transactions of The Victoria Institute* (Elliot Stock, 62, Paternoster Row, London, E.C.).

There were, however, a few men who, having seen and handled some of Boucher de Perthes' specimens, stated they were in agreement with his opinion as to their human origin.

But these opinions made little headway, and for many years the majority of archaeologists held to the belief that the flints found in the valley of the Somme had been flaked by some unknown natural forces.

Gradually, however, opinion began to change. Investigations carried out in river-gravels in this country and elsewhere established the fact of the occurrence in these deposits of similar flaked flints to those found by Boucher de Perthes, and at length, after much searching of heart on the part of the archaeological world, palæolithic man came into his own. With the general acceptance of the palæoliths, a condition of comparative tranquility once more descended upon the question of man's antiquity; and though there were some bold spirits who held that an elaborately flaked palæolithic implement could not, in the nature of things, represent man's first effort in the fashioning of flint, and that in consequence his earlier efforts would probably be found in deposits more ancient than the palæolithic river-gravels, yet these were but "voices crying in the wilderness," and little heed was paid to them. The mutterings of the coming storm, which was to sweep away the orthodox limits set to the antiquity of the human race, were first heard from Ightham in Kent, where Mr. Benjamin Harrison, having found palæolithic implements in the oldest river-gravels of his district, began to investigate still more ancient deposits situated on the high plateau of Kent. These investigations resulted in the discovery of a series of rough flints exhibiting flaking along one or other of their edges. This discovery and the announce-

ment that several archaeologists, including the late Sir Joseph Prestwich, regarded these specimens as affording evidence of the former existence of a race of beings much more ancient than the makers of the palæoliths, initiated another period of unrest, and for the third time the archaeological world was grievously disturbed, and the usual division of forces took place.

Fig. 1

Those who supported Mr. Harrison asserted that the flaked flints he had discovered were undoubtedly the work of man, and represented the type of implement which they would expect to find in such an ancient deposit; while his opponents contended that the flaking on the flints was undoubtedly due to natural pressure or percussion and therefore not indicative of human handiwork. Since the publication of Mr. Harrison's dis-

Fig. 2

Fig. 3

coveries, a number of pre-palæolithic implements which seem to link up the primitive Kentian implements with the later palæoliths have been found in various Pliocene and early Pleistocene deposits in East Anglia, and these specimens, though accepted by an ever-widening circle of prehistorians, have in their turn been regarded by certain investigators as representing the work of nature. It will thus be seen that the science of prehistory has

Fig. 4

Fig. 5

advanced only by slow and somewhat painful steps, and it may be of interest to inquire into the causes of this tardy progress.

The prolonged and heated controversies which have inevitably followed any new discoveries of flaked flints tending to extend the antiquity of the human race indicate clearly that the acceptance or non-acceptance of these specimens has not been actuated by any clear

Fig. 6

Fig. 7

Fig. 8

and definite scientific knowledge of the differences exhibited by flints flaked by man or by the unguided forces of nature.

If such knowledge had existed the controversies which have raged round the neoliths and palæoliths, and which are now being conducted in regard to the pre-palæoliths, would either have not occurred at all or, if commenced, would have been of a less prolonged and heated character. But in the absence of definite scientific data, other and less satisfactory means for arriving at a decision had to be employed. A prehistorian expressed himself to be in favor of accepting or not accepting any given flaked flint, simply by reason of his preconceived views and personal prejudices, and the majority of investigators are still swayed by such unscientific influences. In the long run what is known as "common sense" prevailed in regard to the neoliths and palæoliths, and there seems but little doubt that the pre-palæoliths will also be finally accepted on the same grounds; but the processes of common sense are slow and not necessarily scientifically adequate, and other and more satisfactory means of determining whether a flint has been flaked by man or by nature ought to be available. It appears that the only method likely to lead to definite and satisfactory results would be to conduct a series of experiments in which flints would be subjected to the effects of fortuitous percussion, and pressure, and to critically compare the fractures so produced with others caused by a hammer-stone held in the hand, and used in the manner in which ancient man probably used it. It might be urged that it is impossible to simulate experimentally the effect of natural forces upon flints, but on the other hand it seems reasonable to suppose that fortuitous blows and pressure do not differ fundamentally, whether brought into action by nature or in experiments conducted by a human being. The "mightiness" of natural forces cannot certainly be imitated experimentally, but it has been ascertained that flints will only stand, without disintegration, a certain amount (by no means very large and easily equalled in experiments) of pressure and force blows, and that in consequence when the natural forces in operation are too mighty, the flint must be shattered and reduced to fragments. The author concludes, therefore, that the experiments he has conducted in the flaking of flint are of value in this discussion, and though further researches may be necessary before the problems connected with human and natural flaking are finally and completely solved, yet he feels that the results already obtained may help to dispel some of the doubts and difficulties, and thus place the question of the antiquity of man upon a firmer and more scientific foundation. It is proposed in the first place to tabulate and describe some of the details of flint fracture, to which reference will be made in the description of the experiments carried out. The nature of the experiments in which flints were subjected to fortuitous percussion, and the conclusion arising therefrom, will then be described, and finally in a like manner the experiments in which fortuitous pressure was employed¹

DETAILS OF FLINT FRACTURE

Striking Platforms.—When an ordinary nodule of flint is selected with a view to produce an implement from it by flaking, it is necessary, owing to the difficulty of removing flakes from a rounded surface, to break off a portion of the nodule, so that a flat surface is produced, upon which blows removing flakes can be struck with precision. The flakes which are removed will exhibit, in the immediate vicinity of the point where the blows fell which detached them (known as "the point of impact"), a portion of this flat surface, and this is termed the "Striking Platform."

The Cone of Percussion.—When a certain type of blow is delivered in the center of a flat surface of sound flint, and the portions of this surface surrounding the point of impact tapped gently with a hammer, these portions will generally fall away, revealing a more or less perfect cone of flint. This is known as "the cone of percussion," and is due to an inherent

¹A complete series of the flints fractured experimentally can be seen and examined in the Department of Ethnography at the British Museum (Bloomsbury).

*From *Science Progress*.

property in the flint itself to fracture in this manner. This peculiar method of fracture is also found to be present in some other substances, such as glass, obsidian, etc.

Positive and Negative Cones of Percussion.—When a flake exhibits just below the point of impact a conical protuberance, and the block from which it was struck a corresponding hollow or depression, the former is known as the "positive," and the latter as the "negative" cone of percussion. The detached flake may as often exhibit the negative cone and the parent block the positive, but that does not affect the validity of the description given above.

Éraillures.—If a number of flakes removed by blows are examined it will be seen that many of them exhibit on their bulbar surfaces (that is, the surfaces upon which the positive cone of percussion is visible) a scar, where a small and isolated flake has become detached. This small flake is apparently removed simultaneously with the detachment of the larger flake, but the exact reason for its removal does not appear up to the present to have been satisfactorily explained. This éraillure or "mark" is sometimes seen upon flakes removed by pressure (Fig. 1).

Ripple Marks or Conchoidal Rippings.—These ripple marks, which are often seen upon flakes and implements of human manufacture, and also upon flints flaked by fortuitous means (they appear to be more prominent and frequent upon the latter), are due to the tendency of flint under certain conditions to fracture in a conchoidal or shell-like manner. It seems that these conchoidal rippings are somewhat analogous to the ripples produced upon a surface of water when some heavy object is thrown on to it. The water is "jarred" by the impact and ripples produced, and it may be that certain blows jar the flint and so produce rippings on the fractured surface. Conchoidal rippings are sometimes formed when a flint is fractured by pressure.

Fissures.—If a series of flakes is examined it will be noticed that their surfaces exhibit a series of fissures of varying length and width, which radiate from the point of impact. Some of these markings are easily visible to the naked eye, while others require a strong lens to discern them. It seems that their presence can be accounted for by supposing that when the flake is removed the flint is in a sense torn and that these markings represent the tears produced by the cleaving effect of the blow. Fissures are sometimes produced upon a flint which has been fractured by pressure.

Truncated Flakes.—When the manufacture of a flint implement is commenced, large flakes are removed from the nodule of flint, which in the further process of flaking get reduced in size or truncated. Thus, it is possible to observe the remains of flakes which have been thus reduced in size on various portions of the surfaces of many implements. These flakes are termed "truncated flakes."

Opposing Cones of Percussion.—Under certain conditions a flake removed by pressure may exhibit a well-marked cone of percussion at each end. This is due to the flint having been squeezed between two hard objects, and the cones represent the points at which, respectively, pressure and resistance acted. These cones are known as "opposing cones of percussion."

Experiments with Flints Subjected to Fortuitous Percussion.—Seven or eight flint nodules of various shapes and sizes were placed in a sack and violently shaken about for some little time, and in such a manner that the flints were in almost continual collision. It was found that only the stones of a certain shape—and conforming more or less to a wedge in form (the thin edge of the wedge being usually attacked)—were flaked in this process. The rounded flints in the sack were not flaked, but escaped with contusion of their surfaces only. A careful examination of the flaking produced by these fortuitous means, and a comparison of it with that due to human blows, appears to show that there is a fundamental difference between the two methods of fracture. This difference is, primarily, the angles at which the flakes have been removed. The method adopted to ascertain the direction in which any given flake has been removed from a flint is as follows: A line is drawn down the center of the flake and at right-angles to the ripple-marks (these in many cases require locating with a good lens). Then two other lines are drawn one on each side of the central line, and also at right-angles to the ripple-marks as they curve upwards. The three lines are then continued to their point of junction, which point indicates the area upon which fell the blow that detached the flake (Fig. 2).²

The correctness of this method can be checked by observing the direction of the small fissures or "splits," which gives an additional indication of the angle at

which the flake was removed (Fig. 3). Careful outline drawings were then made of the flints flaked in the sack experiment, and also of those shaped by human blows.

Each flake area was accurately drawn and the direction of the blows responsible for the flaking indicated by arrows.

When this was done it was seen that the flints from the sack had had their flakes removed at divergent angles to the edge of the stone (Fig. 4), while those fractured by human blows had had them removed at a constant angle (Fig. 5). It does not appear possible that fortuitous haphazard blows would remove flakes at a constant angle to the edge of any flint, neither does it seem reasonable to suppose that any human flaker would go to the trouble of making a sharp edge by means of divergent blows, as the result would be much less satisfactory than one produced by the normal method. It appears then that we have here a good and simple means of distinguishing between the work of man and of nature.³

A further examination of the specimens fractured in the sack experiment demonstrated that the flakes which

Fig. 9

had been removed differed in appearance from those due to human blows, this difference being—

- (a) The "squatness" of the fortuitous flakes as compared with those removed by human agency (Fig. 6).
- (b) The fact that the former had cut deeper into the flint, causing a step or ledge to appear at the point of their final separation from the parent block (Fig. 6); and
- (c) The occurrence of numerous prominent ripple-marks upon nearly all of the flakes produced by fortuitous blows, as compared with their comparative scarcity upon the "human" flakes (Fig. 6).

The explanation of these differences may be explained as follows:

In intentional human-flaking, a flat striking platform is produced, and blows are delivered on one or the other side of this platform, and in such a manner that the flakes removed are generally the reverse of squat, being of greater length than width, and thinnest at the point of their final separation from the parent block (Fig. 7). Such flakes are also generally free from prominent ripple-marks, due, in the author's opinion, to the fact

Hollow Scraper
Formed by Pressure

Fig. 10

Hollow Scraper
Formed by Human
Blows

Fig. 11

that the blows which detached them were delivered almost vertically to the striking platform. In flaking produced by fortuitous percussion the great majority of the blows fall, not upon the side of the edge of the flint, but almost directly upon the edge itself, and it is known that blows so delivered remove flakes squat in form and productive of a step or ledge at the point of their final separation from the parent block.

It has been ascertained that consistently to remove flakes exhibiting prominent ripple-marks upon their surfaces, it is necessary to strike the flint an oblique blow. As has already been mentioned, a blow delivered vertically to a flint surface will remove a flake almost devoid of ripple-marks, and it is supposed that such a blow has less jarring effect upon the flint, and consequently produces less rippling of the fractured surface, than one delivered in an oblique direction. The explanation of the frequent occurrence of prominent ripple-marks upon fortuitous flakes, and their com-

parative scarcity upon those produced by human blows may be as follows. It may be assumed that there are 180 angles at which a flake may be removed from any given length of edge, but it is only more or less vertical blows, delivered at a few of the higher angles, that will remove flakes exhibiting insignificant ripple-marks. Thus in fortuitous haphazard blows the chances are that the greater number of blows will be delivered at the lower angles, which are oblique to the edge of the flint, and which, as has been shown, remove flakes exhibiting prominent ripple-marks upon their surfaces. It seems, then, that in the shape of the flakes, their character at the point of final separation from the parent block, and the presence or absence of prominent ripple-marks upon their surfaces, afford further criteria for determining whether any particular series of flints has been flaked by man or by nature.⁴

It was noticed that the edges of all the wedge-shaped stones fractured in the sack experiment showed a tendency to assume a sinuous outline due to blows having fallen upon either side of these edges.

The majority of the stones exhibited this sinuosity in an incomplete manner, but one specimen showed a markedly sinuous edge $4\frac{1}{2}$ inches in length. A critical examination (based upon the tests already described in this paper) of the flaking to be seen upon this flint demonstrated clearly its "natural" origin, but another characteristic of the flaking was detected which seems to still further differentiate it from the work of man. It was found that the edge of this flint, though only $4\frac{1}{2}$ inches in length, exhibited 17 truncated flakes (Fig. 8).

On the other hand an examination of various neolithic and palæolithic implements with sinuous edges demonstrated that the average number of truncated flakes on an edge seven inches long is only six.

The explanation of the greater number of truncated flakes upon an edge produced by fortuitous blows as compared with one formed by human agency may be that in the former case the rain of blows is almost continuous and the flint is continually being refaked and truncated flakes formed. It is, however, somewhat difficult to imagine any human flaker being so inept at his work as to be compelled to form 17 truncated flakes in the production of an edge $4\frac{1}{2}$ inches in length.

The author considers therefore that the proportion of truncated flakes upon a flint may furnish a further criterion for the determination of human as against natural flaking.⁵ Various other experiments in fortuitous percussion, such as the hurling of a flint upon others lying on the ground, were carried out, and the results found to be similar to those obtained in the sack experiment.

Experiments with Flints Subjected to the Effects of Fortuitous Pressure.—Experiments were conducted with an adapted letterpress, and a differential screw press, with which considerable pressure was obtained.

It was found that when a suitably shaped stone was placed upon the floor of the press, and another smaller stone placed on the top of it and near its edge, a flake could be removed from the lowermost flint by forcing the ram of the press down upon the uppermost. Such flakes exhibited "opposing cones of percussion" (Fig. 9), due to the pressure upon the upper portion of the flint being equalled by the resistance afforded by the floor of the press. The presence of a cone of percussion at each end of these flakes differentiates them from those detached by human blows, on which only one bulb of percussion is visible. When a duster was placed upon the floor of the press so that the flint rested upon a soft basis, it was found that a flake could be removed, but only with the exertion of considerably more pressure than was needed in the former case. It was noticed also that such flakes exhibited only one cone of percussion, at the point where the small upper stone impinged upon the flint, as in the case of flakes removed by human blows. The majority of these cones of percussion, however, are flat and appear to be only partly developed, and in this respect they differ from the majority of cones of percussion produced by human blows. Pressure flakes, too, seldom exhibit well-marked éraillures, while the fissures and ripple-marks, if present, are of a different order from those present upon humanly-struck flints.

These differences are readily observable when a representative series of each kind of flake is put out for examination, and appear to be due to the different nature of the two fracturing forces. The human flaker is able to guide the line of fracture, within limits, in any direction he pleases, while in the case of pressure and flint is squeezed until it can stand the strain no longer,

²In arriving at a decision it is necessary to examine not less than twenty or thirty specimens; a lesser quantity examined might lead to erroneous conclusions.

³In the re-sharpening of many neolithic scrapers and Moustier (palæolithic) racloirs a number of small truncated flakes were produced, but these are not of the same order, or comparable to the truncated flakes described above.

⁴This method is adopted only when dealing with a truncated flake; when the actual bulbar cavity is present such a method is not needed.

⁵The author has examined and made flaking diagrams of a large number of flints found upon sea-beaches, and the flakes removed from these specimens by fortuitous battering are at markedly divergent angles.

and finally fractures along the line of least resistance. Future investigations may perhaps modify this explanation, but at present the author regards it as affording a likely reason for the difference, which undoubtedly exists, between flakes removed by pressure and those detached by human blows. Experiments were also conducted in which a hard resistant stone was placed upon the floor of the press, and another with a sharp edge placed against it in such a manner that the descending ram would cause this sharp edge to move under pressure over the surface of the other specimen. By this means a flaked hollow was produced in the sharp edge of the uppermost stone.⁶ Several of these hollows were produced and compared with others formed by human blows. This comparison demonstrated that there was a marked difference in the appearance of the two series of hollows.

⁶A similar result can be obtained by placing a sharp-edged flint upon another stone lying on the ground and turning one's heel round on them, under pressure.

It appears that when the sharp-edged flint is being moved under pressure over the surface of the underlying stone very thin flakes or scalings are removed. This is due apparently to the fact that the pressure is able only to attack a very narrow area of the edge of the moving stone, and in consequence only very thin flakes are removed.

In flaking produced by human blows, it is impossible to strike so near to the edge of the flint as to remove flakes as thin as those detached by pressure, and a hollow flaked by the former means always appears, and is, more uneven and less finished than one produced by pressure.

The pressure hollows appear "smoother" than those of the other kind, because, the flakes being thinner, the ridges intervening between them are less prominent (Fig. 10).

In the case of the human blows, which cut more deeply into the flint, and remove thicker flakes,

these ridges are correspondingly more marked (Fig. 11).⁷

The foregoing experiments in fortuitous percussion and pressure have been repeated many times and the results found to be uniform in each case.⁸ It is to be hoped that prehistorians will be able in future to give more attention to the question of the natural and artificial fracture of flint as affording the only means of arriving at satisfactory conclusions regarding the "humanity" or otherwise of the specimens which are the subject of their study. Mere speculations as to the capabilities of man or nature to flake flints in the remote past are, in the author's opinion, of little value.

⁷The author has lately examined the Eocene flint-bed resting upon the chalk at Bramford, near Ipswich, where many specimens have been fractured by natural pressure in the bed in which they now lie. This examination has shown that the fracture Eocene flints reproduce in a marked degree the characteristics of those fractured by pressure in the experiments described. See *Proc. Prehis. Soc. of E. Anglia*, vol. i, Part IV, pp. 397-404.

⁸Further experiments are in progress, and if the results obtained throw any further light upon the problems discussed in this paper, these will in due course be published.

An Attempt to Separate the Isotopic Forms of Lead by Fractional Crystallization*

By Theodore W. Richards and Norris F. Hall

ALTHOUGH the complete inseparability of isotopes by chemical means has been frequently asserted, the evidence on which this assertion is based has always seemed insufficient. The methods used have been fractional crystallization and precipitation, but these processes have seldom been carried out more than ten times in a particular case, and frequently six or seven crystallizations have been thought a sufficiently thorough test of inseparability. A search of the literature revealed only one investigation that of Radiothorium and Thorium by McCoy and Ross (*Journ. Am. Chem. Soc.*, 1907, xxix, 1709), where as many as 100 repetitions of a given process had been made. Thorium nitrate was dissolved in nitric acid and reprecipitated 100 times with excess of ammonia, but without change in the relative concentration of the isotopes. Precipitation with hydrogen peroxide (40 times) was also without result.

It seemed worth while, therefore, to apply to the important generalization of Fajans, Russell, Fleck, and Soddy, a more searching test, carrying the fractionation further, and using as a criterion of success not only the measurement of radioactivity but also the determination of atomic weight.

The lead from Australian carnotite, with which the kindness of Mr. S. Radeliff and Mr. E. R. Bubb had supplied us, and which had been used in previous atomic weight and density determinations in the Wolcott Gibbs Memorial Laboratory (Richards and Wadsworth, *Journ. Am. Chem. Soc.*, 1916, xxxviii., 1659), seemed well suited to our purpose, since its low atomic weight indicated a composition of three or four parts of the isotope (usually assumed to be radium G) to one of lead; and its β -ray activity showed that it contained sufficient radium D to serve as a basis for ionization tests of the possible separation of this isotope from the inactive varieties. The excellent work on radium D and lead by Paneth and von Hevesy (*Sitz. d. k. Akad. Wiss. Wien*, 1913, 122, IIa, 993) had shown, indeed, that no one of the many and diverse chemical methods applied by them had effected any separation of these substances, but even here no single operation had been repeated often enough to detect the effect of exceedingly small differences in chemical or physical properties.

PRELIMINARY PREPARATIONS OF MATERIAL

About one kilogram of the radioactive lead was taken for this work. This included much of the purified samples A, B, and C of the atomic weight work already referred to, some of which had not been used, and some of which had been recovered from the completed analyses. The samples of metallic lead which had been used in spectrographic tests and density determinations were also added, as well as all the mother-liquors remaining from the preparation of the purified samples. Much of this material was already in the form of nitrate, some in the form of chloride, and some was metallic. The chlorides were converted into nitrates by repeated evaporation with pure nitric acid, and the metal was dissolved in the same acid. All the nitrates were then combined, dissolved in the least possible quantity of boiling water, and precipitated by the addition of large quantities of pure concentrated nitric acid and cooling in an ice bath. The crystals were separated from the mother-liquor with a porcelain funnel, washed with concentrated nitric acid, and the washings and mother-

liquor united. The strongly acid mother-liquors were evaporated to fuming with sulphuric acid, and the sulphates almost completely precipitated by dilution. The sulphates were converted to carbonates by boiling with sodium carbonate, washed nearly free from sodium, and dissolved in nitric acid. The small amount of nitrates thus obtained was evaporated to dryness and added to the bulk of the nitrates. No attempt was made to purify further at this stage, because the unavoidable introduction of much silica during the fractionation (as well as possible accidental contamination) made necessary the thorough purification to which the samples were subjected just before the final tests.

FRACTIONAL CRYSTALLIZATION

The method of crystallizing the nitrate by cooling the boiling nearly saturated aqueous solution was adopted after preliminary experiments on the chlorate and perchlorate of lead had indicated the superiority of the nitrate to these more soluble salts for the work in hand.

This superiority consists in the well-known habit of the nitrate of forming compact, large prisms on crystallization from water solution, which can be very completely freed from mother-liquor merely by drainage, as well as in its slight tendency to supersaturation, and the consequent rapidity with which crystal-liquid equilibrium is attained.

The whole of the nitrate was then dissolved in distilled water and evaporated until a crust began to form. This was called Fraction 1. After cooling to room temperature the mother-liquor was poured off and formed Fraction 2. To the crystals from Fraction 1 was added sufficient water to dissolve them, and the solution was called Fraction 3.

Fractions 2 and 3 were evaporated to saturation and cooled as Fraction 1 had been, the liquor from 2 forming Fraction 4. Fraction 5 was made by adding the liquor from 3 to the crystals from 2, and water was added to the crystals from 2 to form Fraction 6. This process was continued as shown in the diagram until, as occurred first with Fraction 16, one of the end fractions became too small to work with conveniently, and was consequently added entire to the mother-liquor from Fraction 17 to form Fraction 22. This expedient was resorted to wherever the end fractions became inconveniently small. In all, 75 courses or tiers of fractions were obtained, most of the last of which contained 18 fractions to a tier. The number of the last fraction was 1006.

The crystallization was carried out entirely in porcelain, at first in large evaporating dishes, then, as the fractions became smaller, in casseroles covered with watch glasses. The hot casseroles were usually set in evaporating dishes full of cold water, which was frequently changed to hasten cooling. The fractionation was stopped after about 1000 repetitions because, since an accuracy of 2 per cent was hoped for in the final tests, 1,000 fractionations would show whether a difference in solubility amounting to one part in 50,000 for a single crystallization existed or not. In other words, if the end fractions tested at this point showed no difference in the relative concentrations of their components, within the expected error of measurement of 2 per cent we should have proof that no complete separation could be attained by this method in less than fifty thousand fractionations.

Through the courtesy of Prof. Gregory P. Baxter, the

facilities of the T. Jefferson Coolidge Memorial Laboratory were extended to one of us during the summer of 1916, when this part of the work was carried out. Thanks are due to Prof. Baxter, not only for this kindness, but also for many helpful suggestions which his experience with rare-earth separations made doubly valuable.

Repeatedly the advantage of centrifugal separation of mother-liquor from crystals has been demonstrated in cases of ordinary crystallization where the crystals and mother-liquor are very different in composition. It is possible to show, mathematically, however, that where there is no such great difference the advantages of centrifugal draining is very much less, and in the present case it was found by this calculation that the gain in effectiveness of separation was less than the disadvantage caused by the increased time necessary for the centrifugal treatment of the thousand precipitates. Making the reasonable assumption that the difference in solubility in this case is probably not greater than 2×10^{-5} the relative advantage of centrifugal over gravitational drainage is seen to be certainly not greater than 1.5:1, so that it would be hopeless to try to increase the rapidity of separation by this means. Even under the best conditions with multiple-basket power centrifugals which could be stopped almost instantly, the time required would certainly be more than half again as much as for mere gravitational drainage. Therefore, in the present case, centrifugal draining was not adopted. Baxter, in his crystallizations of praseodymium and neodymium compounds, had come to the same conclusion from practical experience. A note discussing this matter mathematically will soon be published by one of us.

PURIFICATION

Because of the smallness of the end fractions it was necessary to unite several to obtain a sufficient quantity of material for purification.

(NOTE.—This action nullified the effect of 19 crystallizations on the more soluble end and 83 crystallizations on the less soluble end. Thus the total number of effective crystallizations was 904. The mother-liquor on the less soluble end resulted from an average of 71 tiers of successive products, and on the more soluble end from an average of 65 tiers. It is to be regretted that this reduction in the efficiency of the process was rendered necessary by the small yields at the two ends of the line, but enough crystallizations remained effective to test the question at issue, at least in a preliminary way.)

From the mother-liquor, or "more soluble" end Fractions 989-992 were taken, united and called Sample A. Sample B was made up of Fractions 990-1006 from the crystal, or "less soluble" end. From this point on the two samples were treated in an identical manner. Chiefly to remove the large amount of silica which had dissolved from the casseroles during the fractionation, the crystals were first electrolyzed in warm concentrated solution between platinum wires. The well-formed flat leaflets of lead were removed from time to time with a glass rod, rinsed with a stream from a wash-bottle, and preserved in quartz dishes until all the metal had been deposited from the solution. The crystals were further washed from time to time, and all washings returned to the electrolysis bath, which was gradually reduced in volume as the concentration of lead became smaller. The lead crystals were then dissolved in a nitric acid

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