Bulletin de la Société belge de Géologie	T. 96	fasc. 1	pp. 15-21	Bruxelles 1987
Bulletin van de Belgische Vereniging voor Geologie	V. 96	deel 1	blz. 15-21	Brussel 1987

THERMOLUMINESCENCE IN MINERAL AND ENERGETIC RESSOURCES EXPLORATION, SOME CASE OF APPLICATIONS

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Abstract - After recalling the thermoluminescence (TL) phenomenon and its application possibilities for mineral, uranium and petroleum exploration, some examples are discussed. TL as applied in uranium exploration uses the recording of a fossil radiation dose, whereas TL as applied in raw materials exploration and more particulary in gold exploration uses the TL properties of quartz in relation to its crystallisation condition. The possibility of using TL in petroleum exploration is mentioned (origin of detrital material, paleo-temperature conditions). Finally a new TL apparatus with a microcomputer system is described. It permits an improvement in possibilities to discriminating between different types of quartz and uses a very low quantity of materials (about 0.01 gr).

Résumé - Après un rappel sur le phénomène de thermoluminescence et ses possibilités d'applications à l'exploration des ressources minérales et énergétiques (uranium, hydrocarbures), quelques exemples sont présentés. La TL appliquée à la prospection de l'uranium utilise l'enregistrement d'une dose fossile de rayonnement. La TL, appliquée à l'exploration des matières premières et plus particulièrement de l'or, utilise les propriétés TL du quartz en relation avec ses conditions de cristallisation. Le problème de la TL en exploration pétrolière est évoqué (origine du matériel détritique, conditions de paléo-température). Finalement, un nouvel appareil TL, couplé à un microordinateur est décrit. Il permet d'améliorer les possibilités de discriminer différents types de quartz tout en utilisant de très faible quantité de matière (environ 0.01 gr).

Mots-clés - Thermoluminescence, métallogénie, or, uranium, hydrocarbures.

Key-words - Thermoluminescence, metallogenesis, gold, uranium, petroleum.

A. TL PHENOMENON AND ITS USE IN MINERAL EXPLORATION

Thermoluminescence (TL) is the light emission of materials heated to temperature below incandescence. The emission of light against temperature is the glow curve which often shows several glow peaks (figure 1).

TL results from a thermal activation of electrons trapped by the lattice defects of a crystal. Therefore, TL requires chemical or physical defects in a mineral and the trapping of electrons by these defects.

The number of electrons trapped which give the TL intensity is dependent on various factors :

- TL sensitivity in relation to crystallization or recrystallization conditions.

- radioactivity which increases the number of electrons trapped but with a saturation level in relation to the nature of the mineral and its radioactive and thermal history.

- thermal or photodesexcitation effects which decrease the number of electrons trapped. Thus, geothermal effect, sunlight effect and paleoclimatological conditions can modify the filling

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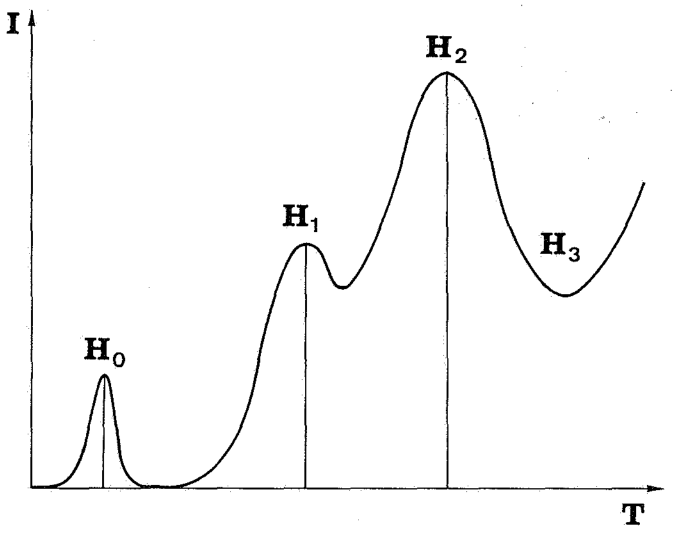


Figure 1. Glow curve of a sandstone.
H_0: glow peak intensity of an internal standard.
H_1-H_2 : glow peaks intensity for the sample allowing the calculation of the shape parameter $R = 100 \times \frac{H1}{H1 + H2}$

rate of the traps.

The number and the features of electron traps or the glow curve shapes are also dependent on various factors :

- crystallization or recrystallization conditions ;

- radiation effects more particularly displacement processes of atoms ;

- thermal effect caused by high temperature events.

The features of the glow curve can therefore be used for various types of applications.

Thermoluminescence can be measure of a radiation dose. Applications for uranium exploration have been developed by our laboratory within the framework of the EEC program in uranium exploration in 1979-1983 and with mining companies since 1984 (Charlet *et al.*, 1982 ; Charlet *et al.*, 1986).

Chemical or physical defects are chiefly related to with the crystallization conditions and so TL can be used to distinguish different types in one mineral species. Applications for raw materials exploration, sedimentology and petrology (more particulary petroleum exploration) can be developed from this point (Charlet & Quinif, 1985). Numerous studies have been carried out over the past twenty years by our research staff on the applications of TL in sedimentology, paleogeography, marine environment and granitoids petrography (Charlet, 1969 ; Charlet, 1971 ; Aloïsi & Charlet, 1975 ; Dupuis, 1973). Recently new research has been performed within the context of the EEC program in mineral exploration. Applications for gold prospection are developing from the district of St-Yrieix (France).

Finally, TL can be used to detect fossil geothermal history (Christodoulides *et al.*, 1971) and in this case, some applications for petroleum exploration in relation with organic matter evolution can be anticipated.

The glow curve of the whole rock is the result of the emission of various minerals. Among the rock-forming minerals, the following show thermoluminescence : K-feldspar, plagioclases, carbonates, quartz, ... The studies performed by our laboratory and by other researchers show that for each of these minerals the TL properties bear more or less a relation to their crystallization conditions (for example, origin of detrital material or their radiative history).

The quartz glow curve generally shows several peaks. This is of interest for using the glow curve shape as a method for discrimination between various origins : paleogeographical or sedimentological problems, quartz veins possibly in relation to mineralization. On the other hand, TL intensity reaches a saturation level after a few millions years so that, in normal conditions, detrital quartz can be only used for the localization of a fossil radiation dose if thermal or optical annealing affects this material (e.g. quaternary sediments which have been the subject of a sunlight effect).

Feldspars show a very poor distinctive glow curve with generally one very broad peak, although our later studies have revealed that the paleodose effect remains clearly evident even after a more or less long period.

So a complete mineral extraction from the whole rock is often of interest although this technique has not been applied in much research work. Besides, it can happen that the percentage of rockforming TL minerals is variable in the sample. Indeed TL of feldspars and carbonates is generally stronger than TL of quartz.

B. APPARATUS AND THERMOLUMINESCENCE TECHNIQUES

The main TL properties are obtained from the glow curve by plotting the light intensity of a mineral powder against the temperature. For some mineral species such as fluorine TL can be visible, but for most of them the light emission is very low and cannot be directly observed. It is accordingly necessary to use an apparatus with a detection system of low light emission.

In 1982, we developed a new TL apparatus (ATL 2000) which presents the following features :

- high sensitivity due to a photon counting technique and an optical system of high quality calculated for this experimental device and the nature of the TL emission. Without any difficulty one can use a very low quantity of material (0.01 gr) with a mineral of low TL such as quartz.

- a linear heating programme connected with a microcomputer (linear slope : 0,125 to 4° C/sec or any previously determined heating cycle).

- an interaction between the photon counter and a microcomputer for acquisition, processing and recording of TL data (Lair *et al.*, 1983). Mathematical computation from the glow curve is possible : smoothing, suppression of the black body emission, automatic research of extrema and inflexions calculus of peak values, peak integrals, shape parameters...

In mineral exploration, to discriminate between different types of quartz, the shape of the glow curve can be used with the number and temperature of the glow peaks, the relative intensity of the glow peak often designated by the factor R (figure 1). One can also use the TL sensitivity of a material for a standard irradiation. For example, in the gold district of St Yrieix (France), the quartz bearing mineralizations show for the same artificial irradiation dose, a sensitivity about ten times greater than that of the barren quartz (figure 2).

Therefore, TL after irradiation is very important. That is named "ATL" or artificial TL, as opposed to "NTL" or natural TL. "NTL" is dependent on the natural irradiation dose of the sample but also on others factors which can change the filling rate of the traps, such as the natural thermal activation or the photodesexcitation effect by the UV sunlight. This last effect can be very important for alluvials and also for some samples collected from old mining districts.

C. URANIUM EXPLORATION

Uranium exploration is based on the recording of a fossil radiation dose in relation with :

- radon migration or accumulation zones in the overburden.

- localization of the passage of the uraniferous mineralized solutions through favourable sedimentary structure (for example : sandstone type deposits, ...).

Numerous possibilities and advantages of this method may be mentioned :

- integration of the radioactive effect on a geological time scale.

- simultaneous use of some natural detectors (feldspars, quartz, ...) showing different TL properties (irradiation effect, sedimentological effect, ...).

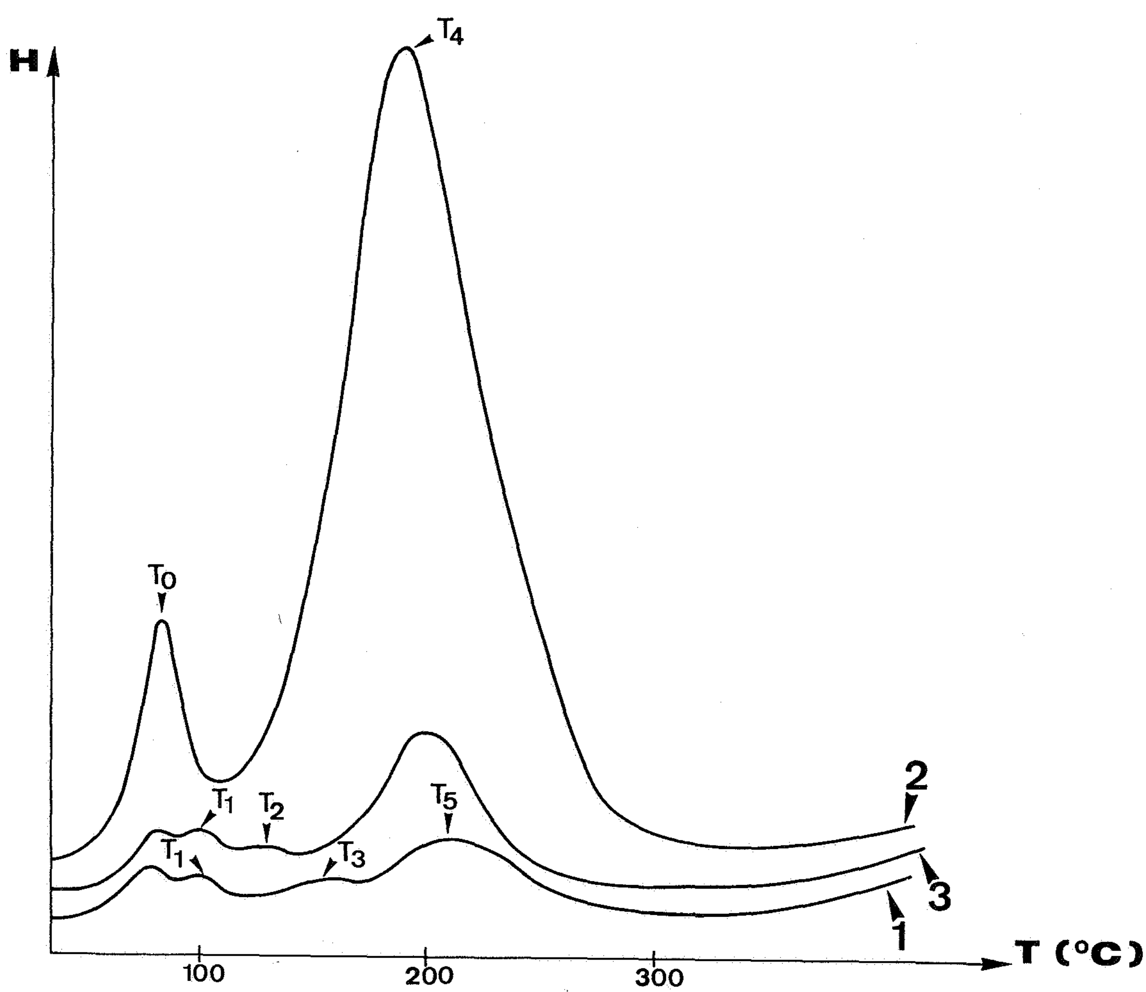


Figure 2. Artificial glow curves of quartz, gold district of St Yrieix.
1. Bull quartz (type 1, see fig. 4) ; 2. gold quartz or directly in relation with the mineralization (type 2, see fig. 4) ; 3. exsudation quartz in the barren gneiss serie (type 3, see fig. 4).

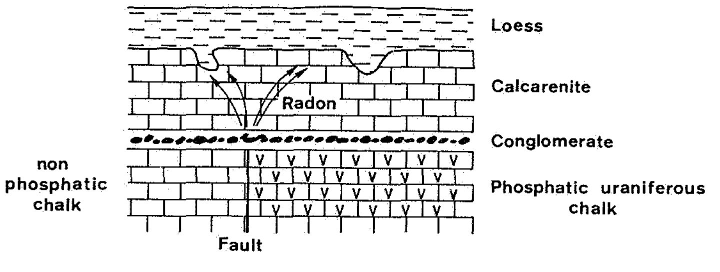
- flexibility because two field operations are not necessary as is the case with most radon methods.

- possibilities for using the cuttings or the samples recovered during drilling or geophysical prospection (e.g. seismic...).

- recording in a drilling survey of the traces marked by the passage of the uraniferous mineralized solutions.

- indication of possible nearby economic deposits during a reconnaissance survey or a detailed exploration.

Applications have been recently performed by our laboratory with various mining companies. A problem in this type of application is the saturation dose. It is necessary therefore to select some mineral species with a suitable reponse for a given radiation dose. We frequently use K-feldspar but a more recent development of this technique based on a methodology similar to the TL dating method of the Quaternary sediments (Wintle & Huntley, 1982) shows that different mineral species (quartz, feldspars) can be used in the superficial formations. From figure 3 a very good correlation may be seen between TL intensities of quartz and feldspars in a loam formation situated over a fault zone which affects an uraniferous phosphatic chalk formation. The high TL intensities correspond to the area with a stronger radon emission located by the gas techniques.



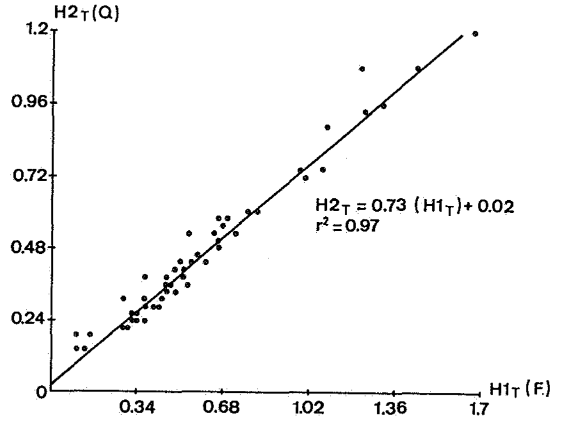


Figure 3. TL properties of a loess overburden (Mons bassin, Belgium). Correlation between the intensity of the first peak of the TL glow curve (peak assigned for the feldspars) and the intensity of the second peak (assigned for the quartz).

D. RAW MATERIALS EXPLORATION, APPLICATION IN GOLD DISTRICT

Raw materials exploration is based on the use of the TL properties to distinguish different types of the same mineral species in relation to its crystallization conditions. Quartz is a very good material. It is a common mineral in ore deposits, superficial formations, alluvials, ... Besides, its glow curve is very complex. Others minerals of gangues could be also used (feldspars, carbonates, fluorine, barytine, ...).

Numerous possibilities and advantages of this method may be mentioned :

- distinction of different types of mineralized structures.

- evaluation of the potentialities of a mineralized district from samples collected in old mining works (dumps, ...), the superficial formations, the alluvials, the cuttings and cores.

- determination of a genetic filiation between the barren quartz and the mineralized quartz and also between different types of mineralizations (for example between stratiform deposits and vein-type deposits).

An application concerning the gold district of St Yrieix (Limousin, France) has been recently developed by our laboratory within the framework of the EEC program in mineral exploration (Charlet & Quinif, 1985). St Yrieix is a district where gold was mined in the Gallo-Roman period and then again at the beginning of the 20th Century. Prospecting by the B.R.G.M. led to the opening of the Bourneix mine in 1982.

Samples have been collected from the dumps, the metamorphic series, barren lodes, alluvials and other superficial formations.

Figure 4 shows in artificial TL, three types of quartz having a different number of glow peaks, different temperatures of glow peaks and different sensitivity for a standard irradiation. It may be seen that type 1 is very frequent in barren quartz, type 2 is typical of mineralized quartz and type 3 is typical of the lenses or quartz veins of the metamorphic series. Therefore, it is possible to evaluate the potentialities of an ore district from the dumps, the quartz pebbles of the superficial formations or the alluvions.

Moreover, if the glow curve of type 2 is a feature of the gold-bearing quartz of St Yrieix, the TL sensitivity may be variable following the samples or the types of veins. At the scale of a regional structure, a diagram plotting NTL as against ATL intensity shows that the experimental point are situated on a straight line. But it is interesting to note that the equation of this line may be different following the structure (f.e., vein structure A'B' with

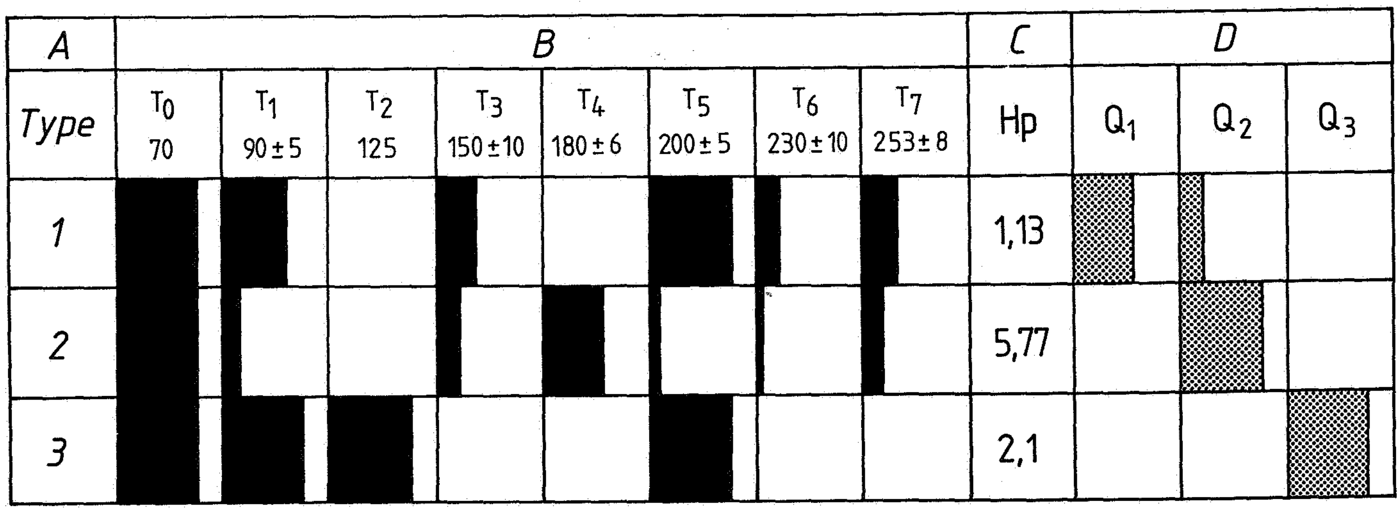


Figure 4. Gold district of St Yrieix (France), types of glow curves and their distribution among the veins quartz and the barren formations. A. type of glow curve ; B. temperature of the glow peaks (°C) ; C. mean TL intensity (in standard unit) ; D. type of quartz (Q_1, mikly bull quartz) ; Q_2. gold quartz or directly in relation with the mineralization ; Q_3. exsudation quartz.

particular minerals assemblages of antimony). From figure 5, it may be also seen that the localization of the experimental points of the barren quartz are situated near the origin.

It is at present impossible to explain these different behaviours in terms of TL mechanism. One can only note that for different structure-bearing mineralizations, there are sometimes different laws for NTL and ATL. So TL sensitivity may be variable from one sample to another. At the scale of a sample, we have sometimes variations of TL sensitivity from one zone to another with the low TL sensitivity generally corresponding to the low grade mineralized zone in spite of a same glow curve shape. The same glow curve shape for the gold-bearing quartz and sometimes for the milky quartz shows that the quartz of later crystallization can memorize the TL features of a mineralization. So the TL properties of the barren quartz can be an indicator of a mineralized zone.

Finally, figure 6 shows the general evolution of the TL sensitivity in the mineralized district of Limoges : very low sensitivity for the quartz of the St Sylvestre granite, the K-pegmatite and the occurence of tin-wolfram, high TL sensitivity spread widely for the gold-bearing quartz and the quartz of the sodic-pegmatite. If the tin-wolfram occurences appear in direct relation with the granite, other factors have probably occured in the metallogeny of the gold deposits. This is in agreement with the recent studies about the St Yrieix district which show that the gold bearing mineralizations are situated in a same lithotectonic unit (lower gneiss unit) of the Limousin metamorphic series (Ahmadzdeh, 1984).

E. MINERAL RESSOURCES IN RELATION TO PALEOGEOGRAPHICAL AND SEDIMENTOLOGICAL PROBLEMS

That TL utilization is based on the property of a mineral (quartz in particular) to acquire very different TL features in accordance with its crystallization conditions and thus with the origin of the detrital material. Various types of applications can be developed from this point : mineral deposits in detrital or volcano-detrital environments, uranium sandstone deposits, petroleum exploration...

We shall only make reference to a study on a COGEMA prospect where a relation occurs between a particular sedimentary structure revealed by the TL properties of quartz and the localization of uranium occurrences revealed by the TL intensity of feldspars. Figure 7 shows a principal lens structure delimited on the left and on the right, and at the top and the bottom by a high value of the quartz shape parameter R ($R > 45$). It corresponds to a sedimentological structure in which some uraniferous levels are developed (N_1, N_2, N_3, N_4). All these levels are not always mineralized at a local scale but the TL properties of feldspars have recorded the passage of uranium

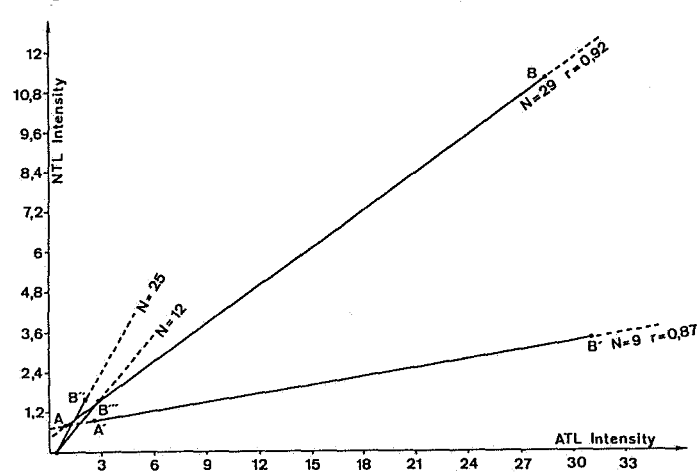


Figure 5. Diagram Natural TL - artificial TL for some quartz veins, gold district of St Yrieix (France).
N : number of samples.
r : correlation
B-B' : mineralized structures.
B"-B'" : barren veins.

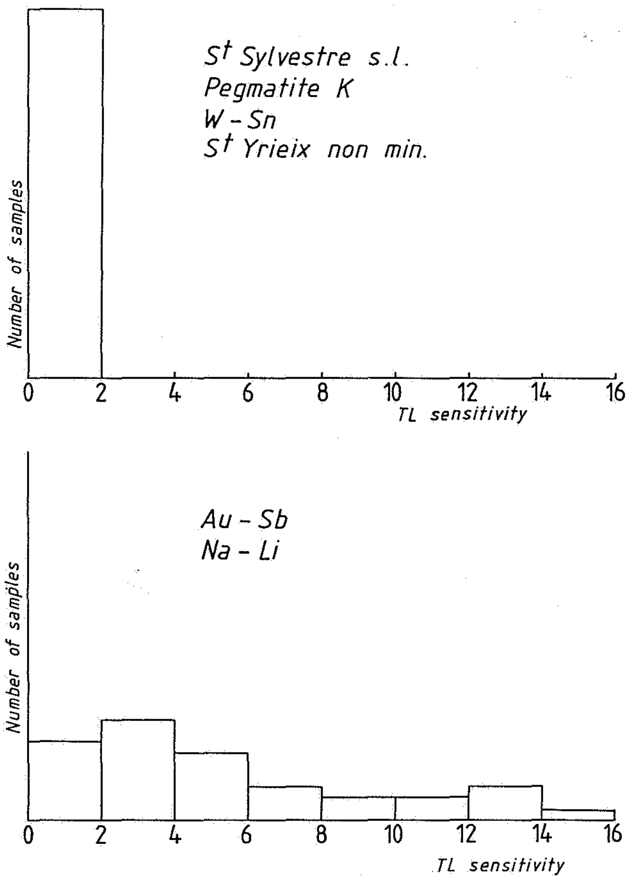


Figure 6. TL sensitivity for the quartz of the mineralized district of Limousin (France).

bearing solutions (high value of TL intensities). So thermoluminescence can prove to be an indicator of possible nearby economic deposits.

In conclusion, these applications of TL in mineral exploration show the possibilities of this method which can be used for various materials as well as for various geological environments. Thermoluminescence is a rather simple and low cost technique altough it takes place in the context of the very actual geospectroscopic methods in relation with the solid state physic of the mineral. However, for a fully application of TL in mineral exploration and mining development it would be again necessary to understand the specific causes of the modifications of the TL properties of quartz.

ACKNOWLEDGMENTS

These studies have been supported by the Commission of the European Communities and the "Service de la Programmation à la Politique Scientifique" (Belgium).

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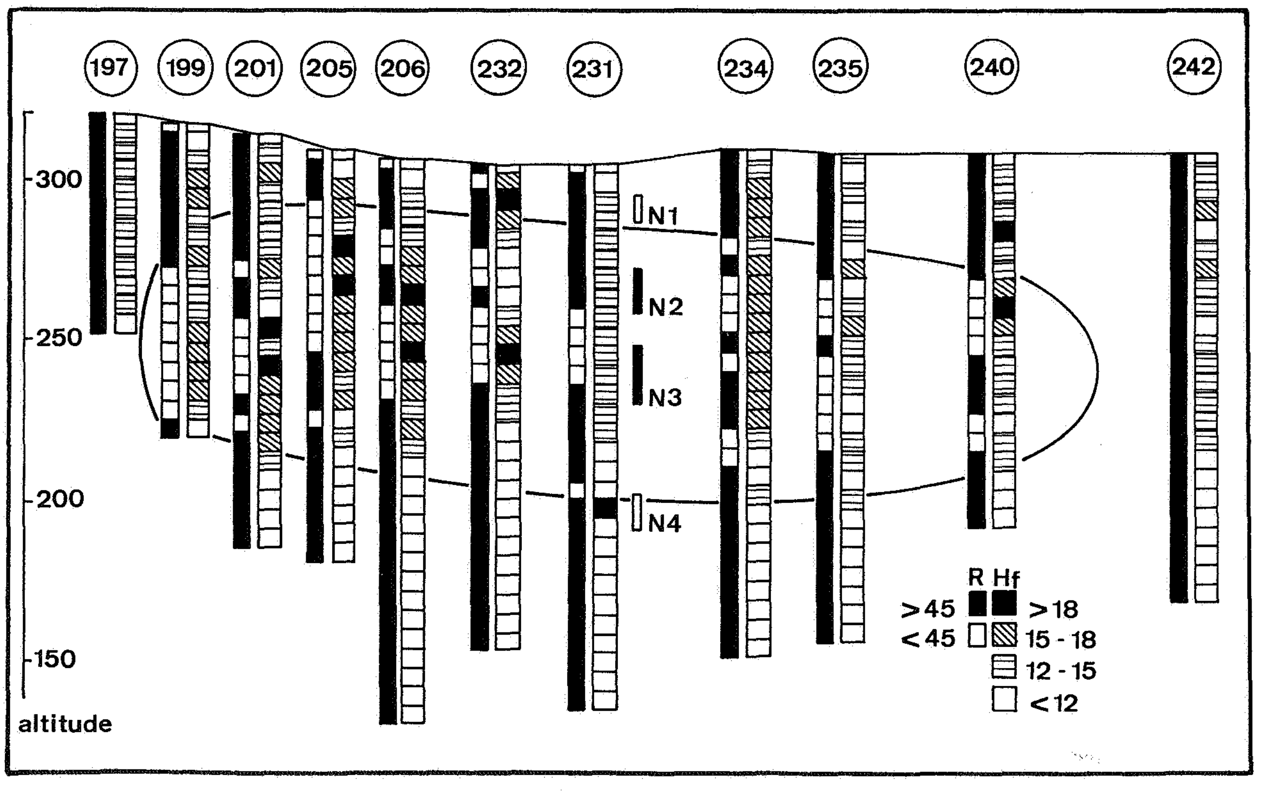

Figure 7. Distribution of TL properties for cuttings of a drilling survey.
R : glow curve shape parameter for quartz.
H_f : TL intensity for K-feldspars.

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