

Thermoluminescent Determination of Prehistoric Heat Treatment of Chert Artifacts



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Science, New Series, Volume 197, Issue 4311 (Sep. 30, 1977), 1359-1362.

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produced during my simulations. The pebbles and new flakes were lithologically similar to the Sozudai material. The new flakes were also stained to make them appear similar to the weathered Sozudai flakes.

The test sample of 18 pieces was shown separately to a total of 27 (11 Japanese and 16 American) archeologists who claimed to be familiar with flake characteristics. The observers were asked to identify the flakes in the test sample. On each piece identified as a flake the observer had to indicate the point of percussion, the bulb of percussion, and the ventral surface. Three of the new flakes served as a control group. If any of these flakes were not accurately described, the observer's opinions on the rest of the samples were disregarded. Only 19 observers correctly identified all of the flakes in the control sample. All of these observers also correctly identified all of the newly made flakes.

All of these 19 archeologists identified at least one of the Sozudai pieces as a flake. Only two of them, however, considered all five of the Sozudai pieces to be flakes. Since each of the 19 observers was presented with five pieces from Sozudai, a total of 95 judgments were elicited. Of these judgments, 61 were in exact agreement with the identifications reached during my own flake identification procedure. Observers were presented with a small, randomly selected sample of heavily weathered Sozudai flakes. Furthermore, these pieces were not presented in the context of a total assemblage and the observers were called on to make their judgments on the basis of only relatively brief examination. It is, therefore, not surprising that their judgments were not in total agreement with mine which were based on long, detailed examination of the entire Sozudai collection. A χ^2 test applied to the results of the observations, however, does indicate a strong positive correlation between my identifications and those of other observers ($\chi^2 = 7.66$, $p < .01$, d.f. = 1). My conclusions on the Sozudai flake sample thus are substantiated by observations of other trained archeologists.

Thus, although some Japanese and American archeologists have questioned the human creation of the Sozudai assemblage, an independently substantiated test procedure indicates that at least 13.6 percent of the assemblage is composed of flakes. Furthermore, when the characteristics of these flakes are compared to the criteria established by A. S. Barnes, the Sozudai assemblage

seems to be clearly of human rather than natural origin. Since the Sozudai assemblage clearly predates Upper Paleolithic industries and probably dates from the early phases of the last glacial period, these conclusions strongly support an early human occupation of southern Japan.

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References and Notes

1. F. Ikawa-Smith, *Asian Perspect.* **18**, 15 (1975).
2. H. Oi, *Hoppo Bunka Kenkyu* **3**, 45 (1965); S. Sugihara, *Shinano* **19**, 245 (1967); V. J. Morlan, *Arct. Anthropol.* **8**, 164 (1971).
3. C. Serizawa, *Kagaku (Kyoto)* **39**, 28 (1969); C. Serizawa, *Proc. 8th Int. Congr. Anthropol. Ethnol. Sci.* **3**, 353 (1970); in *Early Palaeolithic of South and East Asia*, F. Ikawa-Smith, Ed. (Mouton, The Hague, in press).

4. H. Oi, in *Early Palaeolithic of South and East Asia*, F. Ikawa-Smith, Ed. (Mouton, The Hague, in press); V. J. Morlan, *Arct. Anthropol.* **8**, 141 (1971); S. Sugihara, *Kokogaku Janaru* **8**, 2 (1967).
5. C. Serizawa, in *Nihon Bunka Kenkyujo Kenkyu Hokoku* (Tohoku University, Sendai, 1965), vol. 1, pp. 1-119.
6. J. D. Speth, *Am. Antiq.* **37**, 34 (1972); A. Tsirk, *ibid.* **39**, 128 (1973).
7. A. S. Barnes, *Am. Anthropol.* **41**, 99 (1939).
8. R. Ascher and M. Ascher, *Science* **147**, 243 (1965).
9. F. Ikawa-Smith, thesis, Harvard University (1974), pp. 302-304.
10. H. Nakagawa, in *Nihon Bunka Kenkyujo Kenkyu Hokoku* (Tohoku University, Sendai, 1965), vol. 1, pp. 121-141.
11. C. Serizawa, in *ibid.*, pp. 1-119.
12. H. Kobayashi, *Kokogaku Janaru* **78**, 8 (1973).
13. I began this research while I was a Japan Foundation Fellow at Tohoku University. I thank Prof. C. Serizawa, who generously made the Sozudai collection available and freely shared his knowledge of Japanese prehistory. I also thank H. Kobayashi and M. Okamura for their help and support. Dr. A. Bleed also gave valuable help with the framing and execution of the research.

18 January 1977; revised 23 May 1977

Thermoluminescent Determination of Prehistoric Heat Treatment of Chert Artifacts

Abstract. *In recent years archeologists have become interested in the extent to which prehistoric peoples heat-treated chert prior to shaping it into tools. Thermoluminescent determination of the radiation dose accumulated by an artifact since it was formed or last heated provides a simple, reliable test for such heat treatment. This test can be applied to single artifacts without the need for raw source material for comparison. Results on 25 artifacts from four sites indicate that, for many chert sources, color and luster are not useful indicators of heat treatment by prehistoric peoples.*

Chert is the most common raw material in the lithic technology of prehistoric peoples. Various accounts of the use of heat treatment in the manufacture of tools by primitive peoples have been reported since at least the turn of the century (1), but the lack of success in duplicating these techniques led to a general skepticism which prevailed for many years. However, in 1964 Crabtree and Butler (2) reported that, when raw chert was slowly heated and slowly cooled in sand, marked improvements in the knapping properties resulted.

In recent years the belief that prehistoric people sometimes heat-treated chert before shaping it into finished tools has gained support and generated interest in finding a simple, reliable test for heat treatment. A number of techniques have been applied in attempts to determine whether particular chert artifacts were heated by prehistoric people. Probably the most common has been an examination of the visual appearance. A pink or reddish color or a vitreous luster, or both, are often taken as evidence of heating. Many forms of chert display these characteristics upon heat treatment (1, 2). However, in addition to

being a subjective test, these characteristics may also result from nonthermal effects. Raw chert, even within a single outcrop, often varies greatly in color with reds and pinks produced, for example, by exposure to groundwater containing iron; a vitreous luster on a tool may be caused by polishing of the surface from repeated use.

More sophisticated techniques have been used to provide evidence of heat treatment, including electron microscopy and x-ray diffraction. Electron microscopy indicates that high temperatures bring about changes in the microcrystalline quartz structure of chert (1, 3). Weymouth and Mandeville (4) reported x-ray diffraction line-broadening in some (but not all) heat-treated cherts, presumably due to the breakup of the original crystals into smaller crystals or to local strains. However, both these measurements require raw source material for comparison, and the authors concluded that neither provided a reliable, independent test.

In view of the success of thermoluminescence (TL) dating of archeologic ceramics (5), it has been proposed that heat-treated chert could also be dated in

Table 1. Geologic cherts studied.

Geologic source location	Chert formation	Number of controlled test samples	
		Total	Heated
Near Koster, Illinois	Burlington	2	1
Northwestern Iowa	Tongue River	8	4
Dordogne Valley, Peridgord, France	Unknown	4	0

a similar way (6). Chert, however, presents a number of practical difficulties which make accurate dating by TL difficult (7). On the other hand, it should be relatively easy in principle to distinguish between raw unheated chert and chert heated in prehistoric times; an unheated sample should give a much larger TL signal than a recently heated sample. Rowlett *et al.* (8) attempted this determination but found only a difference of a factor of 2 in TL intensity between raw source material and chert heated in the laboratory just prior to measurement. This small difference meant that they could only establish heat treatment in artifacts for which they had raw material with very similar TL sensitivity, a serious limitation because of the large variability in chert samples often found even within a single geologic source. However, as we show here, for all the cherts we have measured with our techniques, the TL from raw chert is typically 100 times greater than the TL from chert heated within the last 5000 years, and the distinction can be reliably made.

The basic principle and techniques used are closely related to those of TL dating (5). Thermoluminescent measurements are used to determine the natural dose that has been accumulated by the sample since it was formed or last heated. We base our technique on measurement of the natural dose rather than the natural TL alone because the natural TL of cherts with similar ages can vary over several orders of magnitude, especially if the cherts are from different sites. This is so because the TL sensitivity is strongly dependent on trace element concentrations, and therefore extremely variable. On the other hand, the natural dose rates vary much less and thus the natural doses of cherts of the same age are consistent within an order of magnitude.

One determines the natural dose by first measuring the natural TL and then measuring the TL produced by a known dose of radiation. The equivalent dose, defined as

$$\frac{\text{Natural TL}}{\text{Artificial TL}} \times \text{Artificial dose}$$

is taken as the measure of the natural dose (9). Raw source material will have received a very large equivalent dose since its formation. In most cases the TL is saturated or near saturation, and the equivalent dose, equal to the saturation dose, is typically 10^5 to 10^6 rads. If a chert has been heated to more than $\sim 400^\circ\text{C}$ in prehistoric times, the equivalent dose is reset to zero. The sample will then begin to reaccumulate a radiation dose but, as we show below, for archeologic times, the reaccumulated dose is much less than, and easily distinguishable from, the saturation dose in an unheated sample.

The apparatus is essentially the same as that used in the dating of archeologic ceramics and is described in detail elsewhere (10). The basic system consists of a Nichrome heating plate heated linearly at 7°C per second in a glow oven which is first evacuated and then filled with oxygen-free nitrogen. The TL is detected by an EMI bi-alkali photomultiplier tube with a Corning 5-60 filter and measured with a picoammeter. Artificial irradiations are carried out with a 1-curie ^{90}Sr beta source.

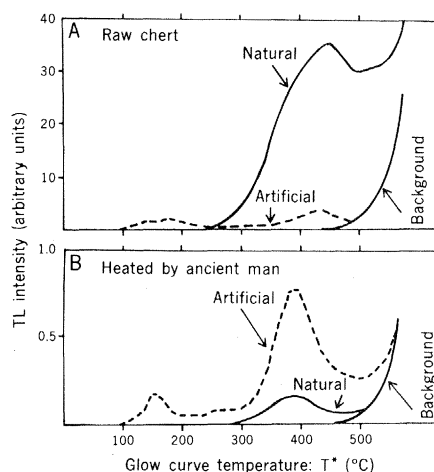


Fig. 1. Natural and artificial (6000-rad beta dose) glow curves of (A) a raw chert from Tongue River and (B) an artifact from Koster apparently heated in prehistoric times (~ 8000 years ago). Chert samples heated in the laboratory in the controlled test gave curves indistinguishable from the background curves shown, composed of photomultiplier tube noise and blackbody radiation.

The artifacts tested in this program typically had dimensions of one to several centimeters. In the sample preparation we followed the technique developed by Göksu *et al.* (6). A low-speed, diamond-impregnated wafering blade was used to cut small slabs, approximately 0.5 by 4 by 4 mm, from the interiors of the artifacts. Since lithic fragments are probably exposed to a considerable amount of sunlight both during their use by ancient man and in their excavation by archeologists, it has been suggested that optical bleaching might reduce the TL intensity (8). However, we found that a 5-hour exposure to bright sunlight induced a large TL signal in several samples corresponding to an equivalent dose of 10^3 to 10^4 rads. For this reason, it is critical that samples tested for heat treatment do not include near-surface material. The transparency of the cherts studied was 0.01 to 0.02 percent per millimeter; consequently, samples were cut at least 2 mm from the surface of the artifact. Because of this effect, small samples less than ~ 3 mm thick are probably not suitable.

The TL method was evaluated on 14 cherts from three geologic sources (Table 1). Half of the samples from Tongue River, Iowa, and Koster, Illinois, were heated by J. Weymouth of the University of Nebraska in a "blind test." Figure 1A shows typical glow curves. In all of the samples measured, the distinction between the unheated cherts, which gave glow curves corresponding to a saturation dose, and the heated cherts, which gave signals indistinguishable from the background curve of Fig. 1A, was very clear. We obtained the correct answer on all of the "blind test" samples. The two most important figures in evaluating the TL method are (i) the minimum detectable dose and (ii) the dose at which the TL saturates. For the cherts tested, the minimum detectable doses ranged from 7 to 70 rads, and the saturation doses from 10^5 to 10^6 rads with the exception of one sample that saturated at $\sim 10^4$ rads. At a glow curve temperature of 450°C , the difference between the raw and heated cherts was a factor of $\sim 10^2$ to 10^5 .

A chert heated in prehistoric times will have reaccumulated an archeological dose. The natural dose rate will be composed of the external radiation of gamma rays plus cosmic rays, and an internal dose rate from beta and alpha particles. The external dose rate will generally be in the range 0.05 to 0.2 rad per year (5). The uranium contents of a variety of cherts were found to be 0.3 to 10 parts

per million, with alpha efficiencies (I_1) of 0.03 to 0.2, giving an effective internal dose rate of 0.01 to 0.35 rad per year. Thus the total effective dose rate is in the range ~ 0.05 to 0.50 rad per year. For this range of total dose rate for samples $< 10,000$ years old, the reaccumulated dose is < 5 krad, which is much less than typical saturation doses. Therefore, any occurrence of prehistoric heating should be clear.

Chert artifacts from four archeologic sites (Table 2) were tested for ancient heat treatment. All the artifacts from Missouri and Bolivia showed natural doses greater than 40 and 80 krad, respectively, a clear indication that these eight artifacts were unheated in prehistoric times. Their glow curves were similar to the curves labeled "natural" and "artificial" in Fig. 1A. Some of the cherts from Koster and Cibola, New Mexico, gave similar glow curves, but others had much smaller natural TL (Fig. 1B) and are apparently unheated.

The data are more fully understood as a graph of equivalent dose versus glow curve temperature (T^*) (Fig. 2). The upper hatched regions represent the results for the raw source material, and the lower hatched regions represent the lower limit of detectable signals and include results for the five control samples heated in the laboratory in the "blind test." Artifacts heated by prehistoric man were expected to fall somewhere in between, depending roughly on their archeological ages (time since heating).

Data from Cibola are shown in Fig. 2A. There were two distinct groups of TL curves: the upper, unheated group

Table 2. Archeologic sources of the cherts studied.

Archeologic site	Number of cherts tested	Approximate age of site (years before the present)
Chiripa, Bolivia	4	2100 to 3100
Foss, Missouri	4	5000 to 7000
Cibola, New Mexico	7	700
Koster, Illinois	10	5000 to 8000

and the lower, heated group with natural doses of ~ 500 krad. With one exception, all the unheated samples are brown whereas the heated samples are red. One artifact was part red and part brown. Samples were cut from each section of this sample, and the TL results showed that the red section had been heated although the brown section had not. However, there was one red artifact which TL shows to have been unheated.

The results for the artifacts from Koster are shown in Fig. 2, B and C. Nine of the ten samples divided clearly into the two groups, heated or unheated (Fig. 2B). There is little correlation between the visual appearance and heat treatment of these cherts; some unheated cherts are very similar in appearance to control samples heated by the archeologists.

The tenth Koster artifact gave the exceptional curve a (Fig. 2C). It has a low equivalent dose at low glow curve temperatures but nearly saturation doses at high glow curve temperatures. Probably this artifact has been heated, but only to a relatively low temperature. Measurements were made on several samples of raw Burlington chert which were an-

nealed for 3 hours at various temperatures. These results are also plotted in Fig. 2C. The annealing drains the TL up to a glow curve temperature approximately 100°C greater than the annealing temperature. By comparison with these results, the temperature at which a partially heated artifact was annealed could be estimated. The chert giving curve a apparently had been heated to a temperature between 300° and 350°C in prehistoric times. The temperature can be determined only approximately as the amount of thermal drainage is also dependent on the annealing time.

To further test the validity of the TL measurement, we measured the radioactivity of the Koster and Cibola cherts and then calculated the expected equivalent doses of heated artifacts. The dose rates for Koster and Cibola are ~ 0.1 and ~ 0.3 rad per year, respectively, giving equivalent doses of ~ 500 to 800 krad and ~ 200 krad, respectively, in heated artifacts. These numbers agree to within a factor of 2 with the values for the heated artifacts in Fig. 2, A and B, and further support the validity of the TL test.

On the basis of the success of the TL technique in the controlled tests described above, it appears that TL provides a relatively simple and reliable test for determining if a chert artifact was heated to $\sim 250^\circ\text{C}$ or more in prehistoric times. However, with this TL test it is not possible to distinguish between a deliberate heat treatment by prehistoric people and an accidental heating, such as in a fire; samples for testing should be chosen by archeologists with this in

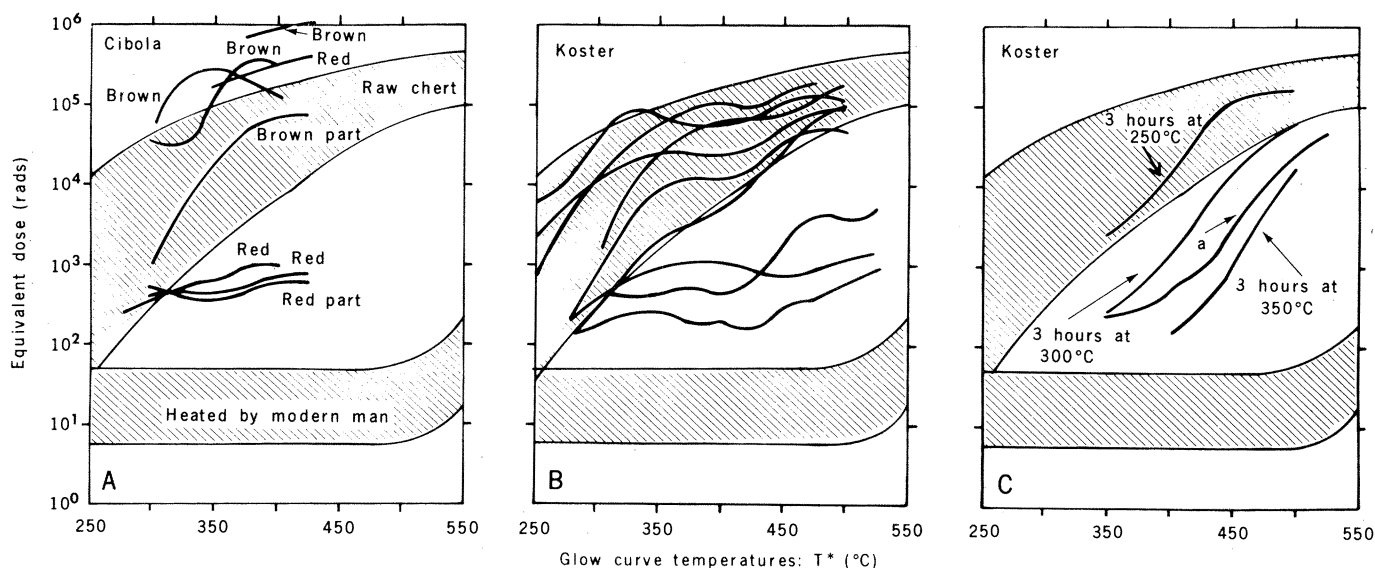


Fig. 2. Equivalent dose as a function of the glow curve temperature of (A) artifacts from Cibola, (B) artifacts from Koster, and (C) an exceptional Koster artifact. Curve a in (C) represents a partially heated artifact. For comparison, several raw chert samples were annealed in the laboratory at various temperatures. The results of annealing for 3 hours at 250° , 300° , and 350°C are shown.

mind. No control samples, such as raw source material, are required. The artifacts tested varied widely in color from white to dark red to brown, and the TL results demonstrate the difficulty of making judgments based on visual appearance alone, at least for cherts from these sites. In the case of the seven Cibola artifacts, judgment based on visual appearance proved to be correct on six samples but incorrect on one. The Koster cherts were carefully selected to contain what were thought to be, based on appearance, both heated and unheated artifacts. Although the TL test showed cherts of both types, the results did not correspond to the visual assessment.

The reliability of the TL determination of heat treatment is dependent on the age of the archeologic site; the older the site, the nearer the reaccumulated dose of a heated chert to the saturation dose of an unheated chert. For example, the difference between the heated and unheated cherts is less distinct for the Koster site (Fig. 2B, 5000 to 8000 years old) than for the Cibola site (Fig. 2A, ~ 700 years old). For sites \approx 10,000 years old, the archeologic dose is at least an order of magnitude less than the saturation dose and the detection of prehistoric heat treatment can be reliably based simply on a determination of the equivalent dose. For older sites, however, more detailed measurements must be made. The saturation level must be determined, the dose rate calculated from the radioactivity of the sample and its environ-

ment, and nonthermal ("anomalous") fading (I2) must be measured.

Although we have used this technique exclusively on chert, other lithic materials such as obsidian and fireplace rocks often give suitable TL and could be tested by the same technique.

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References and Notes

1. M. D. Mandeville, *Plains Anthropol.* **18**, 177 (1973).
2. D. E. Crabtree and B. R. Butler, *Tebuiwa* **7**, 1 (1964).
3. B. A. Purdy and H. K. Brooks, *Science* **173**, 322 (1971).
4. J. Weymouth and M. D. Mandeville, *Archaeometry* **17**, 61 (1975).
5. M. J. Aitken, *Physics and Archaeology* (Oxford Univ. Press, London, ed. 2, 1975), chap. 3.
6. H. Y. Göksu, J. H. Fremlin, H. T. Irwin, R. Fryxell, *Science* **183**, 651 (1974).
7. M. A. Seely, *J. Archeol. Sci.* **2**, 17 (1975).
8. R. D. Rowlett, M. D. Mandeville, E. J. Zeller, *Proc. Prehistoric Soc.* **40**, 37 (1974).
9. This relatively simple estimate of the natural dose ignores a number of factors important in accurate TL dating, such as supralinearity, sensitivity changes with heating, and anomalous fading. However, these effects should be less than a factor of 2 and thus not important in this application. We measured the anomalous fading of the cherts listed in Tables 1 and 2 and found less than 10 percent fading over 8 days.
10. H. P. Hoyt, Jr., J. L. Kardos, M. Miyajima, M. G. Seitz, S. S. Sun, R. M. Walker, M. C. Witteles, *Geochim. Cosmochim. Acta* **3** (Suppl. 1), 2269 (1970).
11. D. W. Zimmerman, *Archaeometry* **13**, 29 (1971).
12. A. G. Wintle, *Nature (London)* **245**, 143 (1973).
13. We thank D. Browman, R. Druhot, M. Fritz, S. Struever, P. J. Watson, and J. Weymouth for kindly supplying the cherts and several anonymous reviewers for helpful comments. This work was supported by NSF grants SOC 73-09029 and BNS 76-82645.

3 March 1977; revised 23 May 1977

Arthropod Invasion of Land During Late Silurian and Devonian Times

Abstract. *Fossil floras and faunas of a Lower Devonian black shale from Alken, Germany, include aquatic, amphibious, and terrestrial forms. The presence of these forms suggests conditions in favor of an invasion of the land. Various arthropod adaptations to terrestrial life are present, including the development of a preoral cavity.*

One of the most important events in the history of the earth was the invasion of the land by higher plants and animals. It probably took place some 400 million years ago at the transition from late Silurian to early Devonian time. Fossil terrestrial plants, mainly psilophytes, are well known from these times, but a more detailed knowledge of the morphology of the plant and animal groups, as well as their ecology, is known only in two cases.

The well-known Rhynie Chert in Scotland, now regarded as Lower Devonian (Emsian), has yielded exceptionally

well-preserved plants and arthropods belonging to the Crustacea, Arachnida, and Collembola, all very small and with appendages intact. Evidently the environment was some kind of a swamp.

The second occurrence, near the village of Alken in the Mosel Valley, Germany, does not present the same excellent preservation of the fossils, but the spectrum of forms is much wider. The fossil flora and fauna comprise marine, brackish, and freshwater forms as well as terrestrial ones. The aquatic fauna includes mostly arthropods, pelecypods, and ostracoderm fishes. The fossiliferous

black shale is of Lower Devonian (Lower Emsian) age and was probably deposited in a lagoon along the coast of a major island (1).

The more or less aquatic plants along the shores of the lagoon formed small "mangroves." This material and the wet plant debris on the beaches evidently constituted conditions very favorable for an invasion and gradual adaptation to a life on land.

A recently completed study of the arthropods (2) demonstrates that a mixture of forms, which belonged to different ecological niches, was present. Eurypterids with swimming legs, such as *Parahughmilleria* (Fig. 1), were probably also able to travel outside the lagoon. The lagoon was occasionally visited by huge and predaceous pterygotous eurypterids, known to be to 180 cm in length. Other eurypterids without swimming legs walked on the bottom and perhaps occasionally walked on land. This might have been possible primarily because the gills (Fig. 2a) were protected by the plate-shaped abdominal appendages and could be kept moist in a way similar to that used by the living horseshoe crab, *Limulus*, when it comes on land.

The gills of eurypterids have been studied in Silurian specimens from Estonia by Wills (3) and in specimens from the Lower Devonian of Scotland by Waterston (4). The respiratory surface of the elliptical gill areas is enlarged by close-set cones with a reticulate surface (Fig. 2a). The ribs surround small areas of very thin chitin through which the respiration probably took place. In several places invaginations between the cones lead into masses of spongy chitin possibly penetrated by five canals (Fig. 2, b through d). Wills assumed that the gas exchange from water to blood took place within the spongy masses. However, the invaginations seem to be too few and limited to be able to take care of the complete aquatic respiration; moreover, water, in contrast to air, has considerable difficulty passing in and out of long narrow passages. Several terrestrial isopods have pseudotrachea in addition to gills, and both kinds of respiratory organs may occur on abdominal legs in one and the same individual. On the basis of a comparison with the isopods, it seems natural to assume that at least some eurypterids might have had both gills and pseudotrachea, the latter represented by the invaginations and spongy masses. The Estonian species *Baltoeurypterus tetragonophthalmus* ("*Eurypterus fischeri*"), showing the structures mentioned, had swimming legs and was thus largely a swimming form, but a fossil eurypterid