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Early medieval iron-smelting furnaces in Western Hungary
and their dating by thermoluminescence

Introduction

During the last 15 years the investigation of the Western-Hungarian slag-sites and the excavation of the iron-smelting workshops has been connected with various physical and geophysical measurements. The different physical dating methods (C^{14} , archaeomagnetic, TL) are even more important because at the sites of former industrial activity we have scarcely enough pottery to determine the age of the objects. These investigations are carried out by the members of the Industrial Archaeology Working Group of the Veszprém Regional Committee of the Hungarian Academy of Sciences.¹

During this project some types of early medieval iron-smelting furnaces were found which resemble more or less those excavated in Moravia by V. Souchopová at Olomučany.²

Therefore the finding circumstances and the possibilities of more exact dating - in this case the results and problems of the TL method - are important in the research of both territories. We hope that the exchange of experiences and the collaboration in the age-determination will help us to solve the problem: where do the various types originate from, and what connections can be observed during their further development.

The archaeological sites

These types similar to the Moravian ones are named in Hungary after the first important sites Nemeskér- and Imola types, respectively. The first means a free-standing shaft-furnace, having a breast-wall made of clay with tuyeres fixed into it. They also have a pit for the flowing-slag in front of them, as the reconstruction of this furnace from Nemeskér shows.³

The precedents of this type can be dated back on the basis of the sherds to the Avar age (Figs. 9, 10). The only difference between these and the later Nemeskér-type is that their basin is dug a little deeper (about 35-40 cm) into the soil and have got a deeper pit in front of them. They have been excavated so far in the outskirts of Avar villages at Tarjánpuszta and at Zamárdi and in 1988 at Somogyvámos.

At Tarjánpuszta (in W-Hungary) the furnaces came to light partly between the houses and baking ovens of the settlement, located without any system. E.g. among the remains of the destroyed stone oven of a house, iron slags were found in secondary position. The sherds date both the settlement and the workshops to the second half of the Avar period. The TL-samples were taken from the hard-burnt basins of the furnaces. The plaster was grey-burnt inside and red-burnt outside (Fig. 1).

The Zamárdi site lies South to Lake Balaton, in the vicinity of an extremely rich Avar cemetery with about 6000 graves. It was an important traffic-junction and perhaps an administrative centre in the Avar times.

At this site remains of the whole iron-smelting process were found: the furnaces are surrounded by oval-shaped roasting and charcoal-burning pits as well as reheating hearths. The surface of these pits is also red- (and partly grey-) burnt, so they are apt for taking TL-samples. Our samples originate from a roasting-pit (No.4), from a typical free-standing furnace (No. 1) and from a reheating hearth (No. 3) On the basis of the post-holes found on both sides, this latter was covered with a roof, so the smelters used it for a longer time.⁴

These burnt surfaces are suitable for cutting archaeomagnetic samples from them as well. The age determined in this way: AD 700-750 is in accordance with the TL-dating.⁵

The C¹⁴ results for this site (made by E. Hertelendi) are the following⁶:

OBJ. No. 6 672-777. A.D.

OBJ. No. 6 660-696. A.D. (Avar)

OBJ. No. 15 342-421. A.D. (Roman)

Another site dated with TL method is at Sopron-Kányaszurdek.

The free-standing furnaces were situated rather close to each other and their coating was built of stone. The reconstruction shows the supposed way of blowing, and that the flowing slag was collected in a pit before the breast-wall. Thus this variant represents a more advanced phase of the 9-10th century Nemeskér-type. Unfortunately only some pieces of the original stone coating remained on the site in situ or in secondary position.⁷ The samples for TL-dating were from the hard-burnt basins of the furnaces.

Since the sherds were not typical or were Roman ones lying in secondary position, the basis of the archaeological dating was the shape of the breast-walls and tuyeres found in situ. These closely resemble the Nemeskér ones.

The importance of the Kányaszurdok-site lies also in the fact, that the mine where the iron-ore possibly originates from, was also found and excavated. It is a pit-field consisting of 227 funnel-shaped pits. They lie in a forest near Kópháza, about 5 km South-East from Kányaszurdok. Here limonitic iron-ore was mined with shaft-working method.⁸

The Imola-type furnaces are built in the side of the working-pit. Since they have no breast-walls, they must have been operated with open breast. This is the feature in which they mainly differ from the Moravian ones. The workshop-complex at Szakony (W-Hungary)⁹ shows well the characteristics mentioned above (Fig. 2). The Imola-type is found all over the Carpathian Basin, mainly in the neighbourhood of early royal fortifications and county-seats. They can thus be considered as remnants of the royal iron-producing organisation. These workshops can be dated well both archaeologically and with the archaeomagnetic method to the 10-11th centuries.

The only example which does not correspond with the date mentioned above was found at Sopron-Május 1. tér. It has lying above the graves of the early Roman southern cemetery of Scarbantia, and was surrounded by later Roman (4th century A.D.) graves. Nevertheless it also belongs to the built-in type and closely resembles the furnaces of the Imola-type.¹⁰

This is why we tried to control the dating. It was made

difficult by the fact that the furnace was taken in situ to the Museum of Sopron. Therefore it was not possible to place dosimeters on the original site. The TL-results, according with the archaeological observations show that this furnace can not be dated as late as the 10th century, rather some centuries earlier. This is the only iron-smelting furnace in the present Hungary that may be dated back to the Roman times. Its shape and functioning raises the very difficult problem of the continuation of the Roman traditions of metallurgy.

Thermoluminescence dating

Minerals which can be found in potteries possess the ability to emit light at high temperatures, usually in the interval from 200 to 400^oC. This weak light emission, called thermoluminescence, is related to the release of trapped electrons from crystal lattice imperfections to lower energy levels. The previous trapping is caused by ionisation due to trace amounts of radioactive impurities. The inclusion technique, one of the methods of pottery dating by means of TL, is based on the phenomenon that mineral inclusions such as quartz and feldspars, which are embedded in the clay matrix, have a much higher sensitivity to "acquire TL" than the matrix itself. On the other hand, the TL observed is a measure of the cumulative dose of radiation to which the inclusions have been exposed since the previous heating. In the case of pottery the event dated is the firing by ancient man¹¹ (Fig. 3).

The accumulated radiation dose and hence the natural TL induced in mineral inclusions originate from alpha, beta and gamma radiations from uranium and thorium chains and potassium which are invariably present in the clay. When the pottery gets buried, radiation from the surrounding soil also adds to the continuous build-up. The shallow traps of the crystal lattice will continuously fade as even they are being continuously irradiated, but the deeper traps giving rise to TL peaks from 300 to 400^oC continue to build up without any fading.

In the quartz inclusion technique, radioactivity-free quartz grains of about 0.1 mm are usually separated from the

pottery. For this grain size fraction, the contribution of alpha radiation is negligible. Alpha particles from the grain/matrix interface have an average range of about 0.02 mm and the outer layer which is affected can be removed by treating the grains with hydrofluoric acid. On the other hand, the alpha particles are much less effective in inducing TL than the same dose of beta and gamma radiation.

Hence, for the extracted grain size fraction, the accumulated dose and the appropriate annual dose originate only from beta particles from the pottery matrix itself, from the gamma radiation of the burial soil and a small contribution from cosmic rays (Fig. 4).

From a measurement of the TL emitted by the quartz sample (natural TL, NTL) and by a proper laboratory calibration giving known artificial laboratory doses (artificial TL, ATL), one can estimate the archaeologically accumulated radiation dose. By comparing this latter with the annual dose (dose rate) received by the quartz grains, the absolute age can be given by the equation as follows:

$$\text{Age (in years)} = \frac{\text{Total dose since kiln-firing}}{\text{Annual dose}}$$

In this simple age equation, many complicating factors are involved. A number of inherent uncertainties imply a severe limitation in improving the accuracy of the TL age. The overall error is between ± 5 and ± 10 % of the age. However, TL is considered as a useful technique in archaeology, being potentially the unique tool to date a great variety of inorganic materials even beyond 50 000 years which is approximately the upper limit for radiocarbon dating. As far as findings of fired clay are concerned, in the present practice TL dating should be reserved for sites on which there is no material enough for radiocarbon or an ambiguity exists in the radiocarbon results.

To apply the above age equation, reliable and very reproducible NTL, ATL and dose-rate measurements are required. During the first period of our investigations (appr. ten years ago), TL measurements were performed with a Harshaw 2000A,B TL analyzer.^{12,13} As far as the main features of this appa-

ratus, in comparison with the standard model, are concerned, the max. temperature was extended up to 600°C, with the possibility of integration between any two predetermined temperatures. The optical filters can easily be interchanged. In addition to this, TL measurements required a higher flexibility in the heating program, too. This has been achieved by developing an appropriate circuitry for preheating.

From 1985 on, the Daybreak photon counting system has been used. This apparatus is designed specifically for TL research in natural materials. It is modular expandable and compatible with a wide range of system components. The temperature controller has an extremely stable, reproducible digital ramp for heating rates 0-25°C, automatic repeat ramp for the background glow curve, a low-power, low-volume glow oven for fast cooling and evacuation, and a pile-up compensating ratemeter that extends the single photon counting dynamic range to beyond ten million counts per second. The system is packaged in two parts: an electronic enclosure and the glow oven assembly with PMT-housing (the PM tube is an EMI 9635QA selected for very low dark count). The glow curves are recorded with a Canberra multichannel analyzer and then evaluated numerically.

As the potteries of prehistoric origin in the Carpathian basin are extremely poor in quartz grains, a special sample preparation technique was developed to get a higher yield in grains. The occurrence of the grains in the appropriate size interval was found to be as low as a part of 10^{-4} of the whole material or less. In our procedure the sherds were crushed to fragments of appr. 0.5 cm, divided into small portions and subjected to repeated attacks in HCl and HF by making use of an ultrasonic bath. The clay matrix was successively eliminated by rinsing in distilled water. The complete decomposition of the fragments was followed by a continuous etching of the residual grains in HF for an hour. After sieving, the fraction from 0.09 to 0.125 mm was used for the TL measurements. Alternatively, the procedure was complemented with a magnetic separation by means of a permanent magnet.

In our standard glow-out technique 2 mg portions of quartz were uniformly spread, as fairly loose monolayers, on stainless steel cups.

Laboratory irradiations were carried out with a beta irradiator (Daybreak model) containing a strontium source of about $2 \cdot 10^9$ Bq activity. Its calibration was performed with a gamma source at the National Office of Measures. In our standard onplate irradiation procedure, the dose delivered per minute to quartz by the beta source quoted above is 2.49 Gy.

Typical glow curves can be seen in Figs. 5 and 6. The principle of the additive technique for the assessment of the total dose (Q+I) is schematized in Fig. 7. For dose-rate measurements thermoluminescence itself was used. In TL dosimetry for radiation protection purposes, a number of phosphors are available which can measure doses as low as 0.01 mFy or less. One of them, synthetic calcium sulphate activated by dysprosium, was selected for measuring gamma dose-rate from the burial soil and beta dose-rate from the clay matrix of the pottery.

In the case of the soil, a copper capsule of wall thickness 1 mm is sufficient to stop beta particles from reaching the phosphor. After filling the capsule with about 0.1 g of $\text{CaSO}_4 : \text{Dy}$ it is made watertight and then inserted into the soil in as similar a situation as possible to that from which the pottery fragment was removed. It is preferable to leave the capsule buried for a year in order to even out seasonal fluctuations of dose-rate which are due to variations in water content.

For measurement of beta dose-rate from the clay matrix, the $\text{CaSO}_4 : \text{Dy}$ phosphor is contained in a polycarbonate tube (internal diameter 1 mm) which is inserted into a small plastic container filled with the powdered sample. By shielding the container against external radiation, after a storage of about a month the TL emitted by the phosphor can accurately be measured. Alpha particles are prevented from reaching the phosphor by the 0.2 mm thick wall of the tube. By appropriate calibrations with samples of known radioisotope concentrations, the correction factor, which is mostly due to beta attenuation in the wall, was determined (the true dose within the sample is appr. two times the phosphor dose).

The beta dose-rate is measured on dry material, therefore it is to be corrected for the water content of the sample in burial conditions. Wetness is expressed in terms of the saturation water content (W) and the actual value of D_{beta} is given by

$$D_{\text{beta}} = \frac{D_{\text{beta dry}}}{1 + 1.25 \cdot WF}$$

where the factor of 1.25 accounts for the higher beta absorption of water relative to that of the sample. F denotes the fraction of saturation to which the average water content corresponds. In this respect, there is a significant uncertainty, hence $F = 0.8 \pm 0.2$ is assumed.

The annual dose is influenced by the radon emanation, too. The escape of the gaseous member of the uranium chain can have a substantial effect on dose-rate and we are limited to rough estimations as far as the emanation extent in the true burial circumstances is concerned. However, in most cases alpha counting proved to be a convenient laboratory approximation. Crushed samples were measured in sealed and unsealed condition as well. When precise dating is required, only those samples should be accepted for which the sealed count-rates do not exceed by more than 10 % the unsealed ones.

When several samples from a context are investigated, the best value for the age of the context is obtained by weighting the individual ages. This has been done for the site of Tiszapolgár-Bastanya.¹⁴ A review of these investigations will be presented in the next future.¹⁵ Preliminary results, suggesting two distinct archaeological periods, have already been described in earlier papers.^{16,17,18} Recent works are in progress to date some other neolithic and copper age cultures in Hungary (Csószhalom, Bodrogzsádány, Tizzaszölös) and in Yugoslavia (Vučedol).

TL dating in industrial archaeology

During the last ten years, several fragments from ancient furnaces and hearths were investigated. Oppositely to the prehistoric potteries, these findings are rich enough in quartz grains, so the sample preparation can easily be performed. In

most cases, unfortunately, large mineral inclusions and various inhomogeneities can also be found in and around the fired material. The true dosimetry situation is strongly affected by these local inhomogeneities which represent a severe limitation in the accuracy of the annual dose. Therefore, in industrial archaeology, the overall uncertainty of TL age amounts to 10 - 20 %, depending on the properties of the context.

Only some of the results are given here (years BP; see also in Fig. 8):

- Iron-smelting hearth, Tarjánpuszta (Fig. 9,10), sample ref. 79/22. TL age: 1400 years (\pm 25 %) ¹⁹

- Smelting-hearth, Kányaszurdok, sample refs. 2/80 and 4/80. TL ages: 1190 (+20 ... - 10 %) and 1260 (+ 20 ... - 10 %) years, respectively ²⁰

- Lime kiln, Pilisszántó. TL authentication: 17th to 18th century ²¹

- Hearts: ²¹

sample ref.	probable TL age (years)
G1/80 (Tarjánpuszta)	1130-1200
G2/80 (Kányaszurdok)	1190
G3/80 (Kányaszurdok)	1260
G4/80 (Kányaszurdok)	1250

- Iron-smelting furnace (Sopron). TL authentication: 10th century or older ²²

- "Red earthwork", Sopron. TL age: 835 years (\pm 12 %) ²³

- Zamárdi, single TL dates for five furnaces: 1440, 1650, 1200, 1150 years (average: 1280 years).

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Jednou z metod, jíž jsou datovány výsledky výzkumů v Západním Maďarsku ze posledních 15 let je termoluminiscenční metoda. Zkoumá objekty železářských pecí podobných pecím z velkomoravského a hradištního období, jak je známe z výzkumů V. Souchopové na Blanensku. V Maďarsku jsou nazývány podle místa jejich prvních nálezů jako typ Nemeskér - jde o šachtovou volně stojící pec s dyšnovou cihlou v hrudi pece a jámou pro jímání tekuté strusky. Vývoj tohoto typu pece sahá až do období Avarského panství např. Tarjánpusta v západním Maďarsku, ale také jižně od Balatonu v Zámárdi. V prvním případě jsou zbytky pecí vtroušeny do chaoticky uspořádaného osídlení, v Zámárdi byla odkryta huť, v níž lze sledovat celý výrobní proces: redukční šachtové pece jsou obklopeny oválnými pražicími pecemi, milířišti a vyhřívacími výhněmi, z jejichž vyzdívek byly odebrány vzorky pro datování. Byly zjištěny kúlové jamky naznačující, že část pracoviště byla zastřešena.

V Šoproni - Kányaszurdoku byly zjištěny tři šachtové pece s kamennou obezdívkou šachty, postavené těsně při sobě se společnou jámou pro strusku. Jsou datovány do 9.-10. století a představují vyvinutý základní typ Nemeskér. V tomto případě bylo patrně nalezeno i hornické dílo, v němž byla těžena železná ruda pro huť. Jde o důlní pole, v němž se zachovalo na 227 válcovitě zakloubených jam. Pece typu Imola jsou vždy zahloubeny do okraje pracovní jámy. Proces byl řízen otevřenou hrudí pece. Tento typ pece se nachází v celé Karpatské kotlině a pece byly datovány jak pomocí archeologického materiálu, tak archeomagnetickými metodami do 10. - 11. století. Nacházejí se většinou v blízkosti královských hradů a lze předpokládat, že jejich činnost byla organizována královskými úředníky.

Jedinou lokalitou, jejíž datování neodpovídá uvedenému datování, je lokalita v Šoproni (Május l.tér), které leží nad hroby raného římského pohřebiště v Scarbantii a je obklopena mladšími římskými pohřby ze 4. stol.n.l. Tato skutečnost byla důvodem k novým měřením, které do jisté míry nepříznivě ovlivnilo přenesení pece do Šoproňského muzea. Termoluminiscenční metoda vyloučila datování do 10. století, naopak posunula stáří pece o několik století nazpět a nevyloučila možnost římského původu. Otázka římské tradice v metalurgii železa a konstrukci pecí však vyřešena nebyla.

Metoda termoluminiscenčního datování je založena na vlastnosti keramických hmot imitovat záření za vyšší teploty obvykle v intervalu 200 - 400°C. Slabé světelné záření zvané termoluminiscence je spojeno s uvolňováním elektronů v defektu krystalické mřížky na nižší energetické hladiny. Předchozí zachycení je způsobeno ionisací vyvolanou stopovými množstvími radioaktivních nečistot. Další z metod Termoluminiscence je metoda vměstková, která je založena na jevu, že minerální vměstky křemene a živců, které jsou zachycené ve struktuře jílu mají mnohem vyšší citlivost k vyvolání termoluminiscence, než vlastní struktura jílu.

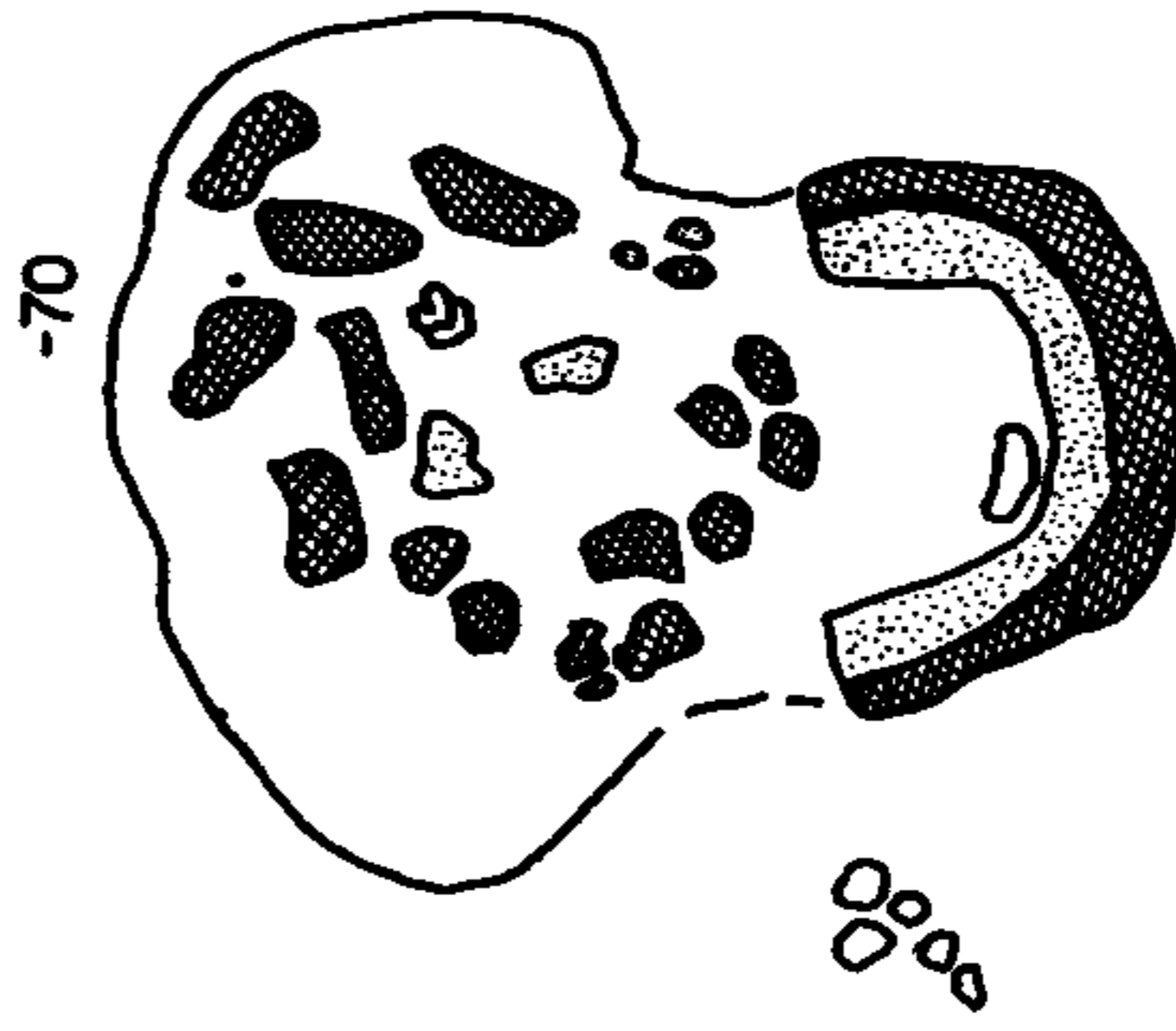
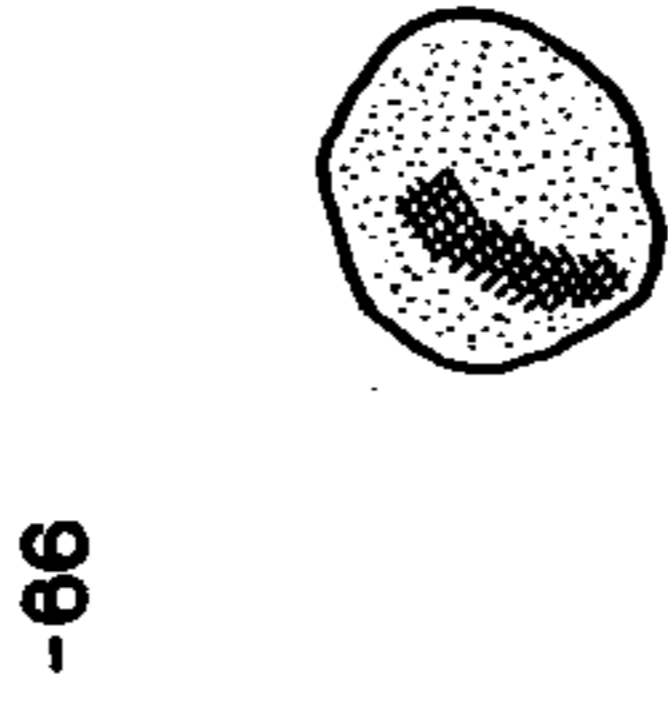
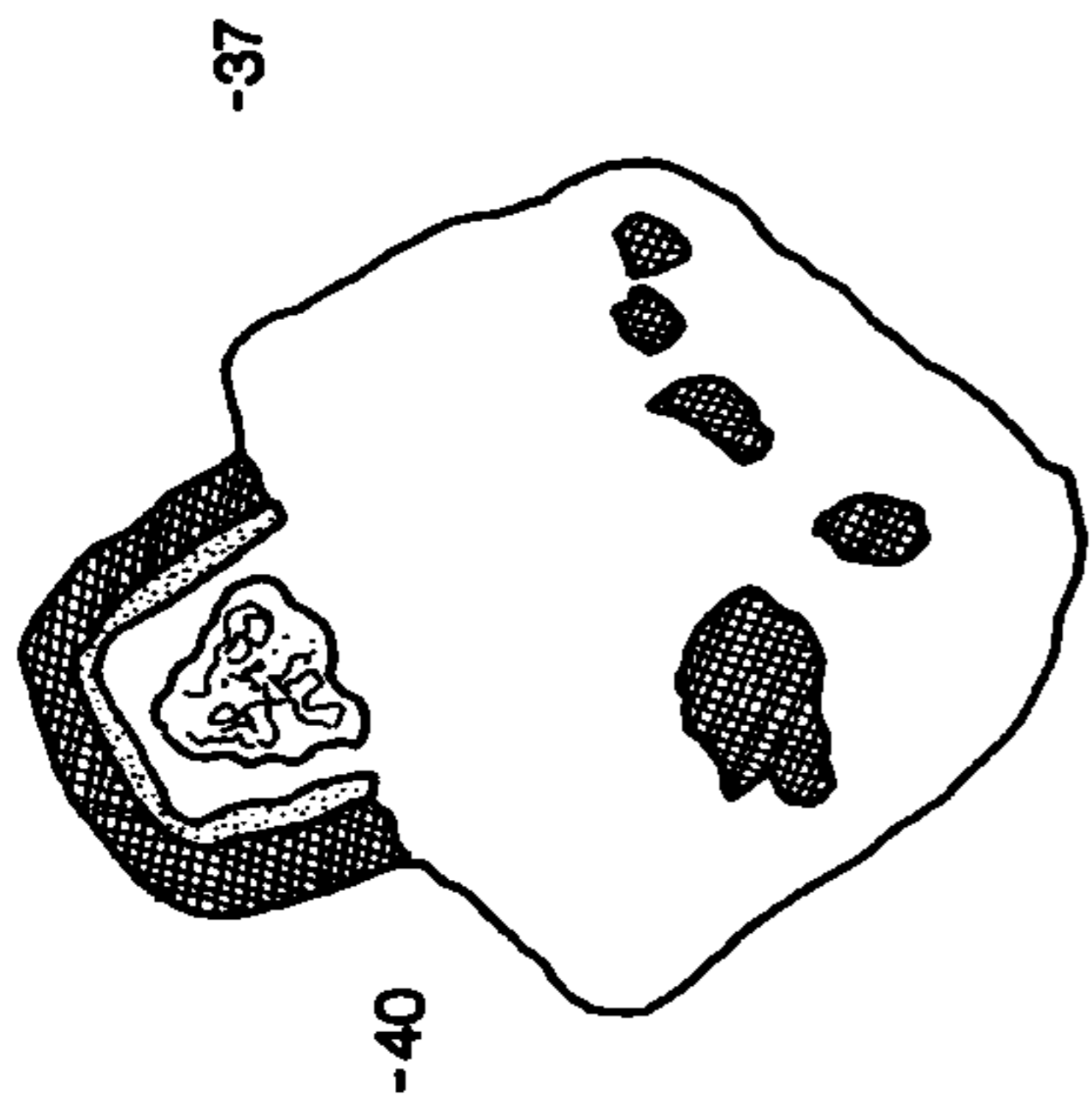
Na druhé straně termoluminiscence je mírou kumulované dávky radiace, již byly vměstky vystaveny od předchozího vypálení. Další metodou je metoda vměstků křemene. Spočívá v tom, že z keramiky oddělí zrna křemene o velikosti kolem 0,1 mm, které jsou prosté radioaktivity a zjišťuje podíl záření alfa, beta a gama vyvolaného okolní hmotou. Zjišťuje přírodní termoluminiscenci a podle kalib. rační křivky (umělé luminiscence) určí archeologickyakumulovanou radiační dávku. Při srovnání této dávky s ročním poklesem radioaktivity pro křemenná zrna lze absolutní věk vyjádřit vztahem:

$$\text{věk/rok} = \frac{\text{totální dávka od vypálení}}{\text{roční dávka}}$$

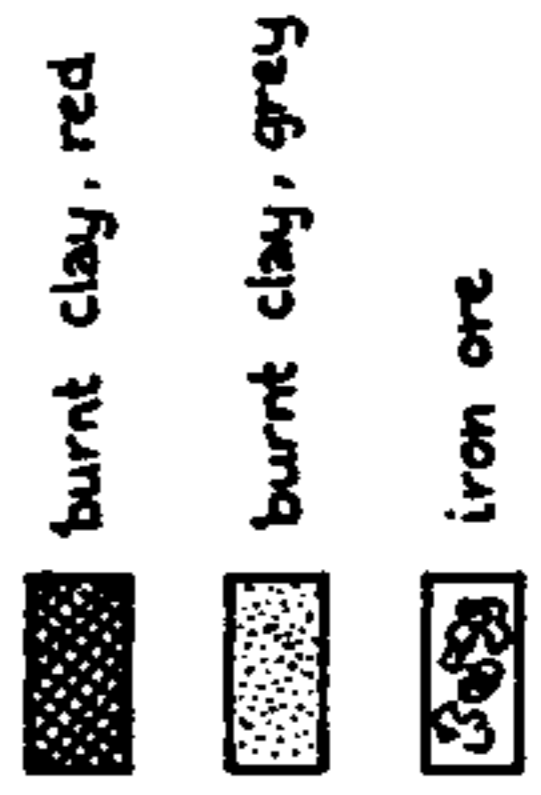
Protože prehistorická keramika z karpatské oblasti je extrémně chudá na zrna křemene, byla vyvinuta speciální technika na získání většího množství (výtěžku) zrna. Jsou podrobně popsány potřebné pracovní operace a metody a zveřejněny výsledky datování archeologických objektů.

- Fig. 1** Tarjánpuszta-Vasasföld II., ground plan of furnaces No. 79/21, 79/22 with a limestone heap (down, left) and a reheating hearth (right), 7-8th centuries
- Fig. 2** Szakony, ground plan and section of workshop No. 6, 11th century
- Fig. 3** Principles of TL-dating
- Fig. 4** Contributions to the dose-rate
- Fig. 4** Contributions to the dose-rate
- Fig. 5** TL-glows and plateau-test (sample No. 53.35.116 from Tiszapolgár-Basatanya), asymmetry is indicative of possible fading
- Fig. 6** TL-glows and plateau-test (sample No. 53.35.116 from Tiszapolgár-Basatanya) after a pre-heat at 320°C
- Fig. 7** Determination of the palaeodose (sample No. 53.35.116)
- Fig. 8** Comparative table of the ages of industrial archaeological sites dated by different physical methods
- Fig. 9** Tarjánpuszta-Vasasföld. Sherds found at furnace No. 1.
- Fig. 10** Tarjánpuszta-Vasasföld II. Sherds found between the iron-smelting furnaces

Fig. 1



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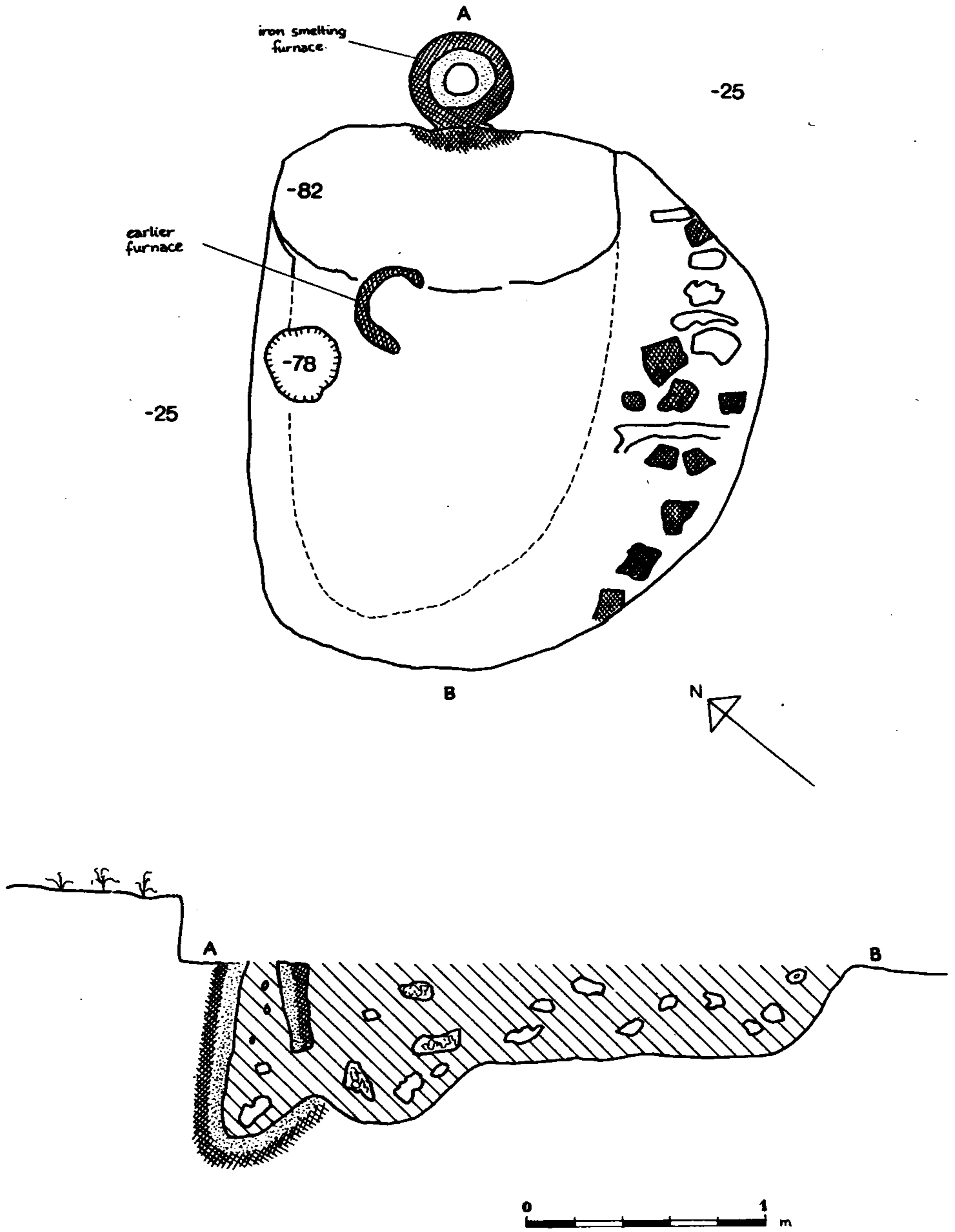


Fig. 2

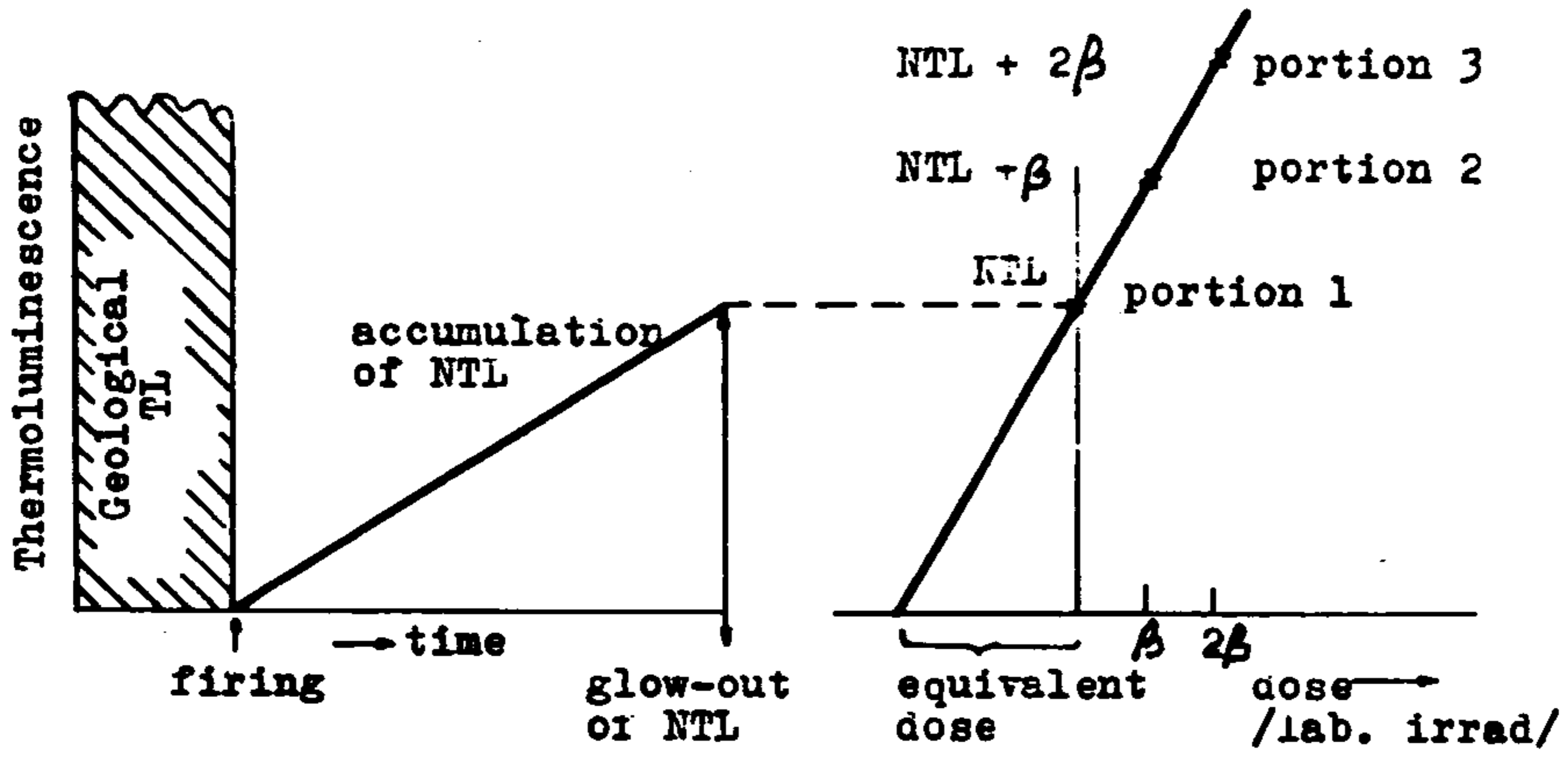


Fig. 3

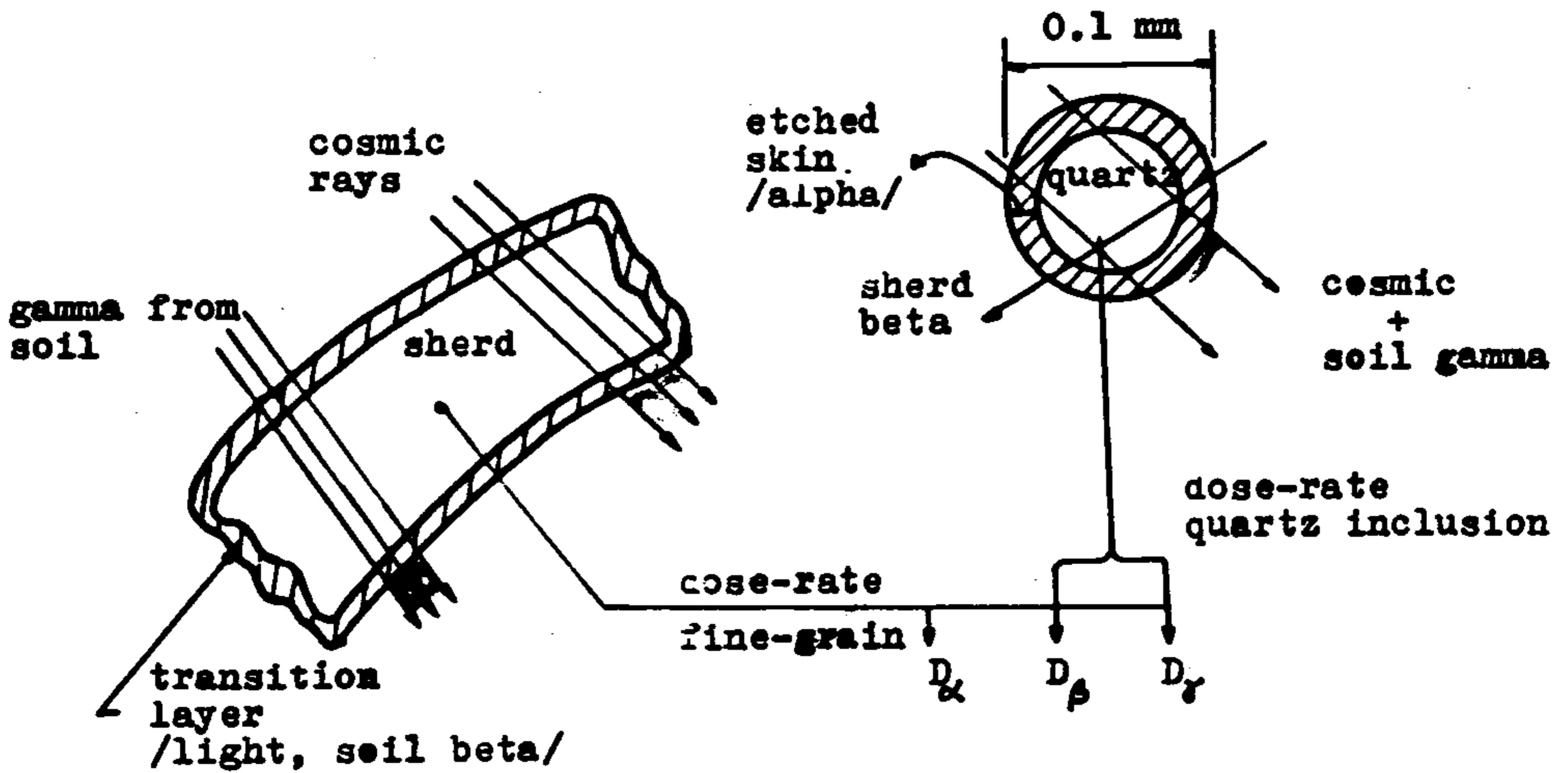


Fig. 4

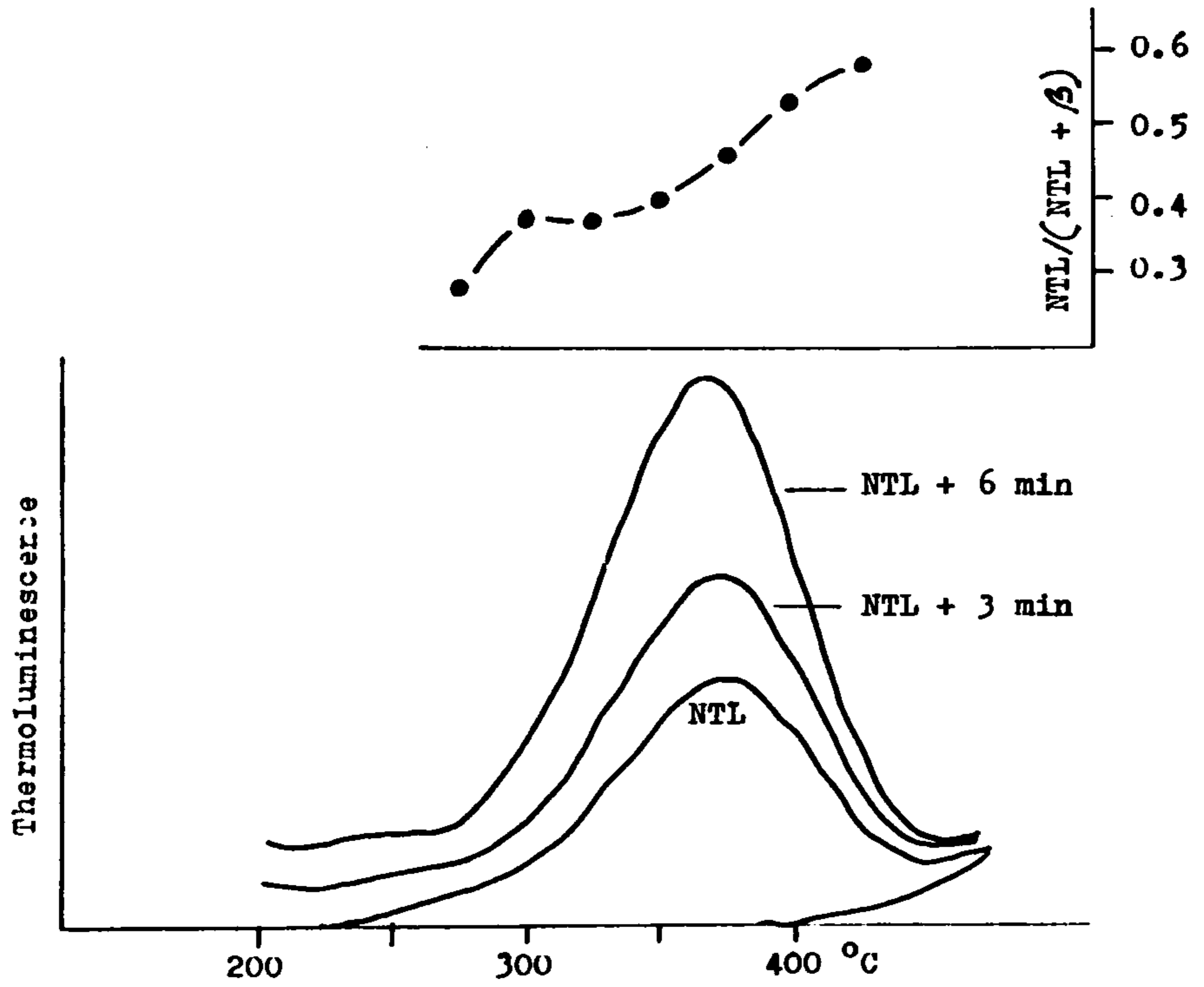


Fig. 5

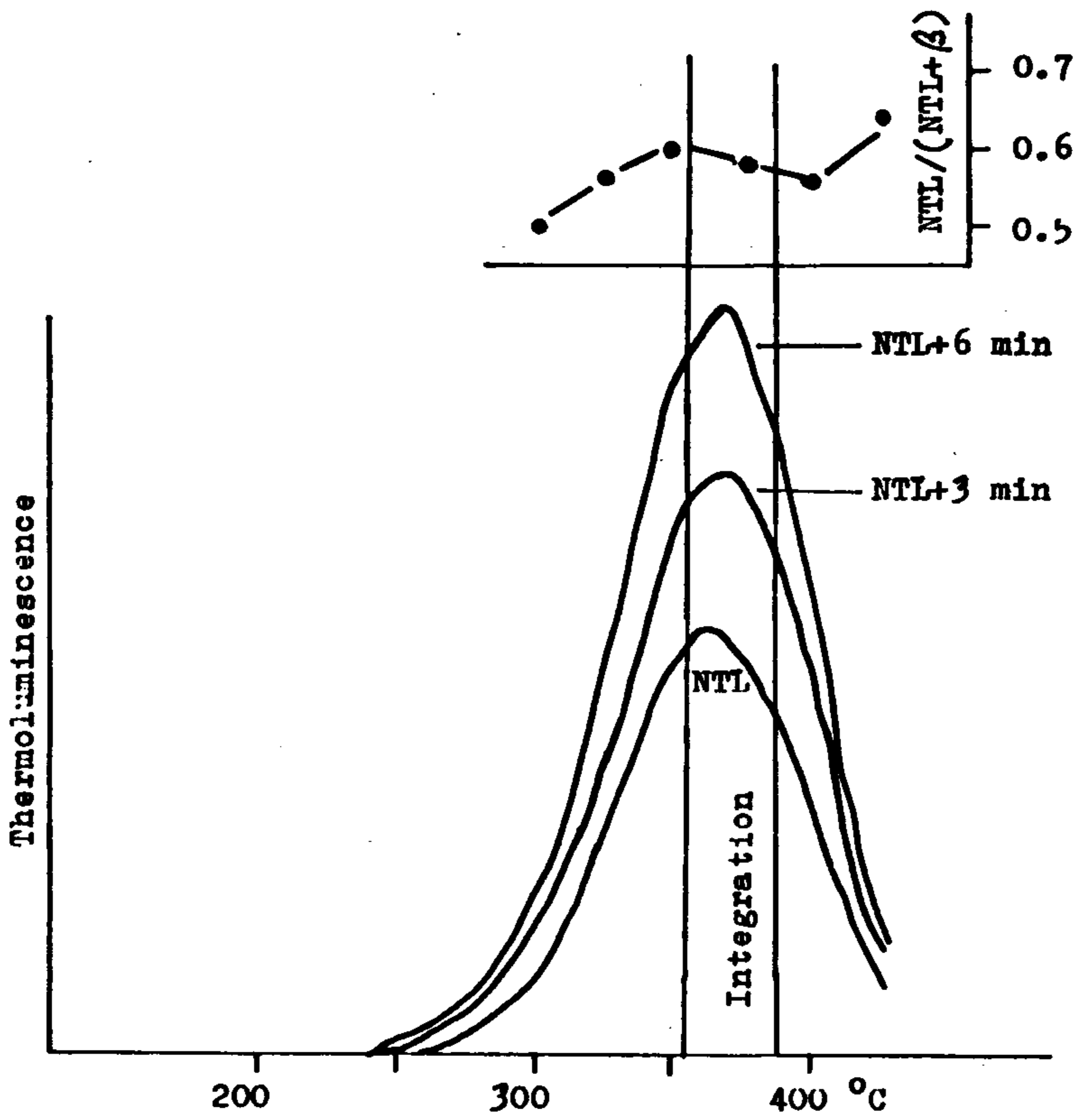


Fig. 6

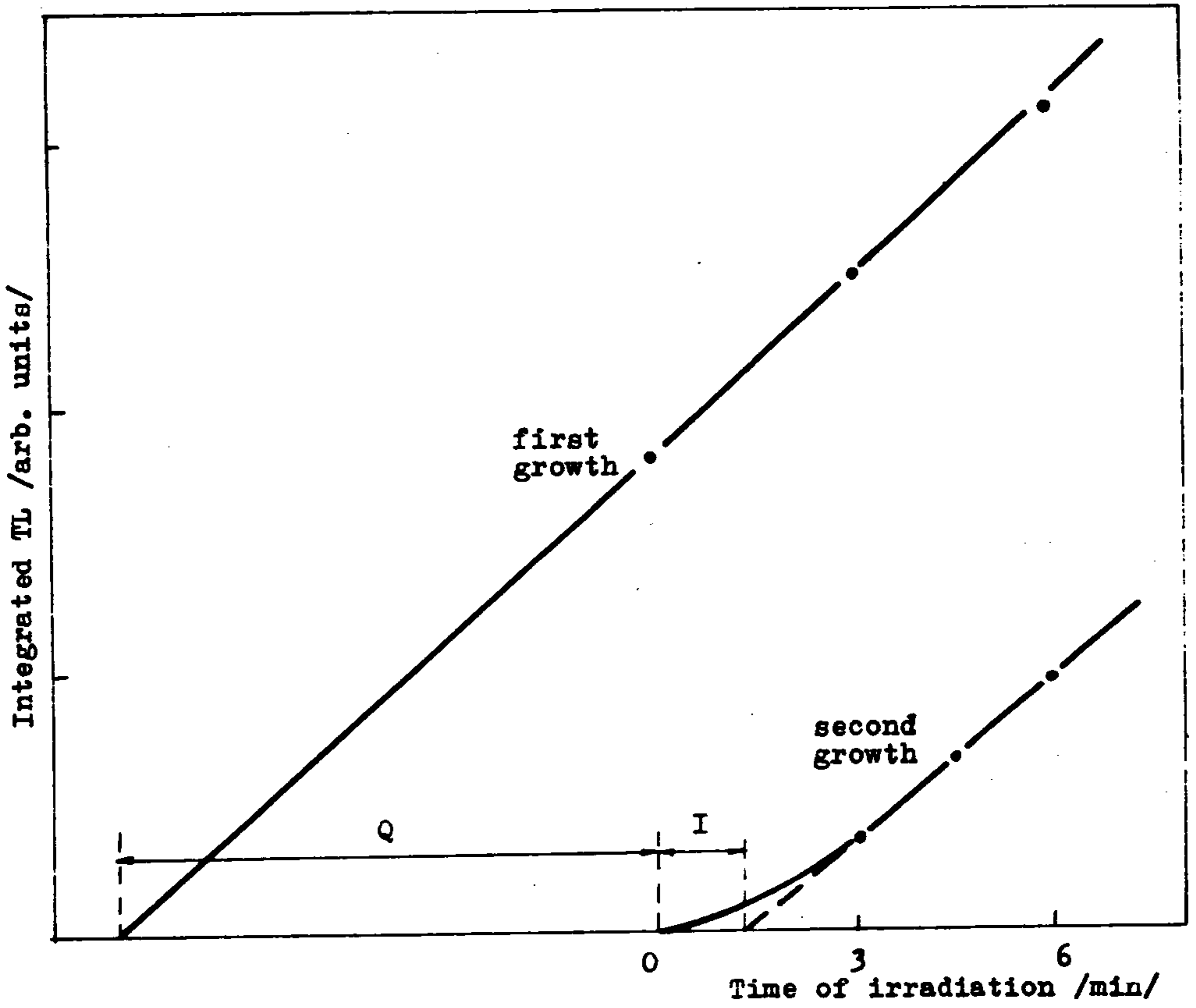
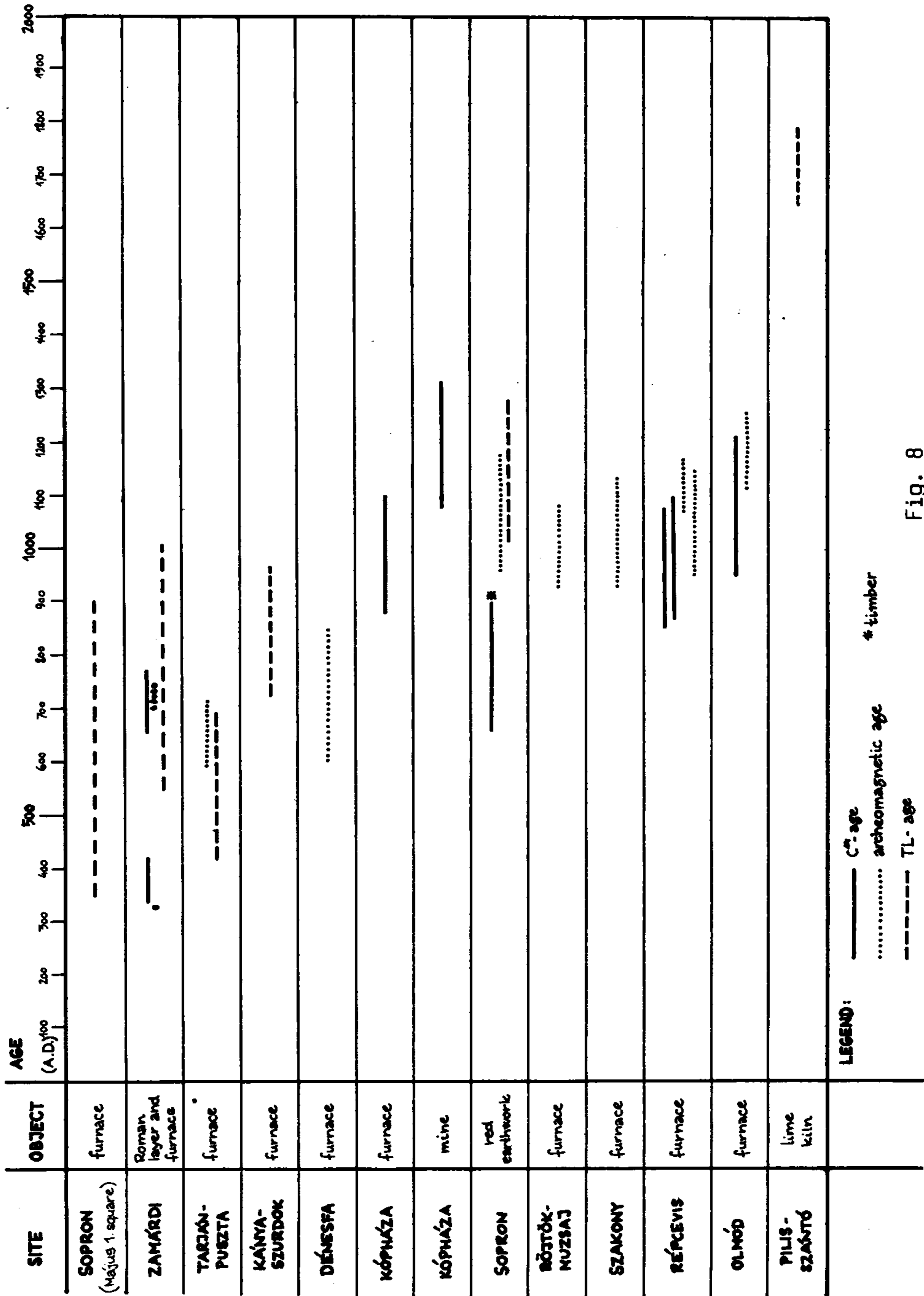


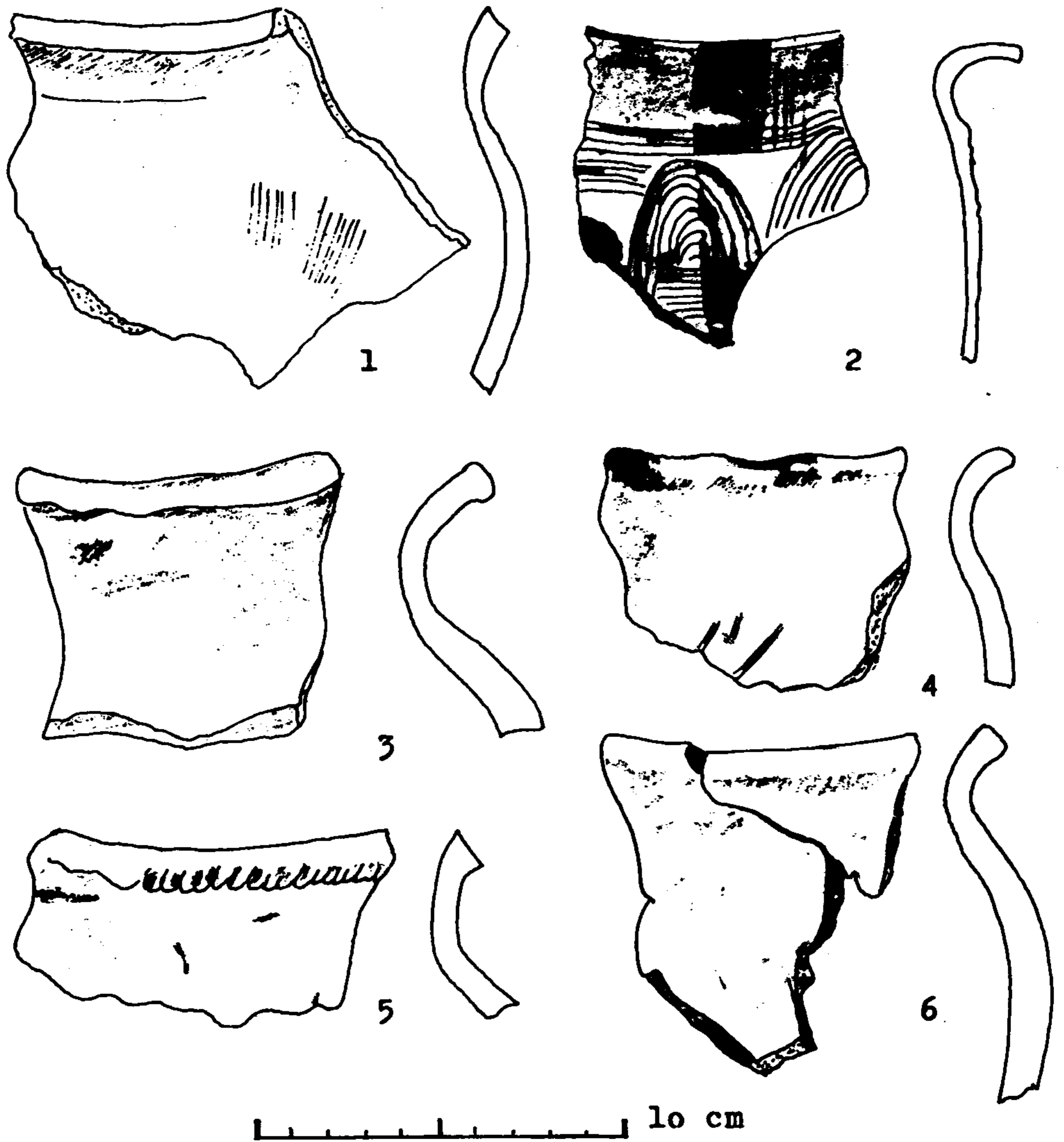
Fig. 7



LEGEND:

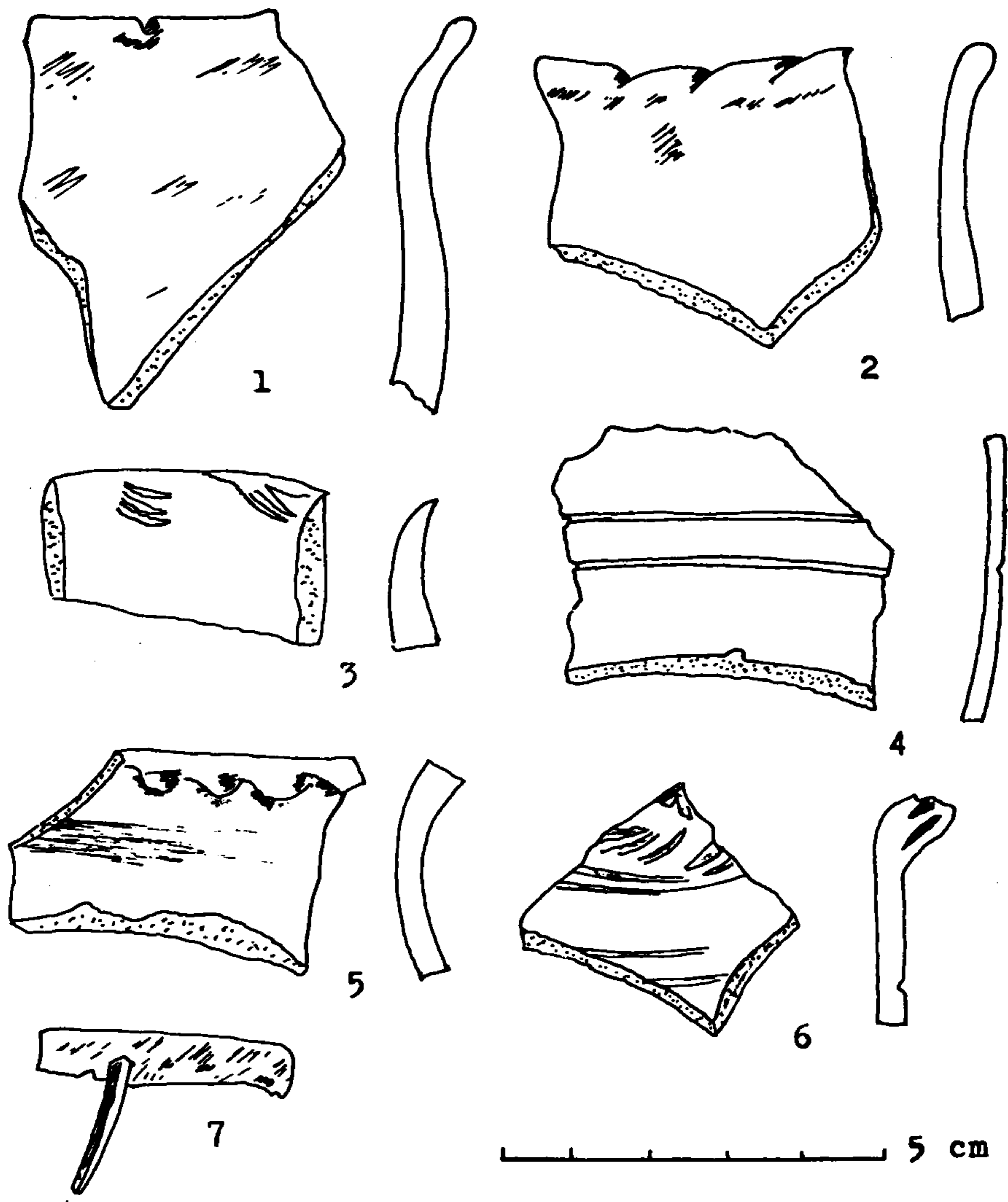
- C¹⁴ age
- archeomagnetic age
- TL age
- * timber

Fig. 8



Tarjénpuszta , Vasasföld
M:1:2

Fig. 9



Tarjánpuszta, Vasasföld II. M:1:1

Fig. 10