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LODESTONE -- NATURE'S OWN PERMANENT MAGNET

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GODDARD SPACE FLIGHT CENTER -



LODESTONE - NATURE'S OWN PERMANENT MAGNET

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ABSTRACT

The early history of geomagnetism - The history of the lodestone, are equivalent statements and though the phenomonology of the lodestone has been known worldwide for several thousand years, there has never been a definition of the lodestone or any explanation why certain iron ores behave as permanent magnets. Presented are magnetic hysteresis and microstructural details which explain why the class of magnetic iron ores defined here as proto-lodestones, can behave as permanent magnets, i.e. lodestones. Certain of these proto-lodestones which are not permanent magnets can be made into permanent magnets by charging in a field greater than 1000 oersted. This fact, other experimental observations, and the vague field evidence coming from antiquity and the middleages, which seems to indicate that lodestones are found as localized patches within massive ore bodies, suggests that lightning might be responsible for the charging of lodestones. The large remanent magnetization, high values of coercive force, and good time stability for the remanent magnetization are all

characteristics of proto-lodestone iron ores which behave magnetically as fine scale (< 10 μ m) intergrowthswhen subjected to magnetic hysteresis analysis. The magnetic results are easily understood by analysis of the complex protolodestone microstructural patterns observable at the micrometer scale and less. The iron ore is magnetically hardened by various natural processes giving rise to proto-lodestone iron ores. The protolodestone ores are then charged by lightning.

INTRODUCTION AND BACKGROUND

'I extremely praise, admire and envy this author for that a conceit so stupendous should come into his mind, touching a thing handled by infinite sublime wits and hit upon by none of them'. This comment on William Gilbert's¹ classic study of nature's only permanent magnet the lodestone - was by Galileo.² Appreciation of Gilbert's classic work extends to present time.³ Since 1600 when Gilbert published the results of his careful studies, there has never been a real definition of the lodestone or any explanation for its permanent magnetic properties. What makes one iron ore a lodestone while another not? How does the lodestone become charged as any magnet must and why does the lodestone hold this charge?

About 1200 AD the Chinese actually tested the strength of lodestones by using weighed Aron objects and preceded Gilbert in observing many of the characteristics of the lodestone. The important Chinese contributions have been placed in proper perspective by Joseph Needham.⁴

He offered valid corrections to the writings of those chauvinistic Western authors without sinological competence who caused the history of magnetism to become quite confused. Western authors have generally misinterpreted, misread, or ignored the Chinese literature to which was difficult to gain access. One notable example can be found in the historical sketch in the treatise on Geomagnetism by Chapman and Bartels⁵ who obtained much of their source and reference from the works of Mitchell⁶. It should be noted that the western authors erred only when attempting to evaluate the historical and comparative context of the Chinese contributions. Many historical studies, giving proper credit to those who preceeded Gilbert in the West such as Peter Peregrinus, Agricola, etc., have been published.^{7,6} The lodestone has been associated with medicinal, mystical, nautical, and scientific activities for several thousand years and for about 600 years prior to 1800 was of considerable economic value. The invention of the electromagnet⁸ placed the lodestone in a position of a curiosity unexplained.

The lodestone up to present has been referred to as magnetite, magnetic magnetite etc.⁹ Such reference was probably valid, since magnetite has been used so loosely in reference to natural magnetic iron ores. However, universal acceptance of this definition, probably as much as anything else, prevented any elucidation of lodestone properties. In fact it would have been difficult for anyone to evaluate

the lodestone before about 1950 when fine particle magnetic theory was being developed.¹⁰ Ore microscopy, usually at low magnification, shed no light on the lodestone because it did not address the problem. Ramdohr's¹¹ only mineralogical comment was simply a reference to all lodestone appearing oxidized (maghematization) a conclusion reached by Mason¹² as well. One way in which matural remanent magnetization may be intensified is by partial oxidation, i.e. the production of maghemite $\langle Fe_2O_3 \rangle$. Maghemite by definition is the defect spinel after magnetite (Fe₃O₄) wherein the iron is in the Fe³⁺ state. The probable role of maghemite in the magnetization of lodestones was first considered by Nagata,¹³ and is important.

Iron oxide powder magnets were produced by Gowin Knight in 1799, and though the similarities, not recognized at the time, between these magnets and the lodestones were never explained we will show that they are basically similar. The Bureau of Mines in 1941 published Bulletin 425,¹⁴ an extensive work dealing with the magnetic separation of iron ores. The Bureau of Mines researchers studied the particle size effects on coercivity, remanence etc. According to Davis,"... the inadequacy of magnetic remanence as a criterion for lodestone is disclosed by the fact that some magnetites which have no appreciable attraction for ferromagnetic substances such as soft iron may be converted into lodestones with strong magnetic attraction and with a remanence as high as natural lodestone by the action of a moderately strong

unidirectional magnetic field." This is the only prior mention of the connection between implied intrinsic properties and charging by a strong field. Clearly the last significant study of the lodestone was by Gilbert in 1600 and in modern times the work described in the U.S. Bureau of Mines Bulletin 425 and in the text <u>Rock Magnetism</u> by Nagata, constitute our prior quantitative knowledge of the lodestone.

All natural materials possess remanent magnetism whether they be terrestrial soils and rocks, lunar samples, or meteorites. It is the intensity of the remanent magnetism, the large coercive force, and the stability of the remanent magnetism which serves to distinguish the lodestone from other natural materials, and in particular the iron ores which do not possess permanent magnet properties. It is convenient at this point to introduce the proto-lodestone definition in view of the fact that there are iron ores which can be made into permanent magnets by charging in a strong unidirectional field. The proto-lodestone, then, is any iron ore which has high coercivity and saturation magnetic remanence, and good magnetic stability, and which can be made into a permanent magnet by charging with an electromagnet.

The existence in nature of uncharged proto-lodestone material, the large values of the ratio, natural remanence (NRM) to saturation remanence (SIRM) for the lodestone and the various references to lodestone finds in the literature of antiquity and the middle ages ¹⁵ indicating that they are found as localized patches in otherwise massive ore bodies would seem to suggest lightning as a possible source mechanism in the

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charging of lodestones. Aside from the definition and explanation of the lodestone, this study provides a description of the magnetic properties of the class of materials - magnetic iron ores-since answering the question - What is a lodestone? requires an unders anding of these iron ores.

Further implications are provided for archaelogical research. Since we provide a definition of proto-lodestone and a probable method of charging, the existence of magnetic science in an early civilization should at least require the existence of proto-lodestone iron ore proximate to the living site or juxtaposed along trade routes. Testing the "magnetic quality" of an iron ore artifact with a hand held magnet is not sufficient, as a distinction cannot be made between magnetite and proto-lodestone material. However, after testing with a magnet, the proto-lodestone material may possess strong attractive properties of its own. Without the lodestone it would be unlikely that an early civilization would discover the basics of magnetic attraction. It is also important to distinguish which civilization made and used 'steal' from those which did not, with or without access to the lodestone. The Chinese for example as early as 1000 AD used thermoremance as did Gilbert to magnetize iron needles. Since the polarity of axial thermoremanence might be experienced by anyone hot-working or casting an iron alloy into rods, swords, etc., it would be possible to develop a compass without the presence of lodestone. Magnetite, lodestone, and other iron ores possessing magnetic moments could be fashioned into magnetic

pointers, but only lodestone is capable of charging pointers by touch. Gilbert also experimentally observed the phenomena of magnetic viscosity, the acquisition of opposite polarities, and the phenomena associated with what is now called the Curie Point. In addition he understood how to magnetically harden steels.

In this paper we present for the first time a clear elucidation of the magnetic hysteresis and microstructural detail which explains the nature of proto-lodestone ores which are capable of being permanent magnets.

THE LODESTONE

The most obvious way to distinguish a lodestone is of course to directly test its permanent magnet properties by using it to pick up paper clips etc. One could become more quantitative by using a technique the Chinese used in 1000 AD, i.e. weighing bits of iron and measuring the pick up distance etc. Using finely powdered Fe_3O_4 , the lodestone magnetic field patterns can be discerned and the position of the poles identified (Figure 1). Magnetostatic effects, the influence of discontinuities, such as cracks, sharp edges, and inclusions, and the sample geometry, are all clearly visualized using this simple 'powder pattern' technique. The original field patterns associated with lodestone samples (outlined at extreme left in Figure 2) M8 and M4, can be modified by application of a 5000 oersted field in the horizontal plane (bottom to top in photo) or vertical (out of the picture). These patterns cannot be observed for similar sized samples of Fe_3O_4

(magnetite). With an appropriate lodestone sample, after noting the position of the poles, o.e can cut pointers as shown in Figure 3. The step by step separation of the rods cut from the original piece is presented to demonstrate the polarity memory as well as magnetostatic effects, noted as the separation distance increases. It should be mentioned here that Peter Peregrinus (1296) and William Gilbert (1600) "machined" spheres of lodestone with north and south poles which they had identified previously.

DEFINITION OF THE PROTO LODESTONE

Before a magnetic iron ore becomes a lodestone by virtue of any charging mechanism, it must have, as we will demonstrate, microstructural characteristics developed as a consequence of exsolution, oxidation induced phase separation, and maghematization which provides for high magnetic coercivity, high saturation remanent magnetization and good time stability. These magnetic characteristics are embodied in the definition of the proto-lodestone as the proto-lodestone is any iron ore possessing the requisite microstructural related magnetic hysteresis characteristics to qualify as a permanent magnet.

Magnetic hysteresis loops for about 30 massive magnetic iron ores, hematitic ores, taconite, single crystals of magnetite and other iron ores were measured on a PAR vibrating sample magnetometer in fields up to 12000 oersted. Samples studied came from widespread geographic locations. The hysteresis loop for a specimen from USNM 99484, a

strong lodestone, is illustrated in Figure 4A. The remanent coercive force, H_R is defined in Figure 4B. All the magnetic parameters discussed in this paper are defined in this figure: H_c - coercive force, H_R remanent coercive force, I_S - saturation magnetization, I_{SR} - saturation remanent magnetization, R_I - ratio I_{SR}/I_S , R_H - ratio H_R/H_C . Table 1 summarizes magnetic parameters for several specimens from 99484 to demonstrate the variation to be found within a strong lodestone mass.

	Spec.	I _{SR} (emu/gm)	R _I	H _C (Oe.)	H _R (Oe.)	R _H
99484	017	16.97	0.24	225	465	2.07
	063	13.01	0.21	262	610	2.33
	062	12,09	0.19	295	750	2.54
	071	16.25	0.25	284	600	2.1

TA	BLE	1

The iron ores which qualify as proto-lodestones:

-/

- have saturation magnetization values \leq 80 emu/gm but \geq 20 emu/gm
- have saturation remanent magnetization values > 5 emu/gm
- have coercive force values ≥ 100 oe up to 350 oe
- have $R_{\rm H}$ values between 2.0 and 2.5 though some magnetically anisotropic lodestones have values as high as 3.5
- have R_{I} values > 0.1, most with values > 0.14 and the c_{L} ingest > 0.20
- the NRM/I_{SR} values are quite large, many exceeding 0.5

Those ores which do not qualify:

- have I values > 80 emu/gm
- have R_H values > 4.0
- have R_T values ≈ 0.05 to ~ 0.01
- have NRM/I_{SR} values ≤ 0.1 to 0.01.

It is useful at this point to consider Fe_3O_4 (magnetite), before explaining the reasons for the proto-lodestone properties. A single crystal of Fe_3O_4 from Algiers (several mm on edge), and compacted one micrometer Fe_3O_4 powder (MO7029 - Pfizer Co.) are contrasted in Table 2.

TABLE 2

Sample	H _c (oe)	RI	
Fe ₃ 0 ₄ - crystal	1.8	< 0.01	
Fe ₃ 0 ₄ - 1µm powder	2 5	0.40	

Note that the critical factor is the effective particle size; in effect the 1 μ m powder can be compacted, then exposed to an unidirectional field and a permanent magnet results. Specimens of lodestones 99484, M24, and proto-lodestone samples Ulmer and M13 were ground to fine particle sizes (down to < 37 μ m) with essentially no change in magnetic hysteresis properties; Fe₃O₄ ground to the same size fractions exhibited continually increasing coercivity etc. <u>Even though a piece of</u> <u>iron ore which behaves as a lodestone may weigh several kilograms it</u> exhibits intrinsic fine intergrowth properties.

The saturation magnetization of all proto-lodestone material was \leq 80 emu/gm, compared to ~ 96 emu/gm for Fe₃O₄. This suggests that substitution of ions such as Ti, Mg, or Al, "or example, or oxidation, or both to be responsible for this observation. Optical studies, at the limits of our optical resolution (~ 1200 X), of polished and HC1 etched mounts using oil immersion and the Nomarski interference contrast technique reveals varied and complex microstructural patterns due to numerous causes such as is shown in Figure 5,6 and 7. Many of the lodestones, contain significant Ti, such as ULMER (\sim 12%) and M24 (~ 8%) (Figure 8). Proto lodestone M13 is an intergrowth of Ti rich $(A_0 = 8.45\text{\AA})$ and Ti poor $(A_0 = 8.40\text{\AA})$ spinel phases. The role of oxidation is much more difficult to evaluate, as oxidation is not necessary to produce a lodestone, but some of the weaker proto lodestones appear to be thus classified because of oxidation and 99484 the strongest lodestone studied contains no titanium but contains two discrete phases identified by x-ray diffraction($A_0 = 8.396$ and $A_0 = 8.376$ A). The phase with $A_0 = 8.376$ A is identified as maghemite. We then attempted to ascertain if there are differences between oxidized Fe304 and oxidized proto-lodestone material in a comparative optical study of natural samples. In Figure 9A,B and C the (111) (0001) Fe_{20_3} decomposition pattern in various Fe_{30_4} samples Fe₂0₄∥ is well defined while the oxidation pattern for lodestones with the primary phase separation is diffuse as shown in Figure 9D,E, and F. These patterns appear to be universal.

The magnetic hysteresis data and the optical miscroscopy survey of the proto-lodestone iron ores and other magnetic iron ores are summarized in Figures 10, 11 and 12. Proto-lodestone iron ores exhibit characteristics of small multidomain to interacting monodomain intergrowths, while other magnetic_iron_ores_exhibit characteristics of coarse multidomain material.¹⁶ The large H_c values, coupled with $\rm R_{H}$ values between 2.0 and 2.5 (see Figure 12), the large $\rm I_{SR}$ value (Figure 11) and the large R_T values (Figure 10), suggest that the proto lodestone iron ores have an ultrafine microstructure, which is supported by the accompanying micrographs. The finer the scale of the phase volumes the stronger the lodestone in terms of its saturation remanent magnetization and coercivity. Samples such as ULMER and M13 which are proto-lodestones as defined were not permanent magnets. If thermoremanence were responsible for charging the lodestone these samples should possess permanent magnet properties. Application of a 4000 field produced strong magnets. This experiment argues for a charging mechanism where strong transient fields are available. The lightning discharge is the only natural source of intense magnetic fields. From the literature of antiquity and the middle ages it would appear that lodestones were found as isolated patches within ore bodies. All of this evidence is circumstantial at present, and in the absence of any definitive data associated with the collection of lodestone, it is difficult to argue from field evidence. Field tests should provide very useful information about

the role of lightning charging, and should be definitive, since the effects of lightning would be local in nature, and irregular in extent and path.

Presently, the role of oxidation <u>per se</u> in lodestone magnetization is incompletely understood, and remains one of the experimental barriers to a total understanding of the lodestone. We do know that oxidation decomposition in magnetite proceeds according to the classical $(0001)_{Fe_2O_3} \parallel (111)_{Fe_3O_4}$ synchoshear mechanism¹⁷ in relatively uniterrupted fashion producing the recognizable pattern (Figure 9A,B, C). In lodestones with a primary evolution pattern the fine scale of primary precipitation prevents this magnetite pattern from developing, thus plates and needles of Fe₂O₃ do not form.

Other important aspects of the proto-lodestone ores involve the role of tectonic stresses, the development of cataclastic texture and the possible development of anisotropic precipitation and oxidation patterns. These results are peripheral to this presentation but important to a total understanding of the lodestone and will be presented at a later time.

ARCHAELOGICAL IMPLICATIONS

Having explained the characteristics of proto lodestone iron ores, and realizing that hematitic, lateritic, taconitic and magnetite ores are not proto-lodestone ores and cannot be made into permanent magnets, i.e. lodestones, except possibly under very special circumstances,

some archaeological implications are evident. Implied here is the recognition that a civilization which worked with steel might have come to appreciate the magnetization phenomenon called thermoremanence, with or without access to or prior knowledge of the lodestone.

As early as, and possibly before, 1000 AD^{4,6,7} it was understood that thermoremanence or lodestone touching would magnetize iron needles for use as pointers in the geomagnetic field. The possible use of bars or such similar objects, made of iron ores which are not proto lodestone ores, for pointers, has been discussed by Carlson.¹⁸ Object M160 - an Olmec Artifact - which he describes has a magnetic remanent vector, but is probably not a proto-lodestone though there may be lodestone objects in the Olmec artifact record. If the object was used as a geomagnetic pointer this is significant. Did the Olmec's know of the lodestone? Was M160 lodestone charged? Since iron was apparently not known to the Olmecs there was no way that they could have discovered magnetic polarity etc. in thermoremanent magnetization of elongate iron objects. It would be hard to argue in favor of the Olmec civilizations familiarity with the magnetic arts if no connection can be made with their knowledge of lodestone.

The definition of proto-lodestone material presented in this paper also points to the possible fallacious use of a hand magnet in rating iron ores as magnetically strong etc. and attempting to

discriminate between lodestones and other iron ores. All materials with characteristics summarized in Figures 10, 11 and 12 would be strongly attracted to a hand held magnet whether they be proto-lodestones or not.

DISCUSSION

Magnetic iron ores can be classified as proto-lodestone based on their magnetic hysteresis properties. There are two categories of proto-lodestone ores, those which have been charged by some magnetization mechanism - the permanent magnet lodestone - and those which have magnetic hysteresis properties similar to the lodestone, but which are not permanent magnets. This latter category can be made into magnets by subjecting them to an unidirectional magnetic field of \sim 1000 cersted or greater. Iron ores which do not fit the protolodestone class cannot be made into a magnet, this includes magnetite. The magnetic hysteresis analyses prove that the proto-lodestone ores have been magnetically hardened, and exhibit 'fine intergrowth' magnetic characteristics. The proto-lodestone are analogous to precipitation alloy magnets such as Alnico whereby hardness is achieved by decreasing the magnetically effective phase volumes and adding shape anisotropy with magnetic interactions figuring to be significant. Microscopic analyses using oil immersion at up to 1200x magnification with the Nomarski interference contrast technique provides graphic evidence for the micrometer and submicrometer scale of the microstructure

responsible for the 'fine intergrowth' magnetic characteristics. Oxidation plays a minor to significant role in hardening the protolodestone. Iron ores rich in Titanium or other elements are hardened by primary exsolution which responds to oxidation, by preservation of the relict primary precipitation patterns, the oxidation proceeds in an optically diffuse manner. In magnetites oxidation precipitation proceeds via the classic (111) $\text{Fe}_30_4 \parallel (0001) \text{Fe}_20_3$. Two categories of lodestones are identified, those which derive their properties via primary exsolution and another which is due to complex maghematization and oxidation induced phase separation. Once the magnetic iron ore is magnetically hardened - the proto lodestone can be made into a lodestone by a lightning strike. The full details of the microstructural phase relations, the significance of oxidation, and definitive experimental verification of the lodestone charging mechanism - including actual lightning strikes will be published later. Each lodestone or proto-lodestone is somewhat distinct from another, but the magnetic hardening is common to all.

CONCLUSIONS

We have presented the first explanation for the magnetic properties of lodestones and the microstructural elements responsible for the magnetic hardening of those irons ores which can become nature's only permanent magnets. Mother Nature has in essence done exactly what a magnet scientist or technologist might, i.e. magnetically hardening the material-then charging it. In fact there are some interesting

parallels with such precipitation hardened commercial magnets such as Alnico, etc. Mechanical and thermochemical alterations either synchronous with formation of the iron ore body, or possibly at some later time produced the complex microstructural patterns responsible for magnetically hardening the iron ore. On cooling through respective Curie points all natural iron oxides acquire thermoremanence which when measured in the laboratory is called natural remanent magnetization (NRM). When a sample is saturated in a strong magnetic field it will acquire saturation isothermal remanence (SIRM). The ratio NRM/SIRM gives some indication as to whether thermoremanence or some other mechanism is responsible for charging a lodestone. The ratios for members of the solid solution series Fe_30_4 - $\text{Fe}_2\text{Ti}0_4$ are ≤ 0.01 whether oxidized or The ratios for lodestones evaluated are > 0.5. Maghematization not. can result in intense magnetization under certain circumstances but there is no definitive information available to discern whether such a large ratio can be due to such oxidation. However if a transient strong field > 1000 versted is applied, this will satisfy the NRM/SIRM results for the lodestones. The only natural mechanism for producing such strong fields is a lightning discharge. This can be effective in making permanet magnet lodestones from previously magnetically hardened iron ores. Laboratory and natural lightning experiments and field evidence should suffice to confirm the lightning charging mechanism for the lodestones.

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Figure Captions

- Figure 1 Magnetic field pattern 9 centimeters above the surface of USNM 99484 (the scale bar is 9 cm)
- Figure 2 Magnetic field patterns at the polished surface of specimens M8 and M4, - the original pattern is indicated to the right of the specimen outline. Patterns after applying a 5000 oersted field in the horizontal plane, and verticel are indicated.
- Figure 3 Magnetic field patterns at the surface of a slab-from sample USNM <u>B8294</u>. (A) original piece (B) Sliced sample (4 pieces) pieces in intimate contact, (C) sliced sample pieces separated (D) sliced sample pieces further separated. X and X' mark the top and bottom of the sample.
- Figure 4 Magnetic hysteresis loops for sample USNM 99484

A - complete loop with parameters defined

B - Definition of the remanent coercive force (H_R)

- Figure 5 Optical micrographs for samples M24 and USNM 108591 -Samples etched with HC1.
- Figure 6 Optical micrographs for sample USNM 99484. Sample etched with HC1.
- Figure 7 Optical micrograph for sample USNM 76464 Sample etched with HCl.

- Figure 8 EDAX chemical spectra (Fe and Ti) for two proto-lodestones <u>M24</u> and <u>Ulmer</u> and corresponding probed regions
- Figure 9 Oxidation patterns (no maghemite) in magnetites (A,B,C) and in proto lodestone (D,E,F)-needle like structure is Fe₂O₃ in A,B, and C. Diffuse whitish areas are oxidized areas in D,E, and F.
- Figure 10 Plot of R_I (ratio of saturation remanence I_{SR} to saturation magnetization (I_S) vs saturation remanence (I_{SR}) for magnetic iron ores. Proto lodestones are indicated by filled circles.
- Figure 11 Plot of saturation remanence I_{SR} vs coeraive force H_C for magnetic iron ores. Proto-lodestones are indicated by filled circles.
- Figure 12 Plot of coercive force H_C vs remanent coercive force H_R for magnetic iron ores. Proto lodestones are indicated by filled circles.

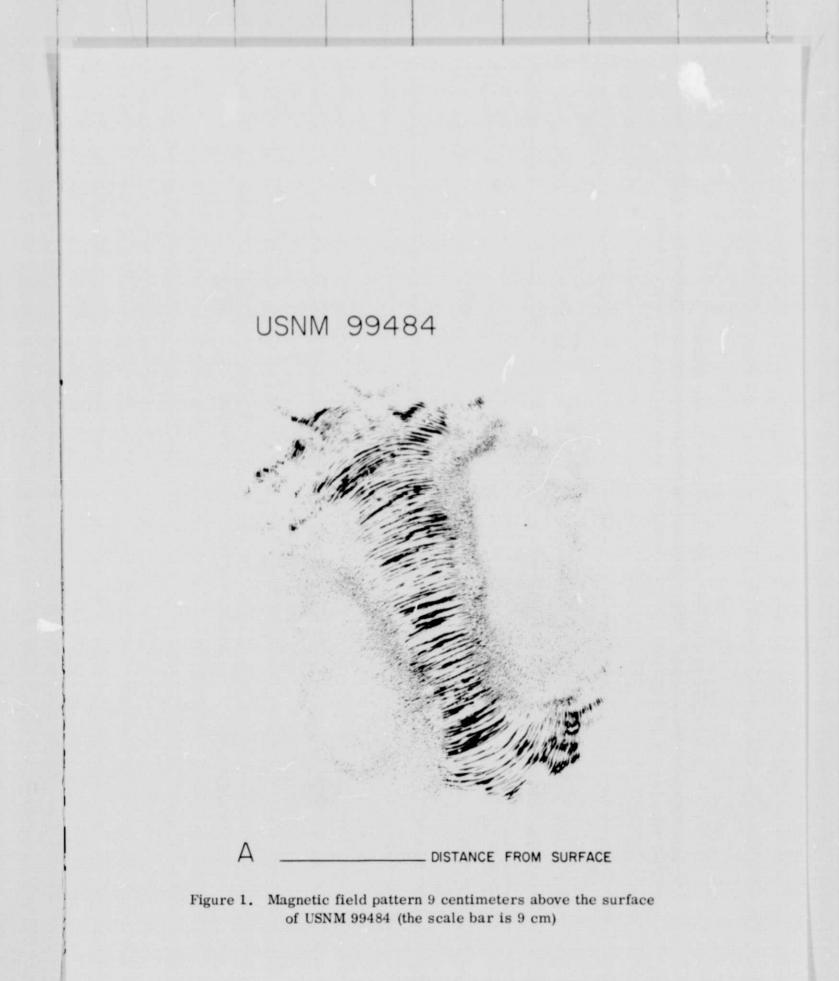
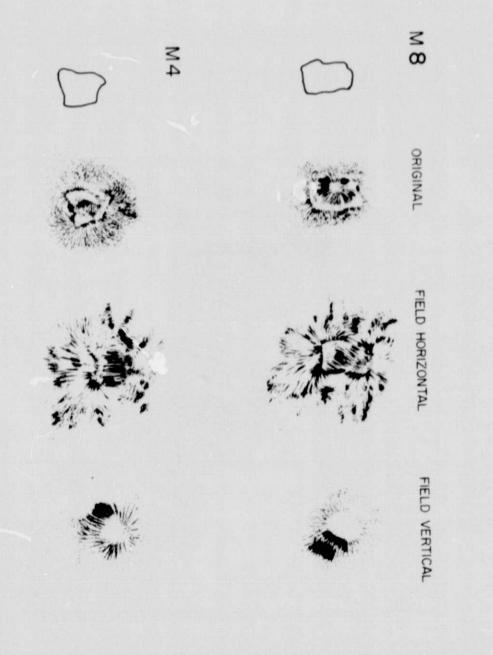
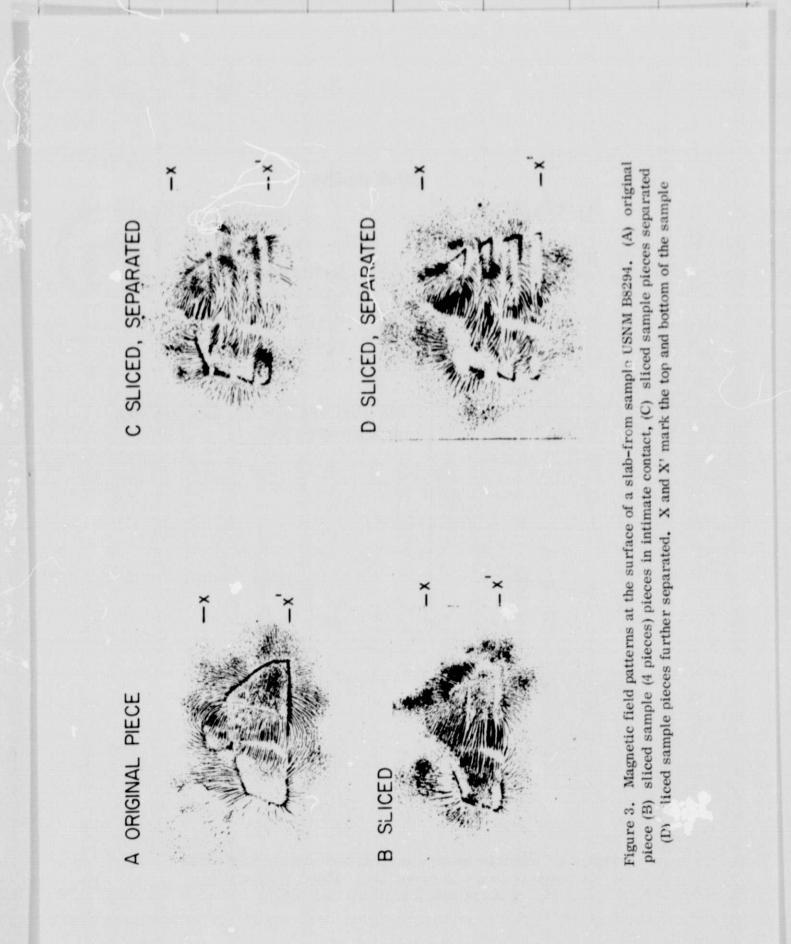
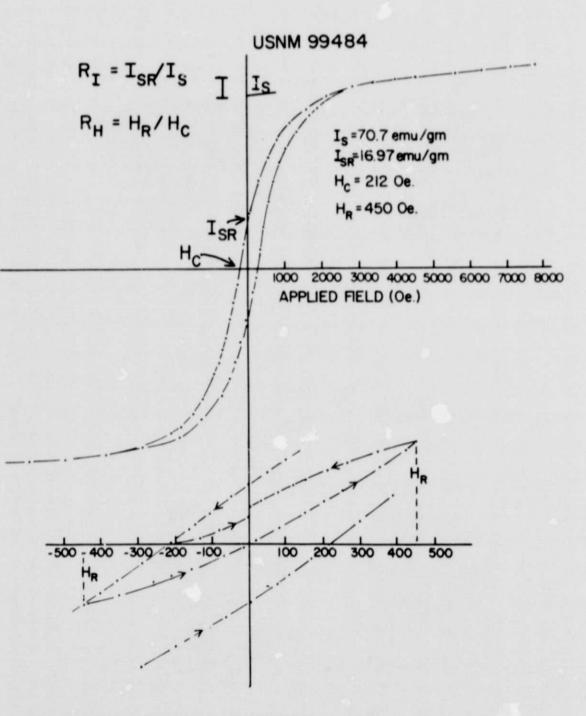
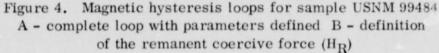


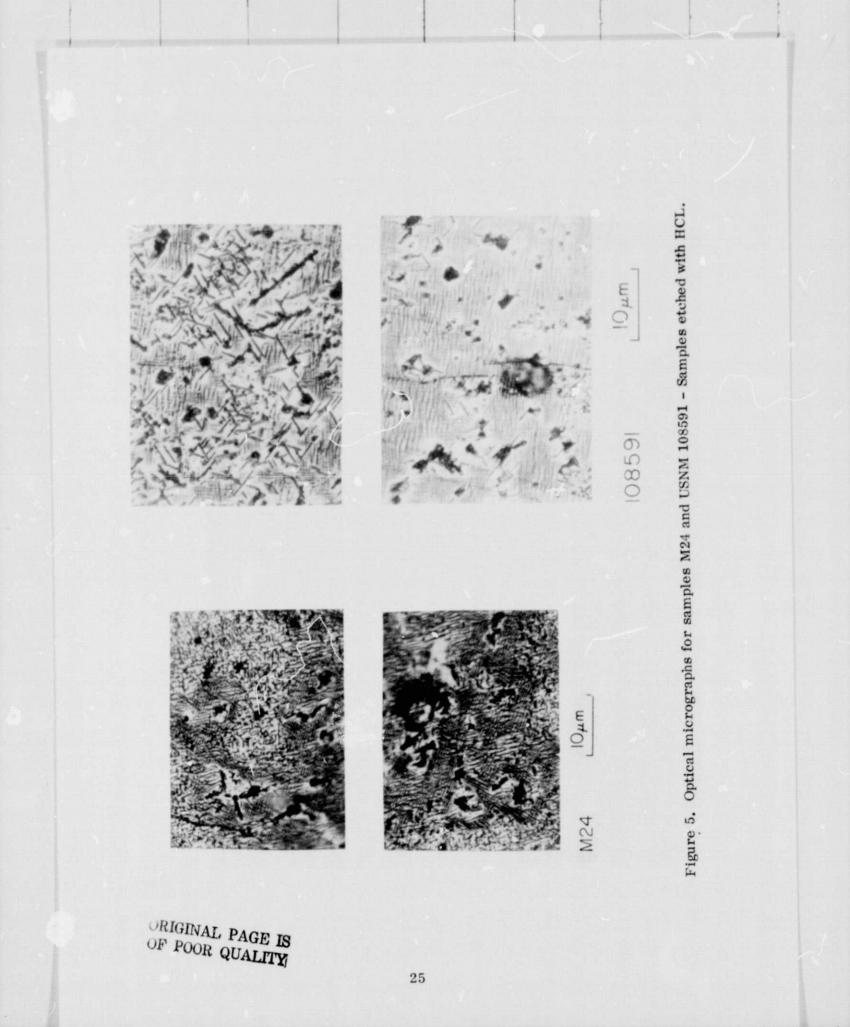
Figure 2. Magnetic field patterns at the polished surface of specimens M8 and M4, - the original pattern is indicated to the right of the specimen outline. Patterns after applying a 5000 oersted field in the horizontal plane, and vertical are indicated







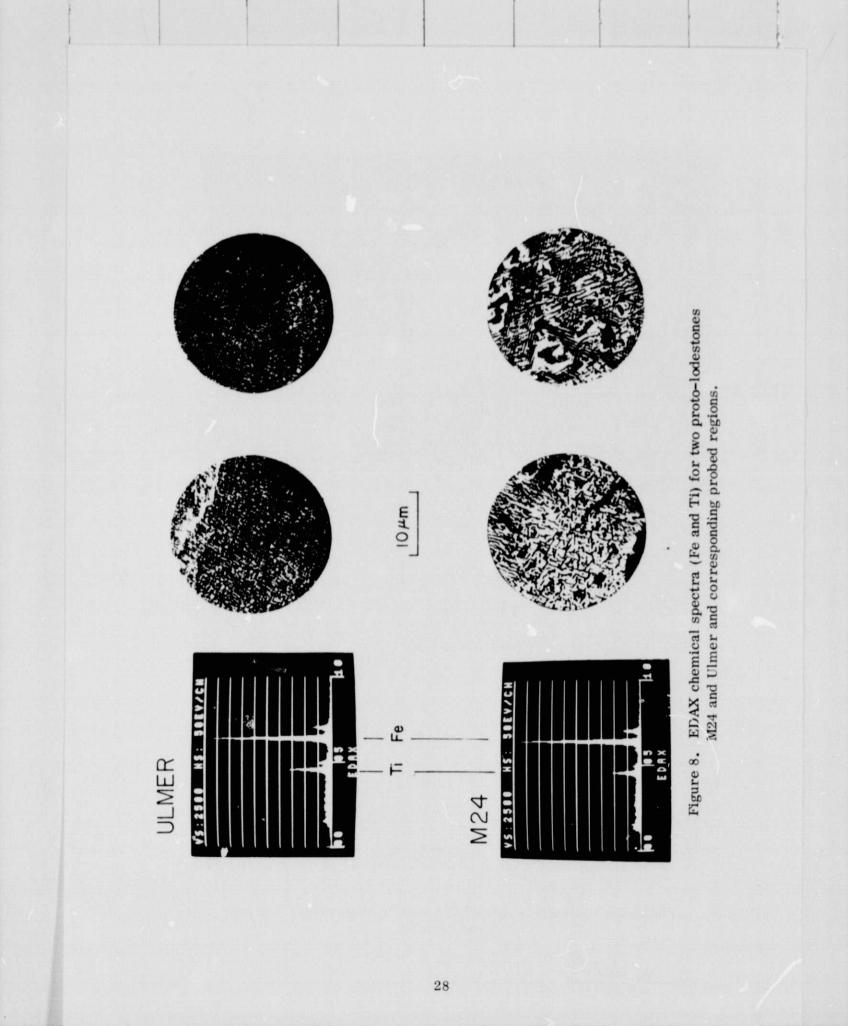


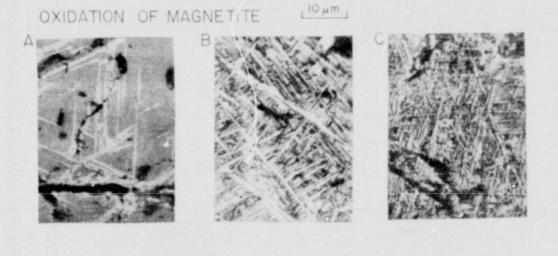






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OXIDATION OF PRIMARY PRECIPITATION MICROSTRUCTURE

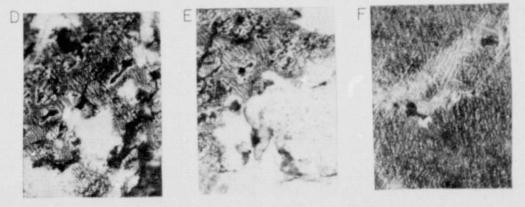
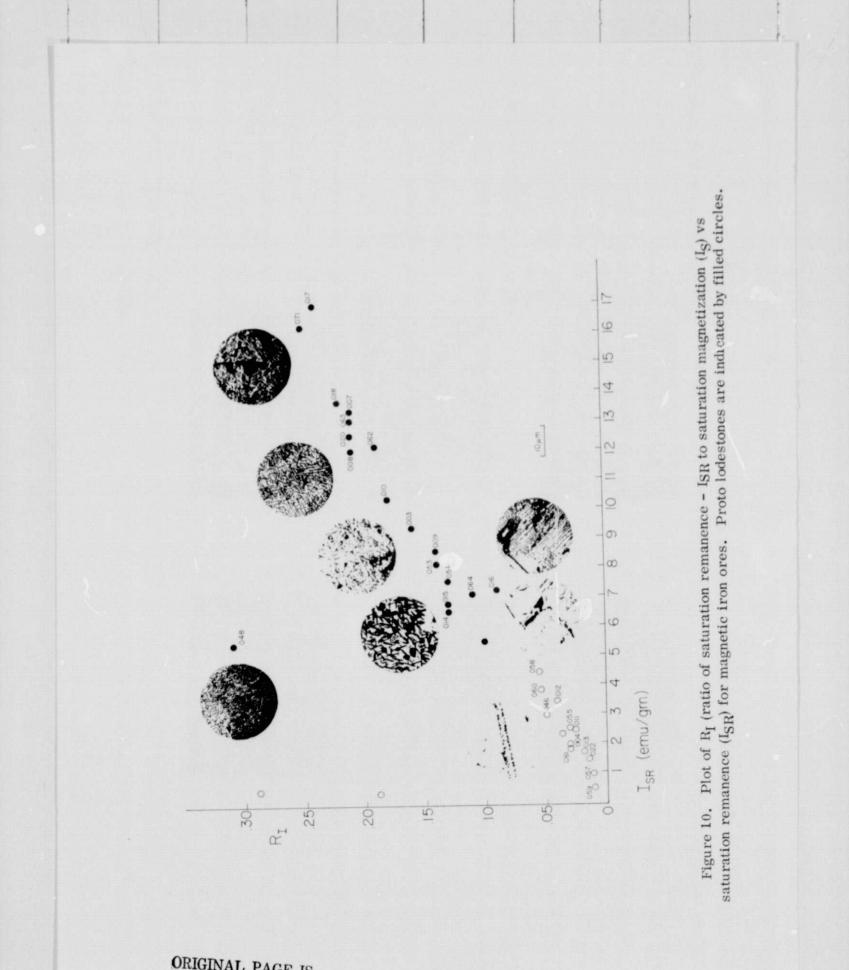


Figure 9. Oxidation patterns (no maghemite) in magnetites (A, B, C) and in proto lodestone (d, E, F) - needle like structure is Fe₂O₃ in A, B and C. Diffuse whitish areas are oxidized ar as in D, E and F.



ORIGINAL PAGE IS OF POOR QUALITY

