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Thermoluminescence in Carbonate Rocks and its Application to Stratigraphic correlation and Geologic Age Determination

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THERMOLUMINESCENCE IN CARBONATE ROCKS
AND ITS APPLICATION TO STRATIGRAPHIC CORRELATION
AND GEOLOGIC AGE DETERMINATION

A thesis
Presented to
the Faculty of the Department of Geology
University of North Dakota

In Partial Fulfillment
of the Requirements for the Degree
Bachelor of Science of Geology

by
E.J. Englerth
May 1958

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Thermoluminescence in Carbonate Rocks
and its application to Stratigraphic Correlation
and Geologic Age Determination

by
E.J. Englerth

Abstract

Thermoluminescence is the temperature dependence of phosphorescence and is common to most carbonate rocks. Although the phenomenon has been known since at least 1663, its use in geology was not investigated until 1948.

Thermoluminescence results from the release of energy stored as displaced electrons in the crystal lattice. Thermal agitation of the displaced electrons causes them to give up part of their energy as light as they move back to their original positions in the lattice. The intensity of the light emitted is plotted as a function of temperature, resulting in a glow curve characteristic of the carbonate sample.

To date, the most promising uses of thermoluminescence in geology are in stratigraphic correlation and geologic age dating. Both short- and long-range correlations have been attempted. No figures pertaining to geologic age dating by this method have been published.

Results of investigations show that, while glow curve data may be of some value, many problems must be solved if the data are to prove reliable. In order to realize the possibilities of thermoluminescence as a geologic tool,

chemical investigations, tectonic studies, and standardized procedures must be carried out and perfected. In the absence of such studies, thermoluminescence must remain a geologic aid of doubtful accuracy.

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Thermoluminescence in Carbonate Rocks
and its application to Stratigraphic Correlation
and Geologic Age Determination

Introduction

Thermoluminescence can be thought of as the ability of a mineral to glow upon the application of heat. The use of thermoluminescence as a geologic tool has been under investigation since 1948, and attempts have been made to apply it to stratigraphic correlation and to geologic age determination. The subject is not adequately treated in stratigraphy texts because of its newness to geology and because of the fact that the results of investigations are widely scattered throughout the literature.

An effort has been made to present this paper from the viewpoint of the geologist. Therefore, the theory, equipment, and procedures have been only briefly treated, and the problems have been presented in a broad outline. For more detailed information, the reader is referred to the references cited.

The writer wishes to thank Professor F.D. Holland, Jr. for suggesting the problem, for providing access to literature ordinarily unavailable through library sources, and for his advice and constructive criticism in the preparation of this paper. The help of Nancy Englerth in preparing the illustrations is also gratefully acknowledged.

The History of Thermoluminescence

Thermoluminescence has been defined as the luminescence stimulated by heat. It has also been defined (Rieke, 1954, p. 1) as the temperature dependence of phosphorescence. The latter definition is preferred, since the source of excitation is high energy radiation and not temperature.

Historically, the phenomenon of thermoluminescence appears to have been first mentioned by Boyle in 1663, followed by Kohler and Leitmeier, Calaját y Leon, Royer, and Saurin (Saunders, 1953). Regarded as a mineralogical curiosity for nearly 250 years, the first quantitative measurements of thermoluminescence were made by Pierce in 1902. Wick in 1927 and Steinmetz in 1934 carried out X-ray excitation studies in this country, while spectral studies connected with quantitative measurements were carried out mainly by Japanese scientists (Saunders, 1953).

In connection with a U.S. Atomic Energy Commission research program, studies under the direction of Professor Farrington Daniels were begun in 1948 at the University of Wisconsin to determine the possible uses of thermoluminescence in geology. The results of these studies will make up the major portion of this paper.

The Theory of Thermoluminescence

The current theory of thermoluminescence was formulated in 1916 by Meyer and Przibram (Saunders, 1953, p. 118) after studying the decoloration and luminescence produced

in different artificial materials by high energy radiation. This theory was accepted by Lind and Bardwell in 1923 as an explanation of the thermoluminescence and coloration they had observed in minerals exposed to radium.

In view of the many explanations of the Meyer-Przibram theory found in the literature, the writer has attempted to synthesize an explanation from the literature intended primarily for the geologist. In so doing, the explanations by Daniels, et al. (1953), Saunders (1953) and Zeller (1954b) have been heavily drawn upon.

Thermoluminescence may be either natural or induced. To exhibit thermoluminescence, a substance must have an ordered structure such as is found in crystals, or a semi-ordered structure such as glass. In addition, it must be electrically a semiconductor or an insulator.

Thermoluminescence is considered to result from the release of energy stored as displaced electrons in the crystal lattice. All real crystals have a certain number of imperfections such as missing ions, interstitial ions, or impurities. Electrons knocked out of their normal orbits in the crystal lattice by high energy radiation such as X-rays or gamma rays may become trapped at these imperfections. The force field around each trap prevents escape of the electrons under normal conditions, but thermal agitation of the crystal ions may provide sufficient escape energy. When the electrons return to their normal positions in the

lattice, some of the energy they possessed while in the traps may be released in the form of light. This light is known as thermoluminescence.

The natural thermoluminescence of minerals is due to electrons driven into traps by the radiation from radioactive impurities in the crystals. Many of the electron traps are thought to be of the missing ion type caused by the dislocation of lattice ions from their normal positions due to alpha-particle bombardment from the impurities. Although the natural rate of production of these dislocations is very small due to the low concentration of radioactive material, the great lengths of time available are sufficient to produce large enough numbers of imperfections to account for the observed thermoluminescence.

Induced thermoluminescence may be effected by any of the common laboratory methods employing high energy radiation.

General Procedure

The technique of measuring thermoluminescence was first developed by Urbach and later modified by Randall and Boyd (Daniels, et al., 1953, p. 344). Samples of carbonate rocks in solid or powder form are heated in an electric furnace at a rate of 1°C per second through a 500°C temperature range. The light emitted is measured by a photomultiplier tube, while the specific temperatures are obtained by a thermocouple. The outputs of these devices are recorded by a potentiometer recorder. The intensity of thermolumines-

cence is plotted as a function of temperature and is called a glow curve. It is by the use of the glow curve that stratigraphic surface and subsurface correlation and age determination are attempted. Typical glow curves are shown in Fig. 1.

Characteristics and Properties

The glow curve of any thermoluminescent material possesses certain properties, some peculiar to the material and others peculiar to the treatment of the material.

A partial list (Daniels, et al., 1953, p. 345) of materials exhibiting thermoluminescence after gamma radiation include:

calcite	quartz
dolomite	glass
fluorite	feldspars
alumina	feldspathoids
magnesium oxide	certain clays
gypsum	ceramics

Any glow curve exhibits a number of light emission maxima, the number being dependent upon the material and its treatment, though in all cases the maxima or peaks are thought to arise from the release of self-trapped electrons in the crystal lattice. As previously stated, there are several types of electron traps thought to exist; and since each type demands a characteristic activation energy, the different peaks in a glow curve represent the different types of traps present in the sample.

The number of peaks varies widely over the range of minerals tested and even within the same mineral. X-ray

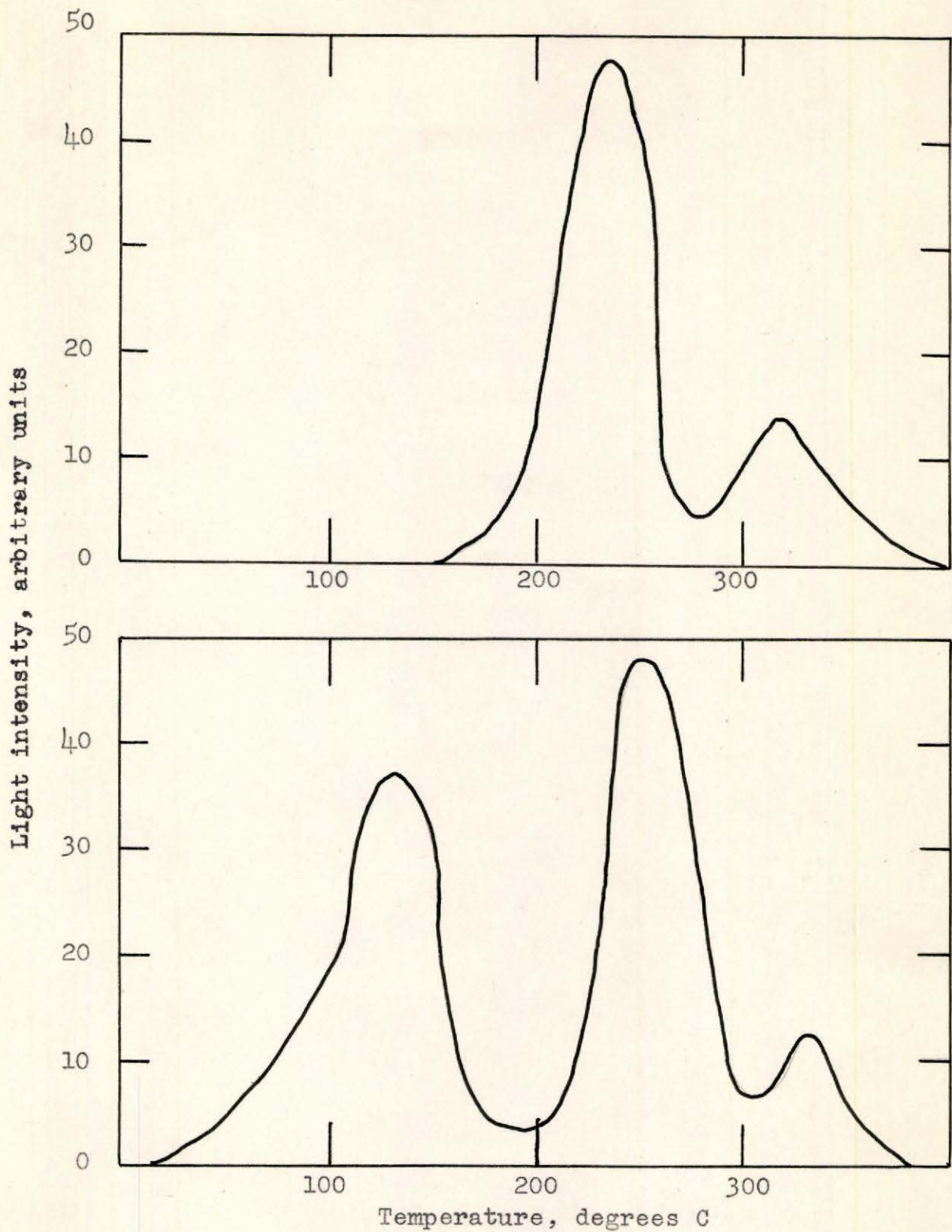


Fig. 1. Thermoluminescence glow curves of a limestone sample in its natural condition and after alpha irradiation. The low temperature peak is drained under ordinary rock temperatures.

Modified after Zeller, 1954c

treatment gives the following results (Keller and Clemmons, 1955, p. 587):

<u>Mineral</u>	<u>Peaks</u>	<u>Temp. Range (°C)</u>
LiF	3	155-310
KCl	1	140
NaCl	3	115-280
CsBr	1	110
NaI	0	-
KF	0	-
KBr	0	-
KI	0	-
RbBr	0	-

Ghormley and Levy (1952, p. 550) found that exposure to light of appropriate wave lengths caused thermoluminescence in some instances. Sylvite (KCl) treated in this manner exhibited five glow peaks between -196°C and 25°C , compared to one peak revealed by X-ray treatment at higher temperatures. Irradiation by X-rays and exposure to ultraviolet light (Hill and Schwed, 1955, p. 654) produced eight glow peaks in sodium chloride (NaCl) crystals between 334°C and 625°C , compared to three peaks found by X-ray exposure at lower temperatures.

In direct contrast, Wick and Slattery (1928, p. 406) concluded that ultraviolet radiation of frequencies very near the visible was capable of completely quenching the ability of a mineral to thermoluminesce.

Samples of rock exposed to gamma or X-rays before heating (Bergstrm, 1956) exhibit greater thermoluminescence than is obtained without previous laboratory radiation. This fact is utilized in stratigraphic correlation.

Pressure is found to have a decided effect upon the thermoluminescence of limestones (Zeller, 1955, and Handin, et al., 1957). Lewis (1956) has attempted to relate thermoluminescence to chemical composition. The significance of these studies will be discussed in conjunction with stratigraphic correlation.

Thermoluminescence in Geology

To date, the most promising use of thermoluminescence in geology appears to be its application to the stratigraphic correlation of carbonate rocks. Work on correlation by this method was begun by Saunders (1953). Other workers are Parks (1953), Bergstrom (1956), Lewis (1956), and Pitrat (1956). Saunders' basis (1953, p. 118) for attempting correlation by this method is as follows:

It can be seen that the shapes of the gamma-activated glow curves of several different limestones from one outcrop locality are different as are the amounts of thermoluminescence as shown by the heights of the maxima.

Although the exact nature of the particular traps which form the several peaks of the calcite glow curve are not known, one may hypothesize that in general the type of glow curve observed for a limestone reflects the composition and crystallization history of the calcite contained therein. Under normal conditions of limestone formation the conditions which control these variables might be expected to be constant over relatively wide areas of one stratigraphic horizon and would vary according to the position in the geologic section. If this is true, then it should be possible to apply the glow curve characteristics as a tool in identifying a certain limestone stratum or sequences of strata in different outcrop localities.

Stratigraphic Correlation

Surface Correlation

Saunders' first attempt at correlation by use of glow curves was made on a large quarry of Niagaran limestone in Wisconsin and employed both lateral and vertical sampling over short distances. The results, based on lateral similarities of glow curve shape and intensity of thermoluminescence in the same formations and differences vertically in glow curve shape between different formations, supported his hypothesis that the method was feasible for short distances.

In a subsequent attempt samples were taken for every three feet of exposed section of three correlative limestones, the Pahasapa, Redwall, and Madison, all of Lower Mississippian age, in an effort to establish long range correlation. The glow curves showed similar trends in each case. Saunders concluded that the results could be taken as a substantiation of previous correlation in thick limestones where long and undisturbed periods of deposition were involved and where depositional conditions varied widely for other limestones in the section. In addition, it was concluded that correlation might be possible in thin cyclical limestones by means of the relative amounts of thermoluminescence of several consecutive formations in each locality. Saunders also found a linear relationship between thermoluminescence and the amount of radioactivity

present as an impurity.

At about the time of Saunders' studies, Parks (1953) began work on the subsurface correlation of certain limestones in the Illinois Basin. Parks, like Saunders, found a linear relationship between thermoluminescence and radioactivity.

Bergstrom (1956) conducted studies of twenty-five limestones of Pennsylvanian age in Iowa, Missouri, and Kansas in an attempt to determine the relative value of natural plus gamma-ray induced thermoluminescence of limestones in stratigraphic correlation. Variations in vertical sections were examined also to determine the possibility of establishing zones. Insoluble residue studies were run on samples of the limestones tested.

Bergstrom agreed that thermoluminescence measurements may aid in stratigraphic correlation but that the technique was definitely not applicable to all limestones. It was found that some limestones could be correlated, some could not be, and that for others the evidence was inconclusive.

Many important conclusions were reached in Bergstrom's studies:

- 1) Glow curves of a limestone of variable composition are highly variable in both shape and amplitude. In such circumstances correlation based on thermoluminescence is impossible even in a small area.

- 2) In general it seems that the consistency of the

physical and chemical characteristics of the limestone is paralleled by a persistence in specific thermoluminescence characteristics. Some limestones apparently show no lateral persistence of specific thermoluminescence.

3) Because many factors apparently control thermoluminescence, a glow curve can be thought of as representing an algebraic total of diverse physical and chemical influences.

4) An insoluble residue content in excess of 15-20% tends to depress thermoluminescence to a point where the glow curve is rendered useless.

5) Dolomitization and recrystallization probably exert a tremendous influence on glow curve properties.

6) The impurities present, which activate or suppress thermoluminescence, are more important in controlling thermoluminescence than is the level of radioactivity.

7) The greatest thermoluminescence for a given limestone is generally found in samples taken from the base of the limestone where the insoluble content (and presumably the amount of impurities present in the calcite lattice) is usually the highest.

8) The level of radioactivity of a limestone sample as determined by alpha-particle count bears no constant relationship to the amount of thermoluminescence.

9) It is possible that thermoluminescence-variation curve anomalies, like self potential and resistivity anomalies in electric logs may have some use as stratigraphic

markers, provided sufficient vertical coverage is available.

10) Some cyclical limestones give cyclical glow curves. This suggests that the glow curves are closely related to the lithology and chemical composition of the limestone. By this theory, some of the limestones studied with a wide range of radioactivity give patterns and intensities comparable in all respects because the limestones were formed under quite similar chemical conditions.

Saunders' suggestion that cyclic limestones could be correlated by glow curve data was rejected. However, it was felt that the apparent sensitivity of thermoluminescence to lithologic or chemical changes in the limestone, and the fact that glow curves reflect changing lithologic types and environments of Pennsylvanian cyclic sedimentation suggest that thermoluminescence might be used in yielding information on limestone composition, depositional environments, and crystallization history.

The problems arising from Bergstrom's studies are very significant and lead to the realization that, in general, the problem of thermoluminescence correlation is far more complex than found in Saunders' and Parks' studies. The fact that a linear relationship between thermoluminescence and radioactivity was not found by Bergstrom emphasizes the need for closer studies of relationships between thermoluminescence and chemical and mineralogical composition, as emphasized by Lewis (1956, p. 698). Further emphasis can

be placed on this need by realizing that half the workers using thermoluminescence as a stratigraphic tool (Saunders and Parks) have found a linear relationship with radioactivity, while the other half (Bergstrom and Pitrat) have not. It is possible, however, that other impurities predominate over radioactivity in controlling thermoluminescence only along the margins of basins closest to source areas (Bergstrom, 1956, p. 941).

A laterally persistent glow curve for a lithologically uniform limestone suggests persistence of the physical and chemical influences, but because the glow curves apparently represent only a cumulative effect of many factors, similar glow curves may be obtained from limestones that are lithologically and stratigraphically different. This difficulty was encountered when two types of glow curves from one formation in Iowa were duplicated by a formation in Missouri despite the fact that the two formations were faunally and lithologically dissimilar. This means that correlation on the basis of thermoluminescence is risky when isolated limestone samples are being considered.

In summary, thermoluminescence studies may aid in correlation when used under the following provisions (Bergstrom, 1956):

- 1) They should be attempted as an aid only in sections spaced to reveal all lateral variations.
- 2) The method of using vertical sequences of thermo-

luminescence variation plus persistent "marker" glow curves wherever they occur has some promise in correlation of thick limestones or alternating limestone-clastic sections. This method tends to mask individual irregularities in the overall glow curve pattern.

3) Identification of a given limestone in an alternating limestone-clastic section is possible only if there are no sudden, marked, lateral lithologic changes in the limestone, since thermoluminescence characteristics change abruptly with such lithologic changes.

Studies were conducted by Lewis (1956) and Pitrat (1956) in an effort to relate the variations in thermoluminescence to the chemical, mineralogical, and radioactive content of the limestone.

According to Lewis (1956, p. 698), the glow curve structure is greatly influenced by any trace impurities and by defect structures in the crystal. Thermoluminescence variations in both calcite and dolomite may reflect both the differences in lattice structure and the distribution of impurities in the lattice.

After evaluating the results of tests made on the Honeycut Bend formation, Lewis stated that the glow curve peaks could be simply related to the mineralogical composition. Highly dolomitized samples had only one peak, while samples containing more than 10% calcite exhibited two glow curve peaks. As a further check, tests were made on samples

of magnesite from localities in the United States and Europe. No detectable luminescence was produced before irradiation. After gamma-ray irradiation, the maximum intensity observed was much less than 1% of any of the irradiated calcite or dolomite samples. Therefore, any contribution to the glow curve by any free magnesite present in the limestone and dolomite samples may be regarded as negligible.

Zeller's work (1954b) on the role of impurities as activators of thermoluminescence supported Lewis' important conclusions regarding the relation of glow curve characteristics to chemical and mineralogical composition. Zeller stated that impurity ions had a marked effect on thermoluminescence. In artificial precipitates magnesium tended to increase middle temperature thermoluminescence at the expense of low temperature thermoluminescence. Iron tended to act as an inhibitor, while strontium and manganese acted as activators.

In attempting to clarify the role of radioactivity and impurities as activators, Pitrat(1956) utilized the low temperature peak of the glow curves obtained from samples of the Mission Canyon and Lodgepole formations. From this study, it was felt that the glow curve shape was more useful in stratigraphic studies than the intensity displayed. Unlike Parks and Saunders, Pitrat was unable to detect a linear relationship between radioactivity and thermoluminescence.

A number of samples were examined spectrochemically by Pitrat in an attempt to discover a relationship between glow curve shape and concentration of natural impurities. Lewis' conclusions were again supported. Only magnesium appeared to bear any direct relationship. Magnesian limestone and dolomite tended to have a higher second peak, whereas the calcitic limestone has a higher first peak. It was thought that the shape of the low temperature part of the curve may be controlled exclusively by magnesium. On this basis, it was concluded that the magnesium content bears an inverse relation to the ratio of peak 1 to peak 2 of the glow curve.

On the basis of his efforts at correlation, Pitrat felt that it was possible to differentiate between the Mission Canyon and the Lodgepole formations of the Madison group. It did not appear feasible to further subdivide either formation by the use of glow curve data.

It was emphasized that extreme care be exercised in working with dolomites, since post-depositional addition or subtraction of magnesium by ground water or hydrothermal activity could have serious effects on the validity of conclusions reached.

Subsurface Correlation

Parks (1953) has supplied very nearly all the information available on subsurface correlation. The Meramecian and Chesteran series of the Illinois Basin were chosen for

the study, as a brief survey of the thermoluminescence of the limestones indicated that sufficient differences existed in glow curve shape to permit identification and differentiation of the limestones.

Because samples came almost entirely from cable tool drillings, their small size necessitated the development of a new sample technique. Rotary cuttings could not be used at all due to their contamination by drilling fluids.

Cable tool samples were washed, sorted, and ground to <100 >200 mesh size. Gelatin capsules of the powder were subjected to fifteen hours of five curie gamma radiation, then kept refrigerated until used in order to minimize the draining of the lower temperature peaks. A thin layer of the powder was cemented to glass and the thermoluminescence measurements obtained as before. Radioactivity of the samples was determined with an alpha-particle scintillation counter.

This method was found to be quantitatively more accurate than the solid sample method and was used also by Pitrat and Bergstrom in their work on surface correlation.

Glow curves were run on both powdered and solid samples. Solid sample curves showed considerable variation in shape and wide variation in intensity. A comparison of solid sample curves with powdered sample curves from the same outcrop showed no appreciable change in curve shape, but did show considerable alteration in overall intensity relation-

ships. The powdered samples eliminated to quite an extent the factor of differences in transparency between limestones of different color and grain size.

When the glow curves of the subsurface samples were run, it was discovered that within any one formation the glow curve shapes and intensities varied through wide limits, including many of the variations apparently characterizing different formations at the outcrop. After further work, it became apparent that the pattern of variation in glow curve shape vertically within a formation would be sufficient basis to separate that formation from other formations and could be used as a basis for correlation. Patterns in variations were put in a form similar to that of electric logs to permit easy visual comparison and correlation.

In general, the glow curves of the Meramecian formations were found to be easily differentiated from those of the Chesteran formations. Only one Chesteran formation curve was found to resemble the typical Meramecian glow curve. Within the Meramecian it was found difficult to differentiate the formations on either the basis of glow curve shapes or overall intensities. Since two of the Chesteran limestones were too low in thermoluminescence to plot, it was felt that the very lack of thermoluminescence in these formations may be a useful correlation characteristic.

Parks' analysis of the patterns of variation anticipated Bergstrom's warning on the use of isolated limestone

samples.

A study of alpha-particle counts indicated a direct relationship, at least in the Illinois Basin, between radioactivity and thermoluminescence, although this relationship was not found elsewhere by Pitrat or Bergstrom. In effect, since all of Parks' sampling was done in the basin and not along the margins, Bergstrom's theory that suppression of radioactive dominance of thermoluminescence takes place along basin margins appears to be contradicted.

In summary, Parks (1953, p. 142) concluded that:

- 1) The pattern of variation in intensity of thermoluminescence, coupled with typical glow curve shapes, may serve to identify and characterize a formation.
- 2) Where several limestone formations occur in sequence with no great lithologic breaks, the intensity and glow curve shape variations may assist in recognizing tops and bottoms of formations.
- 3) Glow curve data may assist in splitting thick carbonate rock sequences into smaller units useful for correlation.
- 4) Once a typical pattern of variation is recognized for a formation, the absence of parts of that pattern at other localities may indicate erosion or non-deposition.
- 5) Within one basin of deposition and within one short period of geologic time, variations in radioactivity of the limestone appear to dominate over variations in activators

and variations in lithologic character in the cause of variations in thermoluminescence.

Pressure is found to have a decided effect upon thermoluminescence. Zeller (1953) found that high pressures applied to artificial precipitates of calcium carbonate caused a progressive decrease in low temperature thermoluminescence and a simultaneous increase in high temperature thermoluminescence. Also, when no pressure was applied the low temperature thermoluminescence was four times as intense as that at high temperatures. In a general way, natural limestones show similar variations in glow curve characteristics with increasing depth of burial.

Experiments on the Yule limestone by Handin, et al., (1957, p. 1218) showed that the effect of compression parallel to the optic axes (translation gliding) was to produce a new glow curve peak at about 280°C. The new peak was found to occur whether or not the calcite had been irradiated before deformation. The 240°C peak was somewhat reduced, but the high and low temperature peaks remained essentially unchanged.

These results agree to some extent with those of Zeller. It seems likely that differences in direction of compression may have some effect upon the retention of the low temperature peaks.

Summary

The feasibility of stratigraphic correlation by thermo-

luminescence as envisioned first by Saunders is not altogether certain. Subsequent work has shown that the technique is not applicable to all limestones, including those of extremely variable composition, and the thin cyclic limestones. On the other hand it is felt that the lack of thermoluminescence in some formations might be used as a correlation tool. The workers in both surface and subsurface stratigraphy are divided as to the relation between radioactivity and thermoluminescence. The writer feels that a tectonic study of this aspect of the problem would be of great help. Lewis' and Pitrat's studies have shown that the relation of the chemical composition to thermoluminescence is of the utmost importance and must be extensively studied and well understood before any further real progress can be made. In any case, the successful application of data to the problem is far more complicated than first supposed.

It is fairly well agreed that stratigraphic correlation by thermoluminescence is possible, but so many limitations have been imposed upon the method as to render it fit for only very restricted use in association with other methods of correlation.

Age Determination

The second possible use of thermoluminescence as a geologic tool lies in its use in age determination. Experimental work was begun chiefly by Zeller, Farrington, Boyd, and Saunders in 1948 and is continuing at the present

time.

The reason for the use of thermoluminescence as an aid in age determination has been well stated by Zeller (1954b, p. 181):

Since the discovery that carbonate sediments show a general increase in natural thermoluminescence with increasing geologic age, attempts have been made in the laboratories of the University of Wisconsin to produce a usable thermoluminescence age-determination method. Experiments have shown a proportional relationship between the amount of light emitted by a thermoluminescent substance and the amount of radiation to which the substance has been subjected. In theory, the geologic age of a sample could be determined by measuring the alpha radioactivity of the sample and the amount of light released. It is possible to bombard samples with alpha particles and thereby determine the amount of thermoluminescence induced in the sample for each unit of artificial irradiation.

Before an age determination can be attempted, it is necessary to determine the shape of the saturation curve and the level of ultimate saturation for each sample, since curve shape varies with chemical composition. The peak in the 300°C range has proved the most reliable, since it is the only peak that persists at ordinary rock temperatures. Saturation is attained through alpha-particle bombardment from a polonium source.

The impurity activated electron traps in any sample can probably hold electrons any time after lithification of the sediment; they will be characteristic of that particular sample, and their number will be finite. A saturation level will be reached eventually and further radiation will not increase thermoluminescence of the sample. If a lime-

stone or dolomite sample has reached saturation under natural conditions (Zeller, 1954b, p. 182) it is not possible to determine its age, although a minimum age could be established.

Following a study of radioactive decay, Zeller (1954b, p. 181) concluded that the effect of natural gamma ray bombardment could be neglected. It was also shown that calcites subjected to very large amounts of alpha radiation showed a reduction in thermoluminescence intensity.

Both Zeller and Farrington and his associates have employed different procedures. According to Farrington, et al. (1953, p. 344), alpha activity of the powdered samples is obtained by the use of a scintillometer. After saturation, the area under a selected high temperature peak in the natural curve is used for comparison. In another method tried, the electron traps thought to be produced by alpha particles are filled by saturating them with gamma rays. The light absorption of each sample is determined experimentally so that corrections can be made for light losses in samples that are not completely transparent. Limestones of known age are used for a calibration scale.

A one curie polonium source of alpha bombardment can be given to a thin surface layer in a second, which is equivalent to a million-year bombardment in a limestone containing a trace of uranium. This technique might cancel out the effect of impurities. Except for radiation damage uncer-

tainties, it was felt that the time required for the polonium source to match the natural thermoluminescence of the rock would give a direct measure of the alpha particles emitted since the rock was formed. Taking all corrections into account and knowing the rate of alpha-particle emission per hour and per year in the rock, the time to produce the observed thermoluminescence could be easily calculated.

According to Zeller (1954c), when the relationship between intensity of thermoluminescence and artificial radiation dosage for a particular sample is known by determining the shape of the calibration curve, the amount of radiation necessary to produce the natural thermoluminescence can be determined as illustrated in Fig. 2. In this figure a natural thermoluminescence of fifteen arbitrary light units intersects the calibration curve at an alpha-particle radiation exposure of 3.30×10^6 ergs. The amount of artificial radiation damage necessary to produce this amount of thermoluminescence is assumed to be equal to the amount required under natural conditions. Natural alpha activity is obtained by the use of a scintillometer and with this value it is possible to calculate the time required for the crystal lattice to receive an equivalent natural radiation dose.

Any process (Zeller, 1954a, p. 370) which causes the sediments to be heated to temperatures greater than 200°C will permit partial or complete drainage of the electron traps. If drainage has been complete, refilling of the

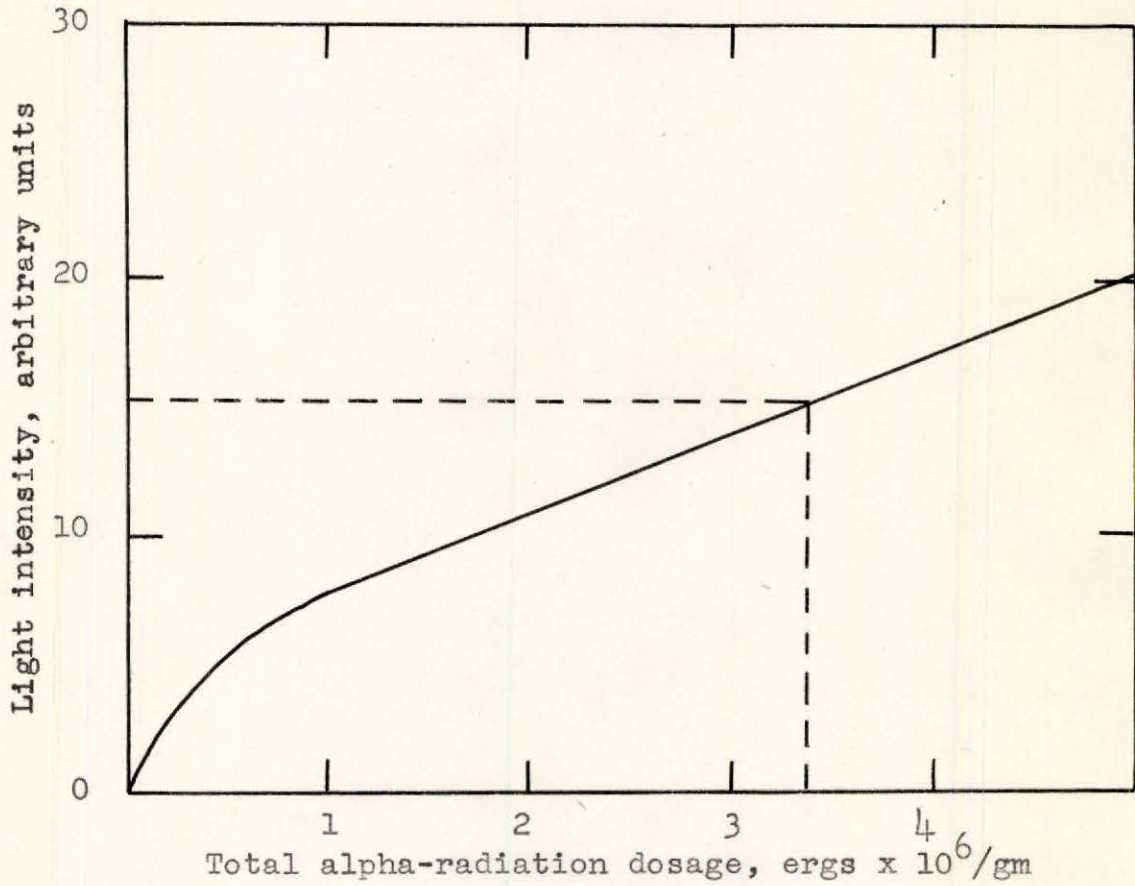


Fig. 2. Typical calibration curve showing the relationship between thermoluminescence light intensity and the total alpha-radiation dosage received by a sample of limestone.

Modified after Zeller, 1954c

traps will occur from the time that the sediments returned to normal earth temperatures. The age obtained for such a sample will be the age of the geologic event which caused the heating, and not the original age of the sample. Structural and igneous activity of sufficient intensity to cause recrystallization of limestone or dolomite might also be dated by this method.

In view of the earth's past diastrophic activity, it does not appear likely to the writer that many original carbonate ages will be obtained outside of sedimentary basins.

It should also be realized that the factors which make the thermoluminescence method applicable to dating periods of heating or recrystallization tend to operate to its disadvantage in stratigraphic work, especially subsurface studies. Well cores may be heated so high at the time of cutting as to drain the electron traps. When such recent heating has taken place it is easily seen in the glow curve, since the high temperature peak cannot be drained without draining the lower peaks. Therefore, any sample which produced a natural curve with only one peak might be suspected of recent heating, and it would not be possible to determine whether it had been partially drained or not.

Summary

Neither Zeller nor Farrington, et al. have published conclusive age-dating results to support their theoretical deductions. The work was done under the assumption that

there existed a direct relationship between radioactivity and thermoluminescence. This assumption was accepted also by Parks and Saunders (1952) in determining the age of mineralization of fluorite.

In view of the division of opinion concerning this assumed relationship and of many other complications previously noted, the reliability of geologic age determination by this method must remain in doubt pending further study of the controlling factors of thermoluminescence.

Summary and Conclusions

It is apparent that the application of thermoluminescence to stratigraphic correlation and to geologic age determination will meet with success only upon the resolution of many major problems. The outstanding problem is that of the nature of the chemistry involved in the mechanism of thermoluminescence. It seems likely that an understanding of the chemical role will lead to identification of the controlling factors of thermoluminescence. Lewis and Pitrat have pioneered these studies, but more work must be done if the phenomenon is to be understood, much less applied. The measurement of physical effects by Zeller and Handin, et al. has yielded information on the nature of the effect of forces within the earth on thermoluminescence. Thus, physical effects, though seemingly less important than chemical effects, cannot be neglected.

In view of the slightly different techniques employed

by different workers in sample preparation and curve determination (not fully enumerated in the text), it is not surprising that comparisons of results have often led to divergences of opinion. Procedures should be refined and made uniform so that the causes of differences may be narrowed down as much as possible. Extreme care must be exercised at all times.

Perhaps better results could be obtained by the use of a uniform procedure applied to a limited tectonic area and then to a succession of similar areas, rather than correlation attempted on the basis of widely scattered samples gathered without regard to the type of lithotope involved. Therefore a tectonic study of an area should precede sampling. Since the lithotope is in part defined by its chemistry, emphasis is again placed on the importance of the chemical role involved and on a thorough understanding of this role.

Due to the many uncertainties involved, the future of thermoluminescence as a stratigraphic tool is, at present, in doubt. Research leading to improvements in techniques and interpretation may someday make thermoluminescence available as a supplementary aid to the stratigrapher and geochronologist, though it may never become an independent method. Because the number of stratigraphic aids available at the present time is limited, every effort should be made to raise thermoluminescence from a curiosity to the status of a member of the family of geologic tools.

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