Reports

Giant Radioactive Halos: Indicators of Unknown Radioactivity?

Abstract. *A new group of giant radioactive halos has been found with radii in excess of anything previously discovered. Since alternate explanations for these giant halos are inconclusive at present, the possibility is considered that they originate with unknown alpha radioactivity, either from isomers of known elements or from superheavy elements.*

A radioactive halo is generally defined as any type of discolored, radiation-damaged region within a mineral and usually results from either alpha or, more rarely, beta emission from a nearby radioactive inclusion containing either uranium or thorium. When the inclusions are very small (≈ 1 μm), the uranium and thorium daughter alpha emitters produce a series of discolored concentric spheres surrounding the inclusion, which in thin section appear microscopically as concentric rings whose radii correspond to the ranges of the respective alpha emitters (*1*). Although the radii of normal uranium and thorium halos vary from 12 to 42 μm in mica, possible evidence of unknown radioactivity exists in the scattered reports of unusual halos with anomalous ring radii (*2, 3*) varying from 5 to 10 μm in the dwarf halos to about 70 μm in the giant halos.

The very few previously reported occurrences of giant halos seem to have been largely ignored, perhaps because either definite information on the presence and size of the halo inclusion was absent (*3*) or because subsequent confirmation of the report was lacking. Hoppe (*4*), for example, was unable to confirm the existence of giant halos found by Wiman in certain Swedish granites, but this is not surprising in view of the large variability in the occurrence of particular halo types and the relatively small number of thin sections that Hoppe examined. Indeed, after a more extensive search in which I examined about 1000 thin sections from these granites, I find that giant halos in the 55-μm range do exist in the biotite along with ordinary uranium and thorium halos. These giant rings invariably occur only around very densely colored thorium halos, a result which implies a correlation of this ring with a high thorium content of the inclusion. Examination of the thorium decay scheme shows that the daughter alpha emitter, Po^{212}, emits a low-abundance (1 : 5500) alpha particle of slightly higher energy (10.55 Mev, compared to a normal 8.78 Mev), whose range may be correlated with the observed giant ring. Although there is some question whether the frequency of the low-abundance alpha particles in this energy range can produce a halo ring, I presently infer this association to be correct. The density of giant halos in these granites is quite low, however, and after a further search I have found a mica sample from Madagascar with uranium and thorium halos, in addition to an exceptionally fine collection of giant halos including all the sizes reported by Wiman as well as several much larger varieties of halos heretofore unreported.

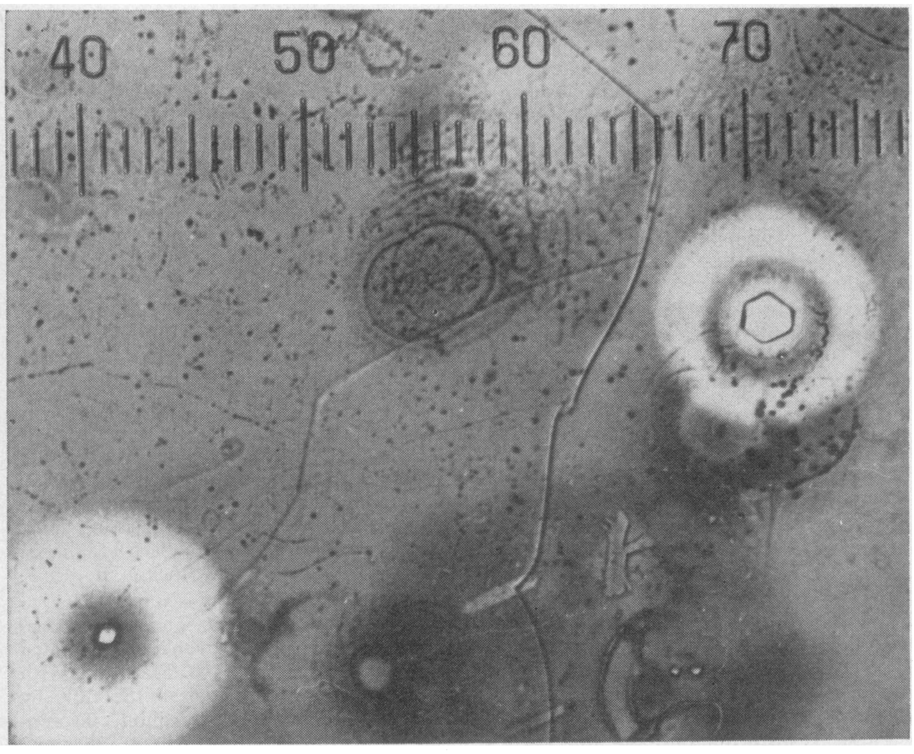


Fig 1. The halo on the right is a combination uranium and thorium halo, with the inner ring radius of 34 μm from the uranium daughter emitter Po^{214} ($E = 7.68$ Mev) and the outer ring radius of 40 μm from the thorium daughter emitter Po^{212} ($E = 8.78$ Mev). The halo on the left with a relatively small inclusion is a giant halo with about a 50-μm radius. One scale division = 10 μm.

The close proximity of occurrence of different halo types in the Madagascar mica provides an excellent range-energy relation which checks with coloration band widths produced experimentally in Van de Graaff helium ion irradiation of the mica matrix (*5*). Whereas the induced coloration bands are darker than the mica, the halos show reversal (bleaching) effects and are generally lighter than the surrounding matrix, except adjacent to the inclusion. Electron microprobe analyses indicate that the inclusions are monazites (*6*), and, since they are somewhat large (> 10 μm in diameter), they do not show ring structure as well as halos with point-like inclusions do. Also, the high radioactive content of some of the inclusions leads to an overexposed condition which tends to further obliterate inner ring structure.

The visual appearance of the giant halos (Figs. 1–3) is similar to that of the combination uranium-thorium halos, and the question arises whether long-range alpha particles have produced the giant halos. The affirmative answer to this question cannot be accepted without a critical examination of other modes of origin, since the magnitude of the giant halo radii involved implies the previous existence of naturally occurring alpha emitters with energies higher than any currently known.

Hence it is considered that the giant halos may have originated from:

1) *Variations in alpha particle range due to structural changes in mica.* Observations show that certain halo inclusions exhibit shapes or structural symmetry not exactly identical to the present outline of the inclusion in the mica matrix, and such deformations of the inclusion from radiation-damage effects might very well alter the structure of the matrix in the vicinity of the inclusion. However, there are numerous sites where uranium and thorium halos of normal size exist adjacent to and, in some cases, actually overlap giant halos (the inclusions of which show no evidence of any expansion or contraction). At least in these cases it would appear that the giant halos do not arise from normal-range alpha particles, which passed through a region of lower mica "density."

2) *Diffusion of a pigmenting agent from the inclusion into the matrix.* Although it is possible that some pigmenting substance may have been present, electron microprobe traverses across the region of the halo revealed no variations in elemental abundances of the matrix. Furthermore, in annealing experiments that were carried out at 450°C for 24 hours the yellowish tint of the halos either remained the same or in some cases became opaque; that is, there was no fading or otherwise any difference between the reaction of the uranium and thorium halos and that of the giant halos. In essence, if a purely chemical diffusion mechanism is operable, it is producing a type of coloration that is thus far indistinguishable from that initiated by radiation-damage effects. [Small crystalline structures (Liesegang patterns) often occur in mica, but these are easily distinguished from radioactive halos.]

3) *Diffusion of radioactivity from the inclusion to the matrix.* Electron microprobe analyses showed that uranium and thorium were confined to the inclusion; techniques by which fission tracks were induced indicated only a background uranium concentration surrounding the inclusion, and autoradiographic experiments with Kodak NTA emulsion showed alpha radioactivity restricted to the site of the inclusion. If diffusion of radioactivity has occurred, it is below the detection limit of these three methods.

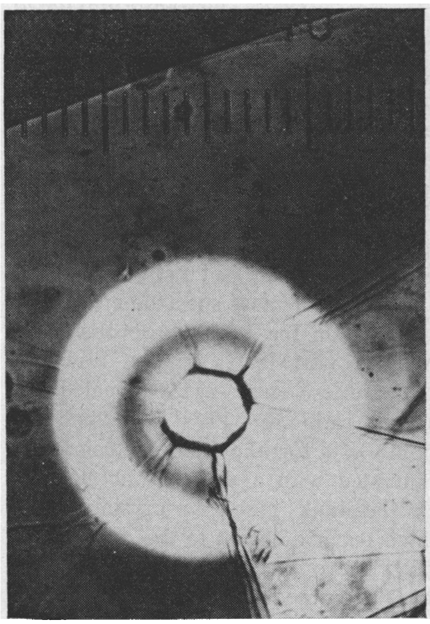
Table 1. Frequency of halo sizes of radii 32 to 110 μm.

Group	Interval of halo radius (μm)	Maximum energy of alpha particles (Mev)	Total No. of halos
I	32–35	7.68	22
II	37–43	8.78	274
III	45–48	≈ 9.5	28
IV	50–58	≈ 10.6	130
V	60–67	≈ 11.7	69
VI	70–75	≈ 12.3	58
VII	80–85	≈ 13.2	30
VIII	90–95	≈ 14.1	10
IX	100–110	≈ 15.1	5

4) *Channeling.* Even though different optical properties in the region parallel to the cleavage plane make it difficult to observe a transverse halo section in any mica, the giant halos do exhibit a three-dimensional structure typical of radioactive halos when successive mica layers are cleaved. The idea that channeling of normal-range alpha particles parallel to the cleavage plane would be instrumental in the formation of giant halo rings is certainly correct in principle. Whether the relatively small number of alpha particles emitted along any given cleavage plane is sufficient to produce coloration is not clear. Furthermore, if channeling were the explanation, a series of successive outer bands corresponding to a given multiple of the ranges of the uranium or thorium daughter alpha emitters, or both, might be expected in a given giant halo. This situation is not observed.

5) *Beta radiation instead of alpha emission.* Laemmlein (7) found beta halos of rather diffuse boundaries with radii up to several thousand micrometers surrounding thorium-containing monazite inclusions in quartz. The fact that many of the perimeters of these giant halos in this mica are well-defined does not favor the association of these halos (Figs. 1–3) with the beta halos; neither do the radii correspond. In addition, Laemmlein noted a correlation between the radius of the beta halo and the volume of the halo inclusion (that is, the thorium content). This is understandable, since energetic beta rays producing coloration at maximum range would emanate throughout the volume of the inclusion. In contrast, no such effect is observed in this mica. Giant halos and uranium and thorium halos occur around relatively small inclusions as well as around larger ones.

6) *Long-range alpha particles from spontaneous fission.* Long-range alpha particles with a broad energy spectrum accompany normal spontaneous fission events from U^{238} in an abundance of about 1:400. Neither of these factors is favorable for the production of relatively sharp boundaries such as are seen in certain giant halos. Upon etching several giant halos with hydrofluoric acid to reveal fission tracks, I have found that fission tracks emanate from the inclusions of some, but not all, giant halos. The tracks emanating from some of the inclusions may be attributed to



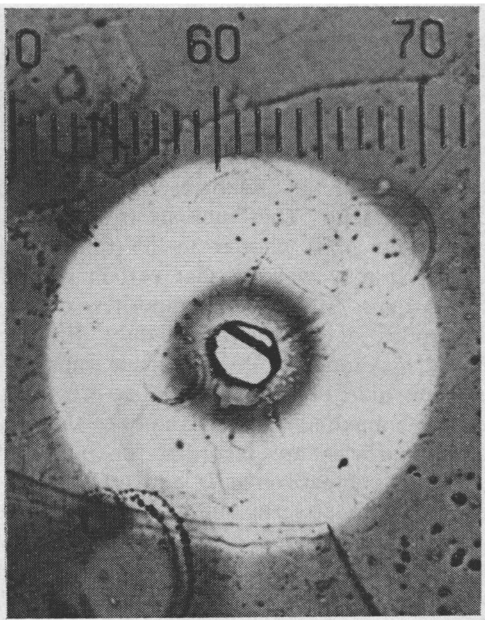


Fig. 2 (left). A giant halo approximately 57 μm in radius, presumably due to the long-range alpha particles from Po^{212} ($E = 10.55$ Mev). One scale division $= 10$ μm. Fig. 3 (right). A giant halo approximately 84 μm in radius, whose origin is unknown. If the halo is due to long-range alpha particles, the energy would be about 13.1 Mev. One scale division $= 10$ μm.

the uranium content of the halo inclusions. The lack of fission tracks in other inclusions implies that at least in these cases long-range alpha particles from spontaneous fission are not instrumental in producing the giant halos.

7) *Alpha particles or protons from* (n,α) *or* (α,p) *reactions.* Mica sandwiches containing halo inclusions were irradiated with a total flux of 5×10^{18} neutron/cm^2. No induced coloration was noted in the mica section adjacent to the inclusion after irradiation. Since this integrated flux is several orders of magnitude higher than would be expected in naturally occurring inclusions, it appears that (n,α) reactions have not produced the giant halos. Calculations show that (α,p) reactions are also insufficient to produce coloration (see *8*).

From the preceding comments it would appear that, although some of the above explanations cannot be definitely excluded, neither can any be presently confirmed as a factor responsible for the origin of the giant halos. Therefore, a few remarks may be made concerning the distribution of halos in this mica and the possibility that the giant halos may have originated with long-range alpha activity either from isomers of known elements or from superheavy elements.

The radii of several hundred halos that were measured with a precision of about ± 1.5 μm are given in Table 1. Greater accuracy was possible but seemed unnecessary, since for halos with large inclusions the actual radius of the halo as measured from the inclusion edge to the halo perimeter will vary up to around 5 to 6 μm with the variation dependent upon the stage of halo development (*9*). Other uncertainties in the radii measurements arise if the inclusion is inclined with respect to the cleavage plane. The intervals of halo radii were thus chosen to be rather broad; it may well be that certain of the groups listed are composites of subgroups of halos with slightly different maximum radii, but further subdivision did not seem justified at present. The maximum energy values of the alpha particles are recorded for purposes of comparison *only* and are *not* meant to necessarily imply that the respective halo groups originated with alpha particles of that energy. There were a few halos which did not fall into any of the above categories, but the number of this type was only a small percentage of the total (2 percent). Halos in groups I and II are the normal uranium and thorium halos, whose maximum radii may be identified with the respective daughter alpha emitters Po^{214} ($E = 7.68$ Mev) and Po^{212} ($E = 8.78$ Mev) of these decay series. Halos in group IV may be associated with the low-abundance, long-range alpha particles from Po^{212} ($E = 10.55$ Mev) in the Th^{232} decay series.

An attempt to relate other groups of long-range alpha emitters of polonium isotopes in the uranium and thorium decay chain with the giant halo radii is more difficult. For example, the 9.5-Mev group of Po^{212}, which conceivably could produce a 48-μm halo, occurs in an abundance of only about 1:30,000; the 9-Mev group (1:45,000) of Po^{214} could produce a 45-μm halo; and there exist still other groups with energies up to 10.5 Mev, but these occur in an abundance of only about $1:10^6$. If it is considered that these alpha particles were emitted in the same abundance as is presently observed, only the halos in group IV may reasonably be attributed to known low-abundance alpha particles of higher energy. G. N. Flerov has suggested that Po^{212m}, an isomer of polonium with a half-life of 47 seconds and an alpha-particle energy of 11.7 Mev, not known to occur naturally, may have been responsible for the halo group in the 62- to 67-μm range, since the energy correlates with the prescribed range (*10*). This identification, if correct, would, first, constitute another example of a rather peculiar phenomenon, namely, the occurrence of halos originating with polonium isotopes apparently unrelated to uranium and thorium daughter products (*11*), and, second, raise the interesting possibility that the other giant halo groups may be associated with unknown isomers emitting high-energy alpha particles in the 10- to 15-Mev range. Kohman has suggested that such alpha emitters, if they exist, may be shape isomers (*12*) of known nuclides.

Very recent mass spectrometric studies in which the Ion Microprobe Mass Analyzer (IMMA) (Applied Research Laboratories) was used revealed an isotope ratio for Pb^{207} to Pb^{206} of about 0.16 for the halo inclusions as contrasted with a value of about 0.35 for the bulk monazite crystals (*13*), which occur adjacent to the mica (both values were uncorrected for common Pb). If subsequent work shows that this difference cannot be attributed to common Pb, this result might suggest that a closer examination be made of possible high-energy isomers, namely, an isomer in a chain decaying to Pb.

The possibility that the giant halos originate with a postulated superheavy element (*14*) in the region from atomic numbers 110 to 114 seems remote, since these elements (i) would not be expected to occur in monazites and (ii) would be expected to exhibit spontaneous fission activity either directly or indirectly (that is, to decay by way of alpha emission to the known spontaneous fission region below atomic number $Z = 105$) (*15*). As noted earlier, some giant halo inclusions do not exhibit background fission tracks. However, of special interest in this context are very recent theoretical calculations by Bassichis and Kerman (*16*), which indicate an island of superheavy element stability at somewhat higher Z (around 120). If such an element exists, it might be expected to occur in a pegmatitic mica.

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References and Notes

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5. I thank Dr. F. Young, Physics Department, University of Maryland, for cooperation in the Van de Graaff experiments.
6. L. Kobren, Goddard Space Flight Center, National Aeronautics and Space Administration, and C. Feldman, Oak Ridge National Laboratory, performed the electron microprobe analyses, which revealed typical monazite constituents.
7. G. G. Laemmlein, *Nature* **155**, 724 (1945).
8. I assumed that (i) (α,p) reactions in the inclusion occur mainly with phosphorus and have a cross section of 0.1 barn, (ii) weight fractions are 0.25, 0.05, and 0.2 for Th, U, and P, respectively, and (iii) 0.2 is the fraction of U atoms decayed (≈ 0.1 from fission-track analysis on the mica); calculations then show that the integrated proton flux from (α,p) reactions in an inclusion 25 μm in diameter is at least a factor of 10^2 below that required to produce threshold coloration ($\approx 10^{13}$ alpha particles per square centimeter from Van de Graaff irradiation) in a giant halo of radius 75 μm. Similar considerations hold for (α,p) reactions on nuclides of low Z in the surrounding matrix.
9. Variations of 1 μm in halo radii are noted even with point-like inclusions, possibly resulting from maximum ionization (coloration) occurring at slightly less than end-point range. In larger inclusions the halo radius appears to increase several micrometers as the halo develops because a greater fraction of the alpha particles are being emitted within the outermost micrometer of the inclusion. Nonuniform halo boundaries also occur and may result from a nonuniform distribution of radioactivity in the inclusion or may be related to one of the unusual development modes previously described herein.
10. I thank G. N. Flerov, Director, Laboratory for Nuclear Reactions, Joint Institute for

Nuclear Research, Dubna, U.S.S.R., for this suggestion.
11. R. V. Gentry, *Science* **160**, 1228 (1968).
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13. This work was performed by C. Andersen, Applied Research Laboratories, Goleta, Calif. In the IMMA a finely focused (10 μm) O_2^- ion beam is used to sputter material directly into a mass spectrometer.
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15. Mass spectrometric analyses of mica containing the giant halo inclusions, performed at Ledoux & Company, Teaneck, N.J., and the GCA Corporation, Bedford, Mass., indicated that, if superheavy elements are present, their abundance must be less than 200 parts per million. The IMMA analysis of the monazite inclusions revealed the presence of what are almost certainly molecular ions with a mass of 303 (possibly CaThP), 310 (possibly La_2O_2) and somewhat higher values.
16. W. H. Bassichis and A. K. Kerman, *Phys. Rev.*, in press.
17. I thank P. Ramdohr, University of Heidelberg, and H. de la Roche, National Center for Scientific Research, Nancy, France, for the Madagascar mica samples; I thank J. Boyle, Oak Ridge National Laboratory, for valuable assistance with the experiments. The research performed at Oak Ridge National Laboratory was sponsored by the U.S. Atomic Energy Commission under contract with Union Carbide Corporation. Part of the research was performed at Columbia Union College, Takoma Park, Maryland, from which I am presently on leave of absence.

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Monosodium Glutamate: Lack of Effects on Brain and Reproductive Function in Rats

Abstract. *Monosodium glutamate was injected subcutaneously in infant rats of both sexes. The lateral preoptic and arcuate nuclei and median eminence were examined by light and electron microscopy for possible monosodium glutamate effects. As adults, treated animals showed no adverse monosodium glutamate effects on the reproductive system and neural morphology.*

Olney (*1*) reported that a single subcutaneous injection of monosodium glutamate (MSG) in newborn mice (2 to 9 days of age) induced acute neuronal necrosis in several brain areas including the preoptic and arcuate nuclei and median eminence region. Newborn mice which received a series of injections of MSG, when adult showed a syndrome which suggested a complicated neuroendocrine disturbance. These mice exhibited skeletal stunting, marked obesity, and female sterility. Acute brain damage was also found in a newborn rhesus monkey after a single administration of MSG (*2*). This report is concerned with both the acute and long-range effects of MSG upon the development of the brain and reproductive function of male and female rats after a single injection of MSG into newborn animals. The rat was chosen for these studies because Olney (*1*) and Olney and Sharpe (*2*) stated that acute hypothalamic lesions also are produced in rats, and also because injections of hormones (*3*) and drugs (*4*) at a "critical stage" of neonatal development can drastically effect hypothalamic control mechanisms as related to reproduction and sexual behavior.

Male and female rats of the Wistar strain (Simonson Laboratories, California), 3 to 4 days old, received a single subcutaneous injection of MSG. The MSG (Mann Research Laboratories) was injected subcutaneously in a volume of 0.1 ml; the total dose was 4 mg per gram of body weight. This concentration is approximately four times greater than the minimum dosage which produced hypothalamic lesions in newborn mice (*1*) and a newborn monkey (*2*). Control rats were injected with an equal volume of saline. In experiments concerned with the acute effects of MSG on the brain, the animals were killed 3 hours posttreatment. To determine long-range effects of MSG, uniform litters (eight pups per mother) were kept in an environmentally controlled room until weaned. The experiment was terminated at 68 days posttreatment for males and 88 days posttreatment for females. Relative reproductive organ weights were recorded (organ weight per unit body weight). The anesthetized rats were perfused intracardially with normal saline followed by either 10 percent buffered formalin or Flickinger's fixative (*p*H 7.2). Brains perfused with formalin were paraffin embedded, sectioned, and stained with cresyl violet for light microscopy. Brains perfused with Flickinger's modification of Karnovsky's fixative (*5*) were sectioned transversely, and the preoptic nucleus, median eminence, and arcuate nucleus were dissected out. The tissue was washed in 0.2*M* cacodylate buffer (*p*H 7.2), postfixed in 1 percent cacodylate-buffered osmium tetroxide for 1 hour, dehydrated in graded alcohols, and embedded in Epon 812. Silver and gray sections were stained with uranyl acetate followed by lead citrate and examined in a Hitachi HS-7S electron microscope.

Light microscopic examination of the

Table 1. Effect of monosodium glutamate (MSG) injection in infant rats on adult reproductive organ weights. Organ values are the mean ratios of organ weight (in milligrams or grams, as noted below) to body weight (in hundreds of grams), plus or minus the standard error of the mean.

	Females			Males			
Group	No. of rats	Ovaries (mg/100 g)	Uterus (mg/100 g)	No. of rats	Testes (g/100 g)	Seminal vesicles (mg/100 g)	Prostate (mg/100 g)
Control	7	34.7 ± 0.9	162.9 ± 9.4	8	1.02 ± .03	244.9 ± 17.0	120.7 ± 9.9
MSG-treated	8	29.3 ± 1.4*	179.8 ± 10.8	6	1.06 ± .06	287.0 ± 16.8	118.3 ± 3.5

* The difference from the value for the control group is significant at the $P < .01$ level.

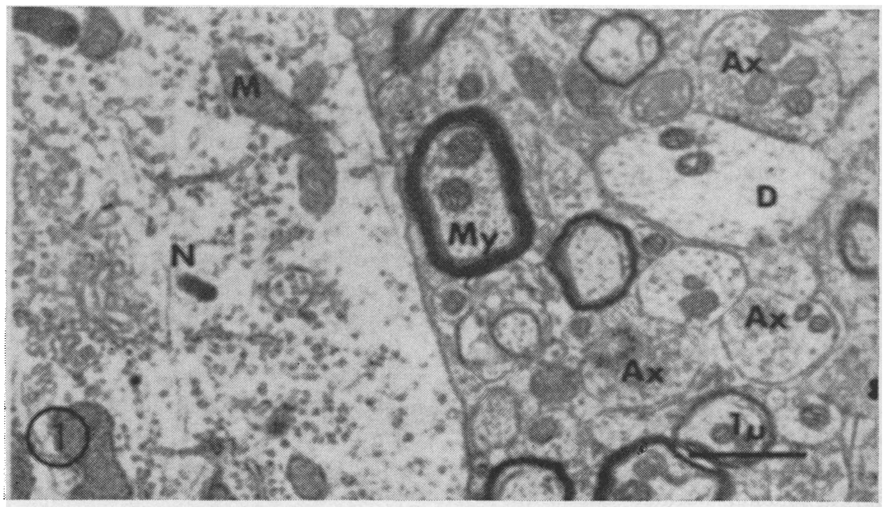

Fig. 1. Neuron (*N*) and neuropil of the lateral preoptic nucleus in an MSG-treated adult rat. *Ax*, axon terminal; *D*, dendrite; *M*, mitochondrion; *My*, myelinated axon (× 16,300).