

ART. XXVIII.—*New Points on the Origin of Dolomite* :* by
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Historical Review.

THE problem of the origin of dolomite has long occupied the attention of geologists and many theories have been advanced for its formation, but no one of these theories has been widely accepted. Von Buch (1)† was the first to seriously attempt to explain the formation of the rock. As early as 1822 in his writings on the dolomite of the Tyrol, he ascribed its origin to the action of volcanic vapors, rich in magnesia, on limestone, and there was some basis for this belief, for the rocks are there penetrated by augite-porphry. Frapolli (2) and Durocher (3) later expressed similar views upon the origin of the rock, and Favre (4), basing his supposition upon the conditions of the experimental production of dolomite by Marignac, concluded that the dolomite of the Tyrol was formed by the alteration of limestone beneath the sea at a temperature of 200° C. and at a pressure of 15 atmospheres, corresponding to a depth of 150 to 200 meters, by magnesium compounds furnished by the action of sulphurous and hydrochloric acids of volcanic origin on the lava of submarine melaphyr eruptions.

In 1834 Collegno (5) pointed out the frequent association of gypsum and dolomite in the St. Gothard region and regarded them both as transformation products resulting from the action of magnesium sulphate in surface waters or limestone. Morlot (6) also favored such a theory of origin.

As early as 1836 Beaumont (7) ascribed the origin of dolomite to the alteration of limestone by circulating solutions of magnesium bicarbonate and, assuming that the replacement was molecular, he calculated that the change should be accompanied by a decrease in volume of the original rock to the extent of about 12.1 per cent. Actual porosity determinations by Morlot (8) on a dolomite sample from the Alps later seemed to confirm this prediction.

In 1843, A. W. Jackson (9) suggested that ascending spring-water bearing magnesium bicarbonate might effect the change. Nauck, Haussmann, Bischof, Zirkel and others, however, subscribed to the view that ordinary circulating ground water bearing magnesium bicarbonate had attacked the limestone.

* The present article is based on a more extended paper which constitutes a portion of volume xxv of the Iowa Geological Survey. The reader is referred to this report for details.

† For numbered references to the literature, see the list at the end of this article.

Van Hise (10) also attaches much importance to dolomitization after the limestones emerge from the sea.

In 1846 Green (11) offered the suggestion that some dolomitic limestones might be formed by the decomposition of olivine sand incorporated in the original limestone and the recombination of the magnesia with the lime. He calls attention to the fact that olivine sand, derived from the action of the waves on lava, constitutes an important constituent of the coral-reef rock about the borders of the Hawaiian Islands and regards this as significant.

Dana (12) in 1843, attempting to account for the dolomite of the coral island of Metia, supposed that it had been formed by the action of magnesium salts of heated sea water on limestone. Twenty-nine years (13) later he expressed the view that the same dolomite had been formed in sea water at ordinary temperatures but perhaps in a contracting lagoon where magnesium and other salts were in a concentrated state. Sorby (14) likewise favored the theory of marine alteration and the same origin has been urged, either for dolomites in general or in special instances, by Von Richthofen, Doelter and Hoernes, Hoppe-Seyler, Mojsisovics, Murray, Skeats, and F. W. Pfaff. In support of this theory are also the observations of Weller (15) who, from a faunal study of the Galena and Niagara dolomites of the Upper Mississippi Valley, concludes that they were deposited originally as limestones and later metamorphosed. More recently, Blackwelder (16) has also advocated the replacement theory for the origin of the Big-horn dolomite of Wyoming, but owing to the very slight porosity of this rock he is led to suggest that the alteration proceeded contemporaneously with its deposition rather than subsequent to its consolidation.

F. W. Pfaff (17) believes that the alteration takes place at considerable depth and in concentrated seas, but Phillipi (18) vigorously controverts this view since he has good evidence that dolomitization may proceed in the open sea and at shallow depths. Skeats' (19) studies of the coral reefs of the Southern Pacific also seem to show that concentration and pressure are not important factors. On the other hand, both Nadson and Walther (20) have suggested that bacteria may play an important part in the alteration.

Still other geologists have supported the theory that dolomite represents a direct chemical precipitate from the ocean. Boné (21) as early as 1831 advocated this method of origin. Bertrand-Geslin (22) and Coquand (23) were also early supporters of this view. That dolomite can be formed as a chemical precipitate is pointed out by Zirkel (24) who shows that the occurrence of crystals of dolomite in veins and druses indi-

cates its possible chemical deposition on a larger scale in nature. Fournet (25) regarded the dolomite beds interstratified with limestone in the Tyrol as original precipitates. His studies showed that the volcanic theory of Von Buch was no longer tenable. Others who have advocated the primary precipitation theory in one form or another are Loretz, Forchhammer, Hunt, Vogt, Daly, Linck, and Sness.

As to the nature and cause of the reactions which have been supposed to give rise to the chemical precipitation of dolomite, there have been differences of opinion. Forchhammer (26) attributed the reaction to the action of calcium carbonate of spring water on the magnesium salts of the sea, while Hunt, (27) basing his views on experimental evidence, regarded dolomite as the product of the action of sodium bicarbonate on the magnesium chloride and magnesium sulphate of the sea. Linck (28) and Daly (29), on the other hand, emphasize the importance of ammonium carbonate furnished by decaying organisms on the sea bottom as the precipitating agent.

Still another primary theory is that introduced by Lesler (30) to account for certain dolomitic layers in the "Calcareous" limestone near Harrisburg, Pa. These he believed to represent ordinary mechanical sediments which were deposited at the time the limestone was laid down. The clastic theory has been adopted more recently by Phillipi (31), who regards certain impure dolomites of the Muschelkalk of Germany as mechanical deposits possibly derived from the residuum of limestones low in magnesia. Grabau (32) has concluded that certain impure dolomitic limestones and waterlimes of the Salinan and Monroan series have had a similar origin.

An entirely different theory of origin is that which was introduced by Grandjean (33) in 1844, to explain the production of the dolomites of the Lahn district. He assumed that by the atmospheric leaching of the lime from an original limestone of low magnesia content, a true dolomite might in time result. Both Bischof and Hardman later demonstrated the plausibility of this theory experimentally, and Hardman (34) immediately accepted it to explain the origin of the Carboniferous dolomites of Ireland. In 1895 Hall and Sardeson (35) applied the same theory in interpreting the history of the Lower Magnesian series of the Upper Mississippi Valley.

Högbom (36) on the other hand, regards surface leaching as of minor importance and emphasizes the effect of marine leaching. He has proven the reality of this process, on a small scale at least, in the modern seas and concludes that some dolomites of former periods may have been formed in this manner. Judd (37) is of the opinion that the weakly dolomitic portions of the atoll of Funafuti may be explained upon the basis of

this theory but regards the magnesia content of the more highly dolomitic portions as having been enriched by reaction with the magnesia of sea water.

Experimental Evidence.

On the experimental production of dolomite there is a voluminous literature. This has been well summarized by F. W. Pfaff (38), and later by Steidtmann (39). Dolomite has been frequently prepared artificially under conditions of high temperature or high pressure, or both, but it has been produced in the laboratory at ordinary temperatures and pressures only in rare instances, and then in minute amounts and under conditions which doubtfully operate in nature, at least on a large scale. It must be conceded then that these experiments furnish little evidence as to the actual conditions obtaining when extensive beds of dolomite are formed naturally. For the purpose of obtaining more accurate data on this point, a series of experiments was begun at ordinary temperatures and pressures early in 1912. In this series it was attempted to simulate natural conditions as near as they could be estimated, and to obtain some quantitative measurement of the effect of time and of concentration in the production of dolomite. In one set of experiments it was attempted to reproduce the conditions which exist in nature when limestone is altered to dolomite beneath the sea by solutions bearing magnesia. In these the effect of solutions of known concentration of $MgCl_2$ and $MgSO_4$, and of mixtures of the salts, both with and without the presence of $NaCl$, on powdered aragonite was tried. The concentration of the magnesium solutions used ranged from two to ten times the concentration of the magnesia in sea water. After a period of six months, residues from the experiments were thoroughly tested for dolomite. The results were entirely negative. No trace of dolomite could be found. Careful re-examination of the residues after a period of nearly three years still gave the same result. The analyses showed that the $CaCO_3$ had reacted slightly with the solutions, but no $MgCO_3$ had been deposited. Apparently the soluble trihydrate of $MgCO_3$ had been formed. It then appears that dolomite cannot be prepared artificially under these conditions.

In a second set of experiments it was attempted to obtain dolomite as a direct chemical precipitate at ordinary temperatures and pressures. First solutions of the bicarbonates of calcium and magnesium, after being standardized, were mixed in molecular equivalent proportions so as to give the same ratio of $CaCO_3$ to $MgCO_3$ as exists in normal dolomite. The solution was then allowed to evaporate spontaneously during a

period of one month. It was noted that the carbonates came down separately with the CaCO_3 much in advance of the MgCO_3 . The precipitate then contained only the mixed carbonates—no dolomite was formed. Scheerer (40) previously obtained the same results in a similar experiment. Negative results were still obtained when a solution prepared as above was inoculated with a crystal of dolomite and allowed to evaporate. Nor could the double carbonate be prepared upon evaporating spontaneously a solution of the two carbonates obtained by the action of carbonated waters on normal dolomite even when a dolomite crystal was introduced and a concentrated solution of sodium chloride and magnesium salts was added.

The experimental evidence so far obtained, therefore, does not suggest the circumstances under which large masses of dolomite can be formed in nature under ordinary conditions either by the alteration of limestone or by chemical precipitation. It is to be regretted that a careful study of the process of dolomitization where it is going on in the seas to-day has never been made. Such a study would doubtless throw much valuable light on the problem. It may well be that bacteria play an important rôle in the production of dolomite as suggested by Nadson.

Field Evidence.

Realizing the importance of careful field studies of dolomitic formations in interpreting the conditions of their origin, the writer undertook a study of the dolomites of the Upper Mississippi Valley under the auspices of the Iowa Geological Survey during the field season of 1912. More recently a grant from the Esther Herrman Research Fund of the New York Academy of Sciences has made possible much more extensive field studies in the eastern and central states. Dolomites ranging in age from the Cambrian to the Mississippian have now been examined and many samples collected for detailed chemical and petrographic study. It is possible to outline in this paper only some of the more important results obtained.

It should be pointed out that the term dolomite is used here in the broad sense to include both normal dolomite and dolomitic limestone. It is not necessary to differentiate between these in a discussion of their origin.

The field studies undertaken during the course of this investigation have alone furnished irrefutable evidence that most of the dolomites examined, regardless of their age, are replacement products. The following facts support this contention: (1) the lateral gradation of beds of dolomite into limestone, sometimes very abruptly; (2) the mottling of limestones by

irregular patches of dolomite on the borders of dolomite masses; (3) the existence of remnants of unaltered limestone in dolomite, and of nests of dolomite in limestone; (4) the irregular boundaries between certain beds of limestone and dolomite; (5) the presence of altered oolites or fossils in many dolomites; (6) the protective effect of shale beds; and (7) the obliteration of structures and textures.

In some instances the relationship of dolomite to limestone is such as to indicate that the alteration was accomplished by solutions which migrated from above downwards after the limestone was formed, or at least in the closing stages of its formation.

It is an interesting fact that certain layers have sometimes been passed over during the dolomitization of adjacent ones, and show little or no sign of alteration. The so-called interstratification of limestone and dolomite cited by some as evidence in favor of some primary theory of origin is then, in some cases at least, rather a pseudo-inter-stratification produced by the selective dolomitization of an original limestone. Some layers which have been passed over have been noted to be coarser grained than the adjacent layers which have been altered and this would seem to explain their greater resisting power. At times, however, the unaltered layers do not appear to differ markedly from the altered ones. The phenomenon is then difficult to account for. Normally the contact lines between such interbedded layers of limestone and dolomite are fairly regular and definite, but in some instances they are known to be very irregular and may even simulate irregular contacts produced by disconformity. A remarkable example of a pseudo-disconformity produced by uneven selective dolomitization has been observed in the St. Louis limestone near Farmington, Iowa. Here a bed of altered limestone is found resting very irregularly on a bed of dolomite. The two beds are very different physically and might readily be taken at first sight for two distinct formations, but when the contact is traced laterally for a short distance the lower bed loses its dolomitic character and passes into a limestone very similar to and continuous with the bed above.

Another striking relationship of limestone to dolomite is exhibited in a certain layer of an interbedded series of limestones and dolomites of the Beekmantown in the old Walton Quarry near Harrisburg, Pa. The beds dip south here at an angle of 30° . The layer in question is represented by dolomite six feet in thickness in the upper part of the quarry face and on each side of it appear good limestone layers. Now in the lower part of the quarry the lower half of this layer passes abruptly into limestone and continues to the quarry floor as

two distinct layers each 3 feet thick. Samples of the dolomite at the point where it passes into limestone yielded 18.1 per cent of MgCO_3 , while the limestone itself yielded only 0.83.

It will be noted that in the above instances the gradation of limestone into dolomite is abrupt, but in many cases the gradation takes place through transition zones of limestone mottled with dolomite. There can be no doubt but that these mottled limestones represent an incipient stage in the process of dolomitization and it is believed that many dolomites have passed through such a stage in the progress of their formation. In most cases the phenomenon of mottling appears to be of purely inorganic origin, having resulted from a process of dolomitization which began at certain favorable centers and spread outwards. In some cases, however, it has been produced by the selective alteration of areas suggesting algæ and fucoids in the limestone first, and the spreading out of the dolomite from these as nuclei. The Tribes Hill limestone, as developed at Canajoharie, New York, furnishes an excellent illustration of the mottling produced by the latter method. All stages of mottling from altered fucoid-like markings to a rock uniformly dolomitic may be traced in this.

It has been observed that the spreading of dolomitization from certain centers in a limestone may give rise to mottling on a large scale if these centers be few and far apart. For example there is a conspicuous bed of dolomite pseudo-boulders in the St. Louis limestone at Alton, Ill., which appears to have been formed entirely in this manner. These boulder-like masses range from a few inches up to six feet in diameter and contain 32.39 per cent of MgCO_3 , while the limestone matrix bears only 3.39. That they were formed in place is clearly indicated by the fact that the contact of the boulders with the limestone matrix is occasionally gradational and that the stratification lines of the limestone may at times be traced directly through the boulders. In a layer of limestone a few feet above the boulder bed here a similar process of local dolomitization has given rise to the development of irregular lenses of dolomite.

It has often been noted during the course of the field studies that many dolomites known to be of secondary origin show little or no evidence of shrinkage and porosity determinations have since shown that the transformation of a limestone to dolomite, even subsequent to its deposition, need not necessarily be accompanied by a decrease in volume as pointed out by Beaumont and consistently adhered to by later writers on the subject. It seems probable, therefore, that the replacement may proceed at times according to the law of equal volumes as enunciated by Lindgren (41) and that the inter-

change need not be molecular. In view of this fact compact dolomites showing no shrinkage effects can no longer be regarded as primary.

Further studies will doubtless show that considerable shrinkage effects produced by dolomitization are not common. It is believed that many vesicular dolomites have resulted from atmospheric leaching long after their formation.

Petrographic Evidence.

The microscopic study of many thin sections of dolomitic limestone has not only further amplified and strengthened the field evidence but has also thrown new light upon the details of the process of alteration. By employing microchemical tests it has been possible to distinguish between calcite and dolomite in the sections and make clear the most intimate relationships of the two minerals. It should be stated, however, that these tests furnish no reliable guide to the exact amount of magnesia in the rock, for crystals containing less than 25 per cent of MgCO_3 may behave essentially like normal dolomite. But this is to be regarded in truth as a distinct advantage, for alterations of only a slight degree are indicated as well as the more marked ones.

It must be admitted that if dolomite has a diverse mode of origin the microscope fails to reveal it. Careful examination of every variety of dolomite fails to show any positive evidence in favor of either the primary chemical or the elastic theory of origin. On the other hand, there is abundant evidence in favor of the alteration theory. It is true that certain dolomites, whose origin is not certainly known from their field relations, possess an extremely fine and uniform texture, and this feature has in fact led Daly (42) to believe that these represent original chemical precipitates. In order to test the validity of this argument the finest grained dolomite of unknown origin encountered by the writer in these studies was compared with the finest grained dolomite of known secondary origin. For example the Jefferson City dolomite of the Ozark region, whose mode of formation is not definitely known, possesses unusually dense and compact layers which are seen under the microscope to be made up of minute granules the majority of which are below $.003^{\text{mm}}$ in diameter, some measuring only $.001^{\text{mm}}$. The granules are suggestive of an original chemical precipitate. The strength of this interpretation is weakened, however, by the fact that a dolomite of known secondary origin has been found in the Middle Devonian of Iowa which is equally as fine-grained. The latter dolomite has resulted from the alteration of a dense, lithographic-like limestone with the approximate retention of the original texture.

As regards the possibility that some dolomites may be of clastic origin, none has been found which exhibits any signs of clastic structure. But that the original structure might have been obliterated during recrystallization is easily conceivable in rocks of this type.

Turning now to the dolomites which from their field relations are known to be secondary after limestone we have much more definite data. Indeed in these, by virtue of the fact that the alteration has frequently been halted before it proceeded to completion, we are often able to trace all stages of dolomitization from a limestone showing only incipient alteration to a good dolomite. Thus, it is possible to describe the steps normally passed through during the transformation of a limestone to dolomite.

So far as the testimony of the microscope goes the fine-grained limestones are more susceptible to alteration than the coarser-grained ones, a fact which is in keeping with the laws of chemistry. The evidence also indicates that the alteration may not proceed in exactly the same manner in the two types of limestone.

The alteration of fine-grained compact limestones seems to be accompanied normally by a notable increase in size of grain. Usually the diameter of the dolomite crystals formed is many times as great as that of the original calcite grains. But in rare cases, such as that of the dense Middle Devonian dolomite referred to above, the original structure and texture seems to be approximately retained. In the dolomitization of such fine-grained limestones the replacement frequently begins at many centers throughout the rock and spreads outwards from these, or if the rock possesses fine stratification the replacement may follow closely these original lines of weakness in the early stages. In those cases where the alteration begins at certain centers and spreads out from these, fucoid-like markings occasionally serve as the nuclei as in the case of the Tribes Hill limestone. But as a general rule no organic influence is noted. Normally the limestone is altered uniformly in the process of spreading from the dolomite centers, but it cannot be said that it is altered completely, for the dolomite patches often possess less than twenty per cent of MgCO_3 . Small remnants of limestone, however, may occasionally escape alteration and become incorporated in the dolomite patches. The boundary between the limestone and the spreading dolomite area may or may not be abrupt. When abrupt, the rock may assume the appearance of a breccia and the term "pseudo-breccia" may aptly be applied. When the boundary is gradational, on the other hand, rhombohedrons of dolomite, variable in size but usually nearly perfect in their development, are disseminated through

the limestone a short distance in advance of the main dolomite area. As the replacement proceeds the dolomite areas grow larger and larger and eventually meet and become confluent thereby giving rise to a rock which is uniformly dolomitic. Further addition of magnesia may then take place by altering the rock more completely.

In the coarse-grained limestones, especially those which were originally coarse-grained, such as the crinoidal limestones, on the other hand, mottling does not seem to be the rule in the early stages of alteration. In these the replacement appears to affect the matrix first and to spread rapidly through the rock. The coarser grains are next affected, being broken down into aggregates of small dolomite grains. In the end a coarse-grained limestone may be changed over into a uniformly fine-grained one.

In the dolomitization of limestones of both types the calcareous skeletons of organisms appear in most cases to successfully withstand alteration and these, owing to their greater solubility than dolomite, are then removed to leave molds by a process of atmospheric leaching when the formation passes into the belt of weathering.

Conclusions.

Considering all the evidence, it seems probable that the great majority of our dolomites had their inception in the alteration of limestones. It will not be denied, however, that some dolomitic formations of minor importance may have had a different origin. For instance, some impure dolomitic limestones associated with shales very probably represent original clastic deposits which have not suffered any alteration whatever, and there is some reason for believing that certain dolomitic limestones high in siliceous material, such as the Silurian waterlimes of New York State, may have had a similar origin.

The importance of marine and surface leaching in increasing the magnesia content of limestones originally low in magnesia should likewise not be overlooked. There can be no doubt that this process has greatly enriched the more vesicular dolomitic limestones in magnesia. But the leaching theory does not explain the ultimate source of the magnesia. It merely shows how the magnesia content of a limestone originally low in this constituent can be enriched.

To return now to the dolomites which have resulted from the alteration of limestone, there are many reasons for believing that the more extensive of these have all been formed beneath the sea, and that dolomitization affected by ground water is only local and very imperfect. Some of the features

which lend weight to this view are as follows: (1) The dolomite areas of mottled limestones are believed to have undergone recrystallization at the same time as the associated limestone areas as suggested by the occasional development of zonal growths of calcite and dolomite. (2) In imperfectly altered limestones the dolomite is seen to follow original lines of weakness rather than secondary structures such as joints or fractures. (3) In most cases of mottling the dolomitization appears to have progressed uniformly as we should expect it to in an uncrystallized rock, rather than to have progressed by forming veinlets and stringers in the early stages. (4) The existence of perfect rhombs of dolomite in many imperfectly altered limestones suggests that the latter had not yet solidified when the dolomite rhombs were formed. (5) The widespread extent and nearly uniform composition of many dolomites indicates that they must have been formed by an agent capable of operating uniformly over wide areas. (6) An adequate source of magnesia for transforming extensive limestone formations into dolomite is found only in the sea which contains many times as much of this constituent as ordinary ground water. (7) Many dolomites are directly and regularly overlain by pure limestone formations or by thick shale beds, proving that they must have been formed before these overlying beds were deposited.

Some dolomites of minor importance, such as those associated with ore deposits and probably most, if not all of those related to fractures (vein dolomites), must have been formed through the agency of ground water. But in general, ground water is incapable of carrying dolomitization far. Study of analyses of ground water, and of river water, shows these to be uniformly low in magnesia, this constituent normally being greatly exceeded in amount by lime. How, then, could such waters dolomitize limestone when they already contain lime far in excess of magnesia? The law of mass action speaks strongly against ordinary ground water being able to accomplish extensive dolomitization. In the case of mineral springs and the mineralizing solutions which are related to ore deposition, however, it is conceivable that magnesia might be present in sufficient proportions to accomplish local dolomitization and doubtless most vein dolomites have been so formed.

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