The Alken fossils include a fossil scorpion, Waeringoscorpio hefteri, which possibly had gills (Fig. 3, a and b). In most Paleozoic scorpions the absence of internal book lungs is indicated by the lack of ventral respiratory openings or stigmata, a feature first appearing in the Carboniferous. The early scorpions are generally regarded as aquatic, breathing by book gills (?) concealed above the ventral plates as in Limulus.

The book gills are regarded as homologous with the book lungs in scorpions and many other arachnids. The embryology of spiders demonstrates the probable transition from gills to lungs, which is a major adaptation of the respiratory organs to terrestrial life.

In my opinion, the final and decisive adaptation to terrestrial life seems to have been the development of a preoral cavity from which fluid containing digestive enzymes could pass to the prey. This assumption is based on paleontological and embryological evidence.

The paleontological evidence for this assumption is demonstrated in scorpions. In Recent forms (Fig. 3c) a preoral cavity is formed by the basal portions of the anterior appendages and by the maxillary lobes of the coxae of the two first pairs of walking legs. As indicated in Fig. 4, the preoral cavity developed gradually from the Silurian to the Carboniferous, a time span which largely corresponds to the time of transition from water to land in these animals. A further phylogenetic support for this idea is derived from the fact that in Recent arachnids a preoral cavity is feebly developed in primitive forms (Palpigrada) and well developed in the advanced forms.

The embryological evidence for a secondary development of a preoral cavity is well demonstrated in the ontogeny of myriapods and insects, the arthropods best adapted to terrestrial and even aerial life. In the Chilopoda (Myriapoda) the primary mouth is situated behind a broad labrum. The anterior part of the sternal area and the limb buds on either side of it gradually sink into the head capsule, forming a preoral cavity with a secondary mouth in front. In Crustacea on the whole, a preoral cavity is lacking in aquatic forms but is indicated in terrestrial isopods by a strong development of the basal portions of head appendages.

The Alken fauna also contains a primitive myriapod, Eoarthropleura (Fig. 5), which might have had a ventral labrum instead of a secondary mouth opening in front of a preoral cavity. Eoarthropleura might have been only about 10 cm long but is evidently related to the Upper Carboniferous Arthropleura, a giant which apparently had a length of up to 180 cm (5). Strangely enough, terrestrial arachnids from Alken also seem to be more closely related to the Upper Carboniferous species 70 million years younger (6). The main arthropod adaptations to terrestrial life are suggested in Fig. 6. LEIF STØRMER

Institute of Geology, University of Oslo, Norway

References

- 1. G. Solle, Notizbl. Hess. Landesamten Boden-
- G. Sone, Noitzbi. Hess. Landesamten Boden-forsch. Wiesbaden 98, 50 (1970). L. Størmer, Senckenbergiana Lethaea 51, 335 (1970); ibid. 53, 1 (1972); ibid. 54, 119 (1973); ibid., p. 359; ibid. 57, 87 (1976). 2.
- ibid., p. 359; ibid. 57, 87 (1976).
 3. L. J. Wills, Ark. Zool. Kungl. Svensk. Vet.-Akad. 18, 93 (1965).
- Akad. 18, 95 (1965).
 4. C. D. Waterston, Fossils Strata 4, 241 (1975).
 5. W. D. J. Rolfe, Treatise on Invertebrate Palaeontology, Arthropoda 4 (Geological Society of America, Boulder, Colo., 1969), p. 607.
 6. L. Størmer, Science 164, 1276 (1969).

Temperature Exposure Measured by the Use of Thermoluminescence

Abstract. A method has been developed to measure temperature exposure based on the use of the termperature-dependent property of thermoluminescence fade. This property provides a relatively simple and inexpensive method for measuring both average and accumulated temperature exposure without the need for sophisticated temperature-recording equipment.

Thermoluminescence (TL) has been used as a research tool in numerous analytical problems ranging from mineral identification to archeological dating (1). Thermoluminescent radiation dosimetry (TLD) is one application that has been widely accepted with the result that techniques have been standardized and various TLD phosphors have been developed (2). We report here a new application of TL in which the temperaturesensitive properties of TLD phosphors irradiated prior to use are utilized to measure temperature exposure. This application provides a simple and inexpensive method for measuring both average and accumulated temperature exposure over extended time periods and at remote locations, as is often re-



Fig. 1. Isothermal fade response observed for CaSO₄:Mn tags at the five temperatures indicated. Data points are the mean TL signal for one tag and have a 3 percent standard error at the 95 percent confidence level. Lines were fitted to the data by a combined computer analysis that specified both a hyperbolic fade response and a fixed relationship between fade rate and temperature.

quired in environmental research such as oceanography and fisheries biology.

The relationship between TL and temperature exposure is apparent from a simplified explanation of TL theory. When TLD phosphors are exposed to ionizing radiation, electrons are dislodged and some become trapped within the crystalline lattice. The probability of an electron's escaping from a trap varies according to trap depth (energy gap), trap frequency factor, and temperature. Heating the material provides the energy to free these trapped electrons, which emit light (thermoluminescence) while returning to ground state. The TL signal is easily determined with a TL analyzer that rapidly heats the phosphor at a uniform rate and measures the light produced. Temperature exposures that occur prior to the TL measurement can prematurely empty the traps and cause a reduction or "fade" in the TL signal.

Fade is generally viewed as an undesirable interference for most TL applications; however, the temperature-dependent characteristics of fade are ideal for measuring temperature exposure. Since fade rate varies with temperature, the total accumulated fade (total signal loss over time) is a direct measure of the past accumulated temperature exposure. The basic procedures for using this relationship are as follows: (i) a TLD phosphor is irradiated to fill a large number of electron traps; (ii) the phosphor is then allowed to fade for a known time period at the thermal conditions of interest; (iii) upon retrieval, the remaining TL signal is measured; (iv) this value is compared with the value that would have been obtained initially after irradiation; and (v) the total signal loss is used in con-SCIENCE, VOL. 197

²⁵ March 1977

junction with predetermined fade rate constants to calculate the past temperature exposure.

To facilitate the use of TLD's for measuring temperature exposure, we have constructed "TLD temperature tags" that contain sufficient TLD materials to allow replicate determinations and thus minimize analytical error. These tags were developed for use in environmental research (3), but they can readily be applied to numerous other research and industrial activities. Tag construction and fade characteristics will vary, depending on the specific application and the TLD phosphor used; however, a description of the tags that we have used for monitoring average environmental water temperatures and the thermal exposure of free-swimming fish will illustrate the conceptual and procedural methods.

The TLD phosphor used for this application was CaSO₄:Mn incorporated in a Teflon matrix. We constructed the tags by sealing 12 CaSO₄:Mn TLD's (microrods containing 8 percent phosphor by weight) in a length of stainless steel tubing. This procedure produces a small, durable tag (30 mm long and 3 mm in outside diameter), at a cost of \$1.30 per tag. All tags were simultaneously exposed to 250 roentgens (60Co) at room temperature and subsequently stored at 26.4°C for 240 hours to empty undesirable shallow traps. The standard error of the mean TL value of any tag (12 individual readings) was 3 percent at the 95 percent confidence level.

We determined fade characteristics by monitoring the fade response at isothermal conditions (Fig. 1). For these tags, isothermal fade between 10° and 26°C approximated a hyperbolic function of the form

$$\frac{1}{S} = \frac{1}{S_0} + m_{\rm T}t$$

where S is the TL signal at time t, S_0 is the initial TL signal, and m_T is the rate of change in 1/S. In this case, the uniform slope m_T was used as the temperature-dependent fade rate and it increased rapidly with increasing temperature; m_T approximately doubled for every 5°C rise. Since fade was retarded at low temperature, the tags were kept at temperatures below -25° C to preserve the stored TL signal prior to their use and analysis.

The increased sensitivity of CaSO₄: Mn tags at higher temperatures is advantageous for certain applications, but it can cause the calculated average temperature to be greater than the true average temperature exposure. This problem arises when the tags are used in



Fig. 2. A hypothetical example illustrating a difference in accumulated fade due to a fluctuating (A) and a uniform (B) average temperature exposure of 15° C. The anticipated TL signals were calculated on the basis of previously determined fade rates for the temperatures indicated. The calculated average temperatures (based on the final TL signal) are given in parentheses.

varying temperature conditions, since exposure to a fluctuating temperature produces more fade than a uniform exposure to the same average temperature. The total difference depends on the temperature range and the apportionment of time between temperatures. Figure 2 illustrates the expected difference in accumulated fade due to exposure to a variable (point A) and a uniform (point B) temperature of 15°C. For this extreme case, where time is equally divided between two temperatures, the calculated average temperature is only 10 percent $(1.5^{\circ}C)$ greater than the true average temperature exposure. Closer agreement is expected for less extreme cases.

If more than one type of TLD phosphor is used in each tag, it would be possible to more accurately define average temperature in a manner similar to that of qualitatively measuring radiation by utilizing the different energy dependence of two phosphors (4). One can obtain a wide range of fade rates by selecting phosphors that are characterized by different trapping depths. It is also possible to adjust the fade properties of individual phosphors by using various preconditioning treatments (5). Ideally, fade rate should remain uniform at a given temperature and be matched to the timetemperature conditions of interest. Further details concerning the use of TLD's for measuring temperature exposure are reported elsewhere (6).

Since TLD phosphors (i) are easy to use, (ii) are small, (iii) are relatively inexpensive, and (iv) have a wide range of fade rates, they provide a valuable research tool that can be used to measure temperature exposures under a variety of conditions.

> G. Patrick Romberg W. Prepejchal S. A. Spigarelli

Ecological Sciences Section, Radiological and Environmental Research Division, Argonne National Laboratory, Argonne, Illinois 60439

References and Notes

- 1. F. Daniels, C. A. Boyd, D. F. Saunders, Science 117, 343 (1953).
- J. H. Schulman, in Luminescence Dosimetry, F. H. Attix, Ed. (publication US AEC CONF-650637, U.S. Atomic Energy Commission, Washington, D.C., 1967), pp. 3–33; J. R. Cameron, N. Suntharalingan, G. N. Kinney, Thermoluminescent Dosimetry (Univ. of Wisconsin Press, Madison, 1968).
- B. G. Oltman, K. F. Eckerman, G. P. Romberg, W. Prepejchal, U.S. Patent No. 3,869,918 (1975).
- 4. J. R. Cameron and G. N. Kenney, *Radiat. Res.* 19, 199 (1963).
- F. R. Moran, Ener. Res. Dev. Admin. Rep. COO-1105-243 (1976).
- G. G. P. Romberg and W. Prepejchal, in Proceedings of the Fifth International Conference on Luminescence Dosimetry (Universität Giessen, Giessen, Germany, in press)
- Giessen, Germany, in press).
 We thank B. G. Oltman and Dr. R. A. Schlenker for reviewing the manuscript. We thank Dr. P. R. Moran for helpful consultation and M. M. Thommes, Dr. T. L. Beitinger, and E. M. Yaguchi for technical assistance. Funded by the U.S. Energy Research and Development Administration.

20 May 1976; revised 17 January 1977

Hydroxyl Radical Reactivity with Diethylhydroxylamine

Abstract. Diethylhydroxylamine (DEHA) reacts with gas-phase hydroxyl radicals on every third collision, whereas the corresponding reaction in aqueous solution is considerably slower. The high gas-phase reactivity explains the predicted inhibitory effect of DEHA in atmospheric smog processes. Results from the studies in the aqueous phase are helpful in predicting the mechanism of the reaction of DEHA with hydroxyl radicals.

Maugh (1) recently discussed the proposal of Heicklen and his co-workers (2) to use N,N-diethylhydroxylamine (DEHA), $(C_2H_5)_2$ NOH, as a radical scavenger in urban airsheds to inhibit smog chemistry. In order to learn how and why DEHA might be effective, we have

initiated a study of the reactivity of DEHA with radical species known to be chemically significant in atmospheric pollution reactions.

The hydroxyl radical (OH) is now regarded as being very important in smog chemistry. Pitts and his co-workers (3)