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Magnetic Characteristics of Sedimentary Rocks from the Tsengwen-chi and Erhjen-chi Sections in Southwestern Taiwan

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ABSTRACT

Four basic magnetic parameters (κ, SIRM, Hcr, MDF_{SIRM}) and two thermal magnetic analyses were investigated in this study to characterize the magnetic properties of magnetite-, pyrrhotite- and greigite-bearing rocks of the Tsengwen-chi and Erhjen-chi sections. These rocks have very similar behaviors in the acquisition of isothermal remanent magnetization (IRM) in spite of bearing with different magnetic minerals. Because of the difference in magnetic mineral concentration and grain size, greigite-, pyrrhotite-, and magnetite-bearing rocks have relatively high, intermediate and low parameter values (κ, SIRM, Hcr, MDF_{SIRM}), respectively. Thermomagnetic analysis in a nitrogen atmosphere has further revealed that each magnetic mineral has its own diagnostic magnetization curves and that greigite could have transformed to pyrrhotite and magnetite, pyrrhotite to magnetite, and magnetite to hematite. Bulk magnetic susceptibility has also shown remarkable drop in greigite-bearing rocks below 320°C, while there are minor and almost non-effective susceptibility changes in pyrrhotite- and magnetite-bearing rocks below 400°C, respectively.

1. INTRODUCTION

Magnetic minerals in sedimentary rocks or sediments generally make up less than 0.1 percent of the volume and constitute only a minor portion of them. However, these minerals have the capability of carrying fossil magnetism and can provide invaluable information about magnetostratigraphy, tectonic reconstruction and geomagnetic field behavior. In addition, the species of magnetic mineral, their mode of concentration and distribution of grain size also can provide clues to the early history of host deposits, including sediment sources, diagenetic processes and sedimentary environments. In the past, by using synthetic or natural material,

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many workers studied rock magnetic properties to characterize their relations with magnetic mineral composition, grain size and concentration (Parry, 1965; Day *et al.*, 1977; Cisowski, 1980; Dankers, 1981; King *et al.*, 1982; Maher, 1988). Some useful parameters have been applied to study natural diverse environments (Banerjee *et al.*, 1981; Özdemir and Banerjee, 1982; Thompson and Oldfield, 1986; Lund *et al.*, 1992).

In a recent publication (Horng *et al.*, 1992), it was pointed out that three major magnetic minerals, detrital magnetite (Fe_3O_4), authigenic greigite (Fe_3S_4) and pyrrhotite ($FeS_{1.1}$), are commonly present in the two Plio-Pleistocene marine-deposited sections (Tsengwen-chi and Erhjen-chi) in southwestern Taiwan (Figure 1). It was also shown that the occurrence of these magnetic minerals is intimately associated with lithology. Greigites, having relatively high concentration, are always prevalent in fine-grained mudstones. Magnetites, showing very poor concentration, are often present in sandy formations. And pyrrhotites, being in moderate concentration, are usually existent in the muddy siltstones.

All of these three magnetic minerals are ferrimagnetic. Magnetite is one of the most widespread iron oxides in the Earth's crust and its magnetic properties have been understood over several decades of studies (Parry, 1965; Stacey and Banerjee, 1974; Banerjee and Moskowitz, 1985). Among iron sulfides, non-magnetic pyrite is thermodynamically stable relative to pyrrhotite and greigite (Berner, 1971). Therefore, pyrite is so common and easily found in reducing sediments that greigite and pyrrhotite seem to have been neglected before. It was not until 1964 that the first natural greigite was discovered in a lacustrine sequence (Skinner *et al.*, 1964). In this sequence, greigite is restricted to clay and fine silt layers and occurs as tiny grains evenly dispersed throughout the host clay. Later on, greigite was reported not only in lacustrine but also in marsh and marine deposits (Dell, 1972; Demitrack, 1985; Snowball and Thompson, 1988; Horng *et al.*, 1989; Tric *et al.*, 1991). By scanning transmission electron microscopy, Mann *et al.* (1990) and Farina *et al.* (1990) further indicated that ultrafine particles of greigite and possible pyrrhotite can be present in multicellular magnetotactic bacteria, showing a possible biogenic source of these iron sulfides. But so far, studies on the magnetic properties of greigite and pyrrhotite are still few (Clark, 1984; Snowball and Thompson, 1990; Hilton, 1990; Tric *et al.*, 1991).

Some basic rock magnetic parameters are very useful, although not straightforward, in reflecting magnetic mineral concentration and grain size. Initial volume magnetic susceptibility (κ) and saturation isothermal remanent magnetization (SIRM) are commonly used as magnetic mineral concentration indicators (Thompson and Oldfield, 1986), while remanent coercivity force (Hcr) and median destructive field of SIRM (MDF_{SIRM}) are mainly correlated with the grain size (Parry, 1965; Dankers,1981; Banerjee and Moskowitz, 1985). Thus, it is believed that we can use these four parameters to characterize and differentiate the two section rocks which have quite different magnetic mineral composition and grain size. Besides, in the heating process, chemical reactions due to instability of magnetic and ferrous minerals would cause changes in bulk magnetic properties. In this study we will also present evidence of these changes by using thermomagnetic analysis and thermal magnetic susceptibility method.

2. LABORATORY WORK

A total of 28 cored specimens (1 inch diameter, 2.2 cm in length) from the two sections, with known magnetic carriers (Horng, *et al.*, 1992), were used for a series of nondestructive magnetic investigations. First, natural remanent magnetization (NRM) and κ were measured with a Digico spinner magnetometer and a Molspin minisep instrument, respectively. Then, each specimen was put in a pair of electromagnetic poles. Direct magnetic fields were

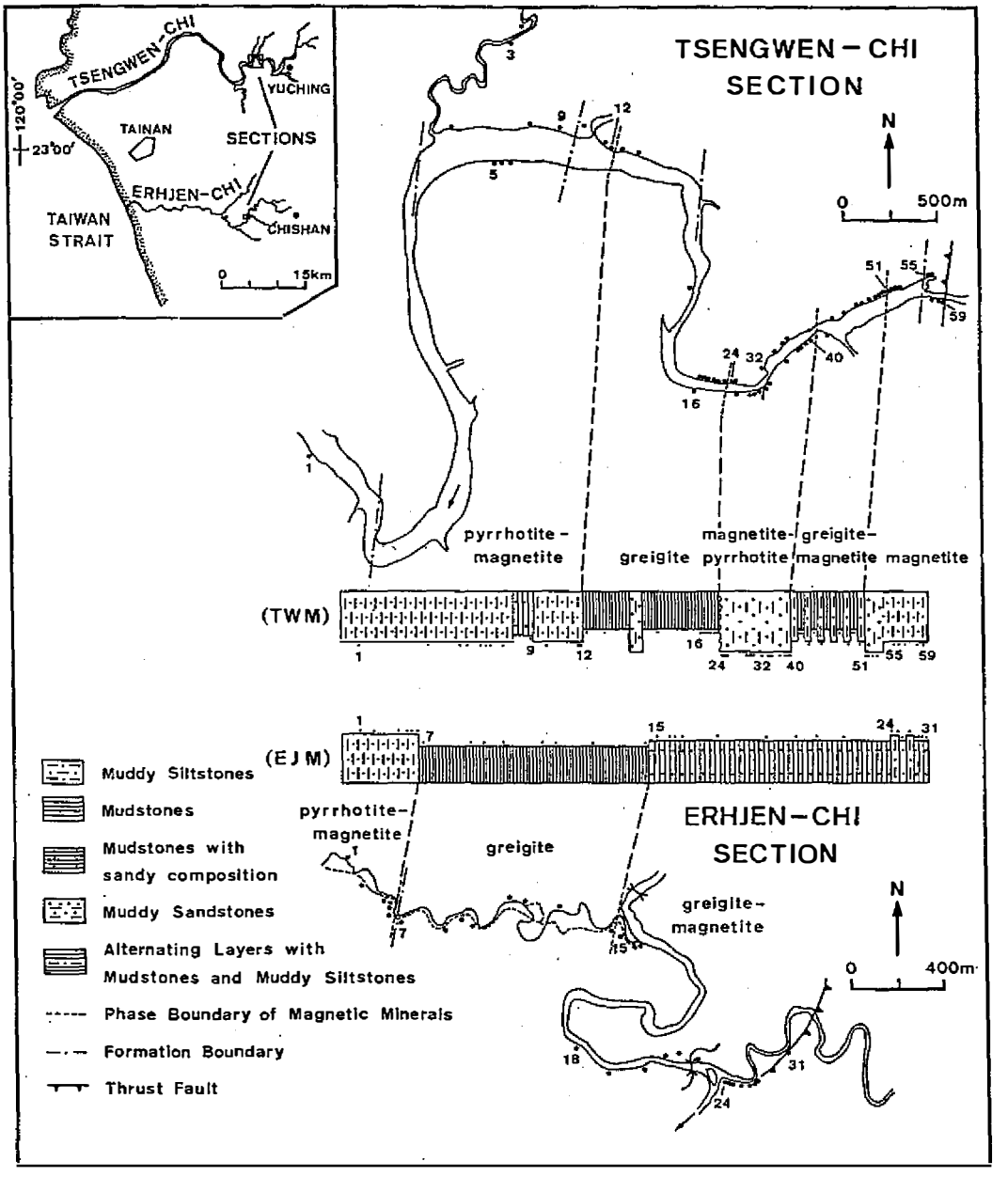

Fig. 1 Lithological distribution and magnetic mineral zonation of the Tsengwen-chi and Erhjen-chi sections. Both sedimentary sequences dip westward normally. Magnetic mineral zonation is based on magnetic mineral composition of 59 and 31 samples in each section (Horng *et al.*, 1992). For expressional reasons lithological columns are laid down horizontally.

applied and increased stepwise to make the specimens acquire magnetization. During each magnetization step, the isothermal remanent magnetization (IRM) of specimen was measured immediately with the spinner magnetometer. After acquiring SIRM in high fields (up to 1.5 Tesla), the specimen was applied again with stepwise increasing reverse fields. Its remanent magnetization was also measured after each step. The decreasing remanent magnetizations display a coercivity curve and Hcr, the field which is required to reduce SIRM to zero, can be obtained by interpolation. When another SIRM in the high reverse fields is acquired, the specimen's median destructive field of SIRM (MDF_{SIRM}) was determined by eliminating one-half of the SIRM through stepwise alternating field (a.f.) demagnetization procedures.

In order to obtain additional features and to show chemical changes of magnetic minerals, thermomagnetic analysis and thermal magnetic susceptibility measurement were carried out. Thermomagnetic analysis of magnetic extracts obtained from previous extraction (Horng *et al*, 1992) were performed by a horizontal magnetic balance, a design described in detail by Housden *et al.* (1988). It was mostly done in a nitrogen atmosphere to reduce oxidation. After the first cycle run (heating up to 650°C and then cooling down with a rate 20°C/minute), the products were identified with a Philips X- ray diffractometer. For thermal magnetic susceptibility variation, another set of cored specimens was put in a Schonstedt TSD-1 thermal furnace and heated in the air in stepwise increasing temperatures. After each cooling, bulk magnetic susceptibilities were measured with the Molspin minisep instrument.

3. RESULTS AND DISCUSSIONS

3.1 Rock Magnetic Properties

Table 1 lists the values of four magnetic parameter (κ, SIRM, Hcr, MDF_{SIRM}) of 28 specimens and their magnetic carriers. Representative diagrams of the acquisition of IRM are shown in Figure 2. It indicates that despite their different magnetic mineral composition (magnetite, pyrrhotite and greigite), nearly all of the specimens (27/28) obtained more than 90% of SIRM below 0.2 Tesla, a case clearly displayed on the inset of Figure 2. Because magnetite saturates easily below 0.2 Tesla and hematite does not saturate until field of over 1-3 Tesla, this nondestructive and convenient method was used effectively in the past to distinguish the presence of Ti-poor titanomagnetites (magnetites) and Ti-poor ilmenohematites (hematites), the two commonest magnetic minerals in terrestrial rocks (Tarling, 1983). Based on this principle, many workers adopted this method to interpret possible magnetic carriers or mixtures in their samples. Therefore, rock specimens having easy saturation behaviors as Figure 2 were generally considered to contain magnetites. Nevertheless, in this study we found that greigite- and pyrrhotite-bearing rocks also have easy magnetic saturation behaviors as magnetites. In other words, magnetic carrier interpretation should be approached cautiously when we deal with sedimentary deposits prior to real magnetic mineral identification.

In another aspect, the magnitude of SIRM value can be normally used as a concentration measure in a given magnetic mineral. Thus, for the greigite of the two sections (Table 1), we can conclude that there is more greigite concentration in the Erhjen-chi section where more fine-grained mudstones are distributed. Table 1 and Figure 2 also show that the SIRM values of greigite-bearing rocks are much higher than those of pyrrhotite-bearing rocks which again are usually higher, but to less of an extent, than those of magnetite-bearing rocks. Based on SIRM values and magnetic mineral occurrence, the concentration of greigite, pyrrhotite and magnetite in mudstones, muddy siltstones and muddy sandstones of the two sections, respectively, would be $C_{greigite} >> C_{pyrrhotite} >= C_{magnetite}$. This proposition is consistent with what we have observed in the magnetic extracts from different lithology (Horng *et al.*, 1992).

Table 1. Magnetic properties and magnetic carriers of sedimentary rocks from the Tsengwen-chi and Erhjen-chi sections.

Sample	κ (10^{-6} S.I.)	SIRM (10^{-3} A/m)	Hcr (mT)	MDF_{SIRM} (mT)	Magnetic Carriers
Tsengwen-chi section :					
					magnetite
TWM26	20.44	1130	57.5	31.0	M
TWM54	29.40	3405	61.0	34.5	M
TWM56	41.24	3745	50.0	20.0	M
TWM58	27.76	1196	38.0	17.5	M
TWM59	19.77	564	42.5	19.5	M
					pyrrhotite
TWM06	20.83	2358	51.0	32.0	P
TWM38	19.65	4367	62.0	38.5	P
					greigite
TWM14	22.79	8381	77.5	46.0	G>P>M
TWM18	30.41	9518	77.5	43.0	G
TWM19	44.59	24590	76.5	45.0	G
TWM20	31.68	17569	68.0	41.0	G
TWM36	29.49	15461	62.5	37.0	G
TWM45	35.31	21612	77.5	47.5	G>M
Erhjen-chi section :					
					pyrrhotite
EJM01	19.46	7030	87.5	58.5	P>>M
EJM04	16.21	2069	59.0	37.0	P
					greigite
EJM03	44.74	53294	91.0	57.0	G>>P
EJM07	30.86	30101	97.5	62.0	G>>P (?)
EJM09	55.20	90730	86.0	49.0	G
EJM10	49.62	46084	86.0	56.0	G
EJM11	48.91	55759	86.0	55.0	G
EJM12	41.47	15053	89.0	58.0	G
EJM17	31.44	16233	74.5	44.5	G,M
EJM19	22.49	5152	75.0	49.0	M>G
EJM21	50.35	69408	76.5	46.0	G
EJM25	57.07	50097	63.0	37.5	G>>M
EJM28	56.70	59011	73.0	43.0	G
EJM30	35.33	21661	76.5	46.0	G>M
EJM31	44.63	16920	69.0	41.0	G>M

The relationship between magnetic parameters κ and SIRM of these minerals is displayed in Figure 3. In greigite- and magnetite-bearing rocks, a rough trend is that higher κ values mostly correspond to higher SIRM values. But there is too little data in pyrrhotite group to make any inference. For a given κ value, it corresponds to a much higher SIRM value in greigite than magnetite. This results in very high SIRM/κ ratios for greigite-bearing rock, as have mentioned by Snowball and Thompson (1990) and Hilton (1990). The nonzero

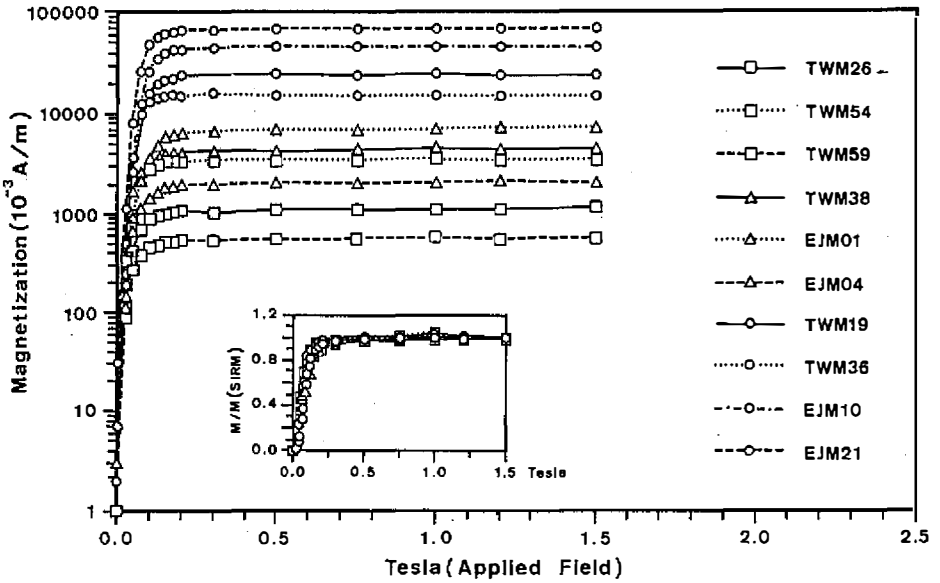

Fig. 2 Representative diagrams of acquisition of IRM for magnetite-, pyrrhotite- and greigite-bearing specimens, with symbol squares, triangles and circles, respectively. All of the SIRM are obtained below 0.2 Tesla in spite of different magnetic mineral constituent in the rocks. The inset shows their normalized IRM acquisition curves.

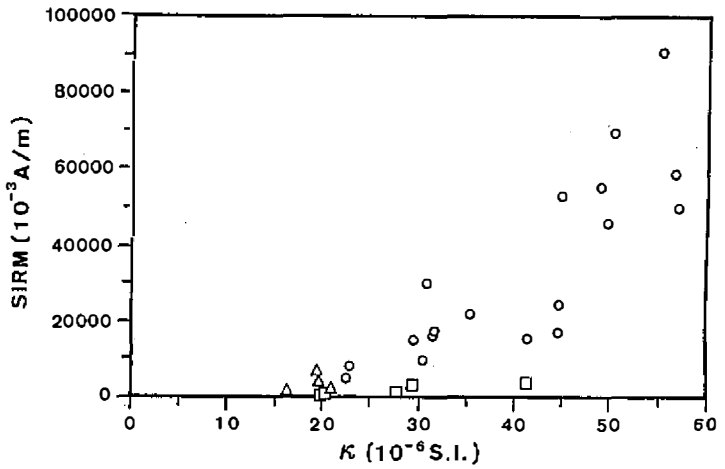

Fig. 3 κ versus SIRM plot, showing that higher κ values mostly correspond to higher SIRM values in greigite- and magnetite-bearing rocks. Symbols are the same as Fig. 2.

intercepts of κ further indicate that when magnetic minerals are a few percent or less, non-magnetic minerals, being the largest percentage material of rocks, can make partial contribution to magnetic susceptibility measurement. Therefore, κ parameter of sedimentary rocks seems not so sensitive as SIRM in magnetic mineral concentration.

In the introduction, we mentioned that magnetite, greigite and pyrrhotite occur in different grain-sized lithology of two sections. This seemingly implies that their particle size distribution is basically controlled by its host sediment. Although greigite and pyrrhotite are authigenic and were formed in the early stage of diagenesis (Horng *et al.*, 1992), the growth of crystals, probably through biogeochemical processes, should be limited by original porosity. For detrital magnetites, since more or less dissolution occurred on the minerals in early diagenesis (Canfield and Berner,1987; Karlin, 1990a and 1990b; Leslie, 1990), coarser-grained magnetites can survive after partial dissolution and therefore can still be found in muddy sandstones. However, the finer-grained magnetites which had suffered complete dissolution become scarce or absent in mudstones. Thus, based on magnetic mineral occurrence of the two sections, it is proposed that the average grain size of magnetite, pyrrhotite and greigite would be relatively coarse, intermediate and fine, respectively.

For magnetite, the relationship between magnetic domain sizes and remanent coercivities has been well established. Magnetite, ranging from about 0.1 micron (stable single domain region) to hundred microns (multidomain region), shows increasing Hcr values as the grain size decreases (Thompson and Oldfield, 1986). If the principle is still valid for greigite and pyrrhotite, we will expect higher Hcr values in them, due to their finer grain sizes than that of magnetite. Figure 4 presents typical coercivity spectra of the three magnetic mineral-bearing rocks. As we have proposed, Figure 4 and Table 1 all indicate that values of Hcr are high (63-98 mT) in greigite, intermediate (51-88 mT) in pyrrhotite and low (38-61 mT) in magnetite-bearing rocks. Another grain-size dependent parameter, MDF_{SIRM}, also indicates a similar result (greigite : 37-62 mT; pyrrhotite : 32-59 mT; magnetite : 18-31 mT) (see Figure 5 and Table 1). In addition, we found that there is a linear correlation between Hcr and MDF_{SIRM} (Figure 6), further indicating that either parameter can be used to verify the variation of magnetic grain sizes.

3.2 Thermal Magnetic Properties

Thermomagnetic analysis of magnetic extracts with magnetites, pyrrhotites and greigites reveals not only their diagnostic characters but also thermochemical reactions in them (Figure 7). For magnetites (Figure 7a), the induced magnetization on heating decreased gradually to zero around 570-580°C, implying that they were mainly composed of almost pure or titanium-poor magnetites. However, the irreversible cooling curve also indicated that certain chemical reactions had occurred. Although the possible reactions are various and depend on factors such as the purity of magnetic extracts and the heating conditions etc., it is suggested that for the case of Figure 7a, the lower induced magnetization on cooling was probably due to further oxidation of some magnetites to hematites (Tarling, 1983). For the heating curve of pyrrhotites (Figure 7b), it displayed rapid decrease in magnetization below 320°C and then showed a small hill-shaped curve above this temperature. Upon cooling, much higher induced magnetization was acquired. All of these indicate that pyrrhotites might be destroyed and transformed to other magnetic phases in the cycle. X-ray analysis showed that most of them had changed into magnetites with only small amounts of residual pyrrhotites (Figure 8a). In greigites (Figure 7c), the magnetization upon heating decreased slowly below 300°C and then dropped down rapidly between 300-440°C. Over this temperature range, it decreased slowly again. Upon cooling, the magnetization was lower and showed an inflection around 300°C. X-ray analysis indicated that greigites had been changed into major pyrrhotites and minor magnetites (Figure 8b).

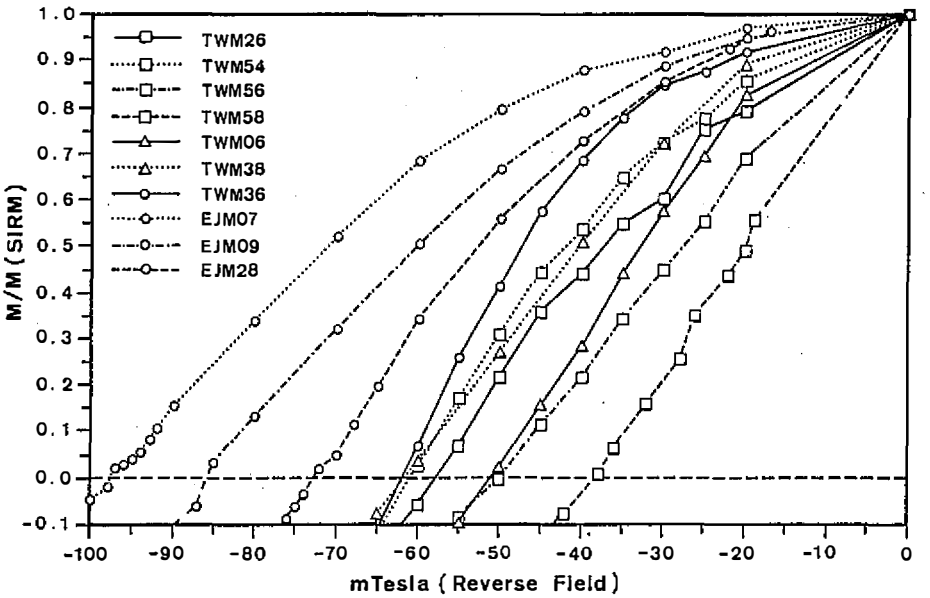

Fig. 4 Typical coercivity plots of SIRM versus reverse applied field for magnetite-, pyrrhotite- and greigite-bearing specimens. Symbols are the same as Fig. 2. Their coercivity forces (Hcr) can be determined when magnetizations reduce to zero.

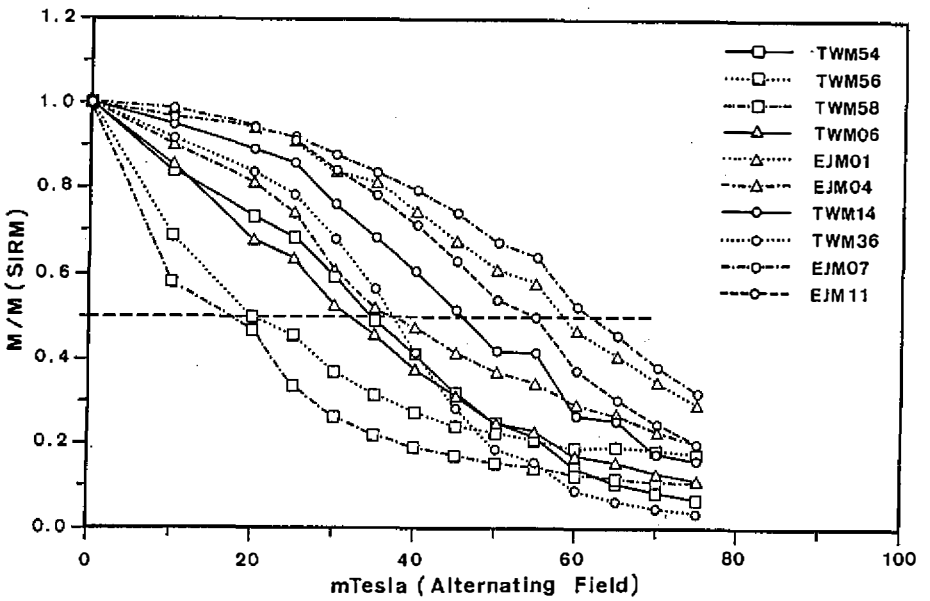

Fig. 5 Typical normalized alternating field (a.f.) demagnetizing curves of SIRM for magnetite-, pyrrhotite- and greigite-bearing specimens. Symbols are the same as Fig. 2. MDF_{SIRM} is the field strength which is required to eliminate one-half of SIRM.

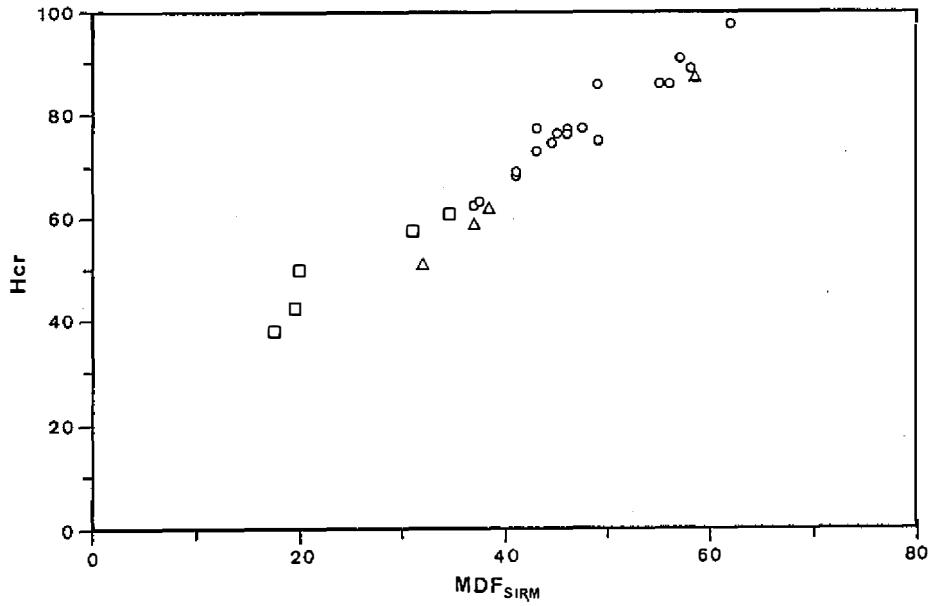

Fig. 6 MDF$_{SIRM}$ versus Hcr plot, showing a linear relationship between them. Symbols are the same as Fig. 2.

Another method which can be used to detect phase changes of these three magnetic minerals in the heating process is to monitor their bulk magnetic susceptibility variations. Some typical changes are presented in Figure 9. Because greigite is the least stable among the three magnetic minerals (Berner, 1971), it is expected to be sensitive to the thermal magnetic susceptibility change. As shown in Figure 9, greigite-bearing rocks have remarkable drop below 320°C, while those of pyrrhotite- and magnetite-bearing rocks respectively show minor and almost noneffective changes until 400°C. It is postulated that the susceptibility variations in greigite- and pyrrhotite-bearing rocks in low temperatures (<320°C or 400°C) are mainly due to magnetic mineral instability. However, the changes at high temperatures (>320°C or 400°C) may result from thermochemical reactions of the three magnetic minerals or of some non-magnetic minerals such as pyrites and Fe-chlorites.

4. SUMMARY

1. In the Tsengwen-chi and Erhjen-chi sections, greigite- and pyrrhotite-bearing rocks have easy magnetic saturation behaviors as magnetites. Most of them obtain more than 90% of SIRM below 0.2 Tesla.
2. For the basic magnetic parameters (κ, SIRM, Hcr, MDF$_{SIRM}$) of the two section sedimentary deposits, we obtained relatively high values in greigite-bearing rocks, intermediate values in pyrrhotite-bearing rocks and low values in magnetite-bearing rocks. These rock magnetic characters are related to their different magnetic mineral concentration and grain sizes.
3. Thermomagnetic analysis in a nitrogen atmosphere has further revealed that each magnetic mineral has its own diagnostic magnetization curve and that greigite may be transformed to pyrrhotite and magnetite, pyrrhotite to magnetite, and magnetite to hematite.

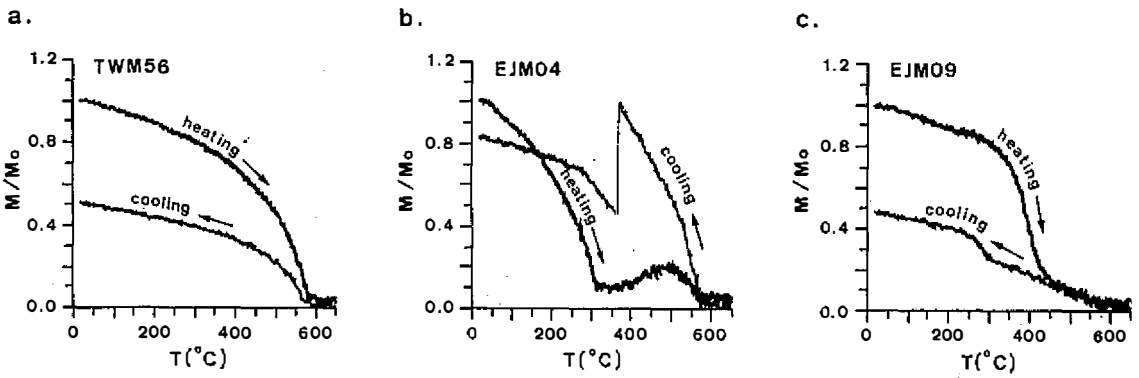

Fig. 7 Thermomagnetic curves of magnetite (a), pyrrhotite (b) and greigite (c) in a nitrogen atmosphere with a heating-cooling rate 20°C/minute. Magnetizations (M) were normalized by dividing the initial value (M_o, room temperature). In Figure 7b, the interruption of cooling curve at 370°C is due to a man-made break of applied field to avoid an over-scale of increasing magnetization.

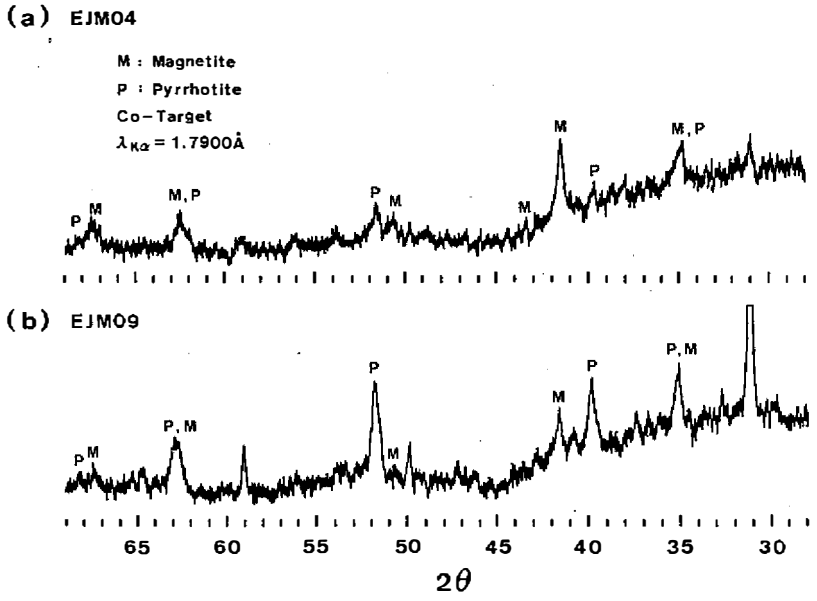

Fig. 8 X-ray diffraction spectra, revealing that after the first cycle run of thermomagnetic analysis, most of pyrrhotite has been transformed to magnetite (a), and greigite to pyrrhotite and magnetite (b). The original pyrrhotite and greigite X-ray diffraction patterns before thermal treatment had been shown in the previous study (Horng *et al.*, 1992).

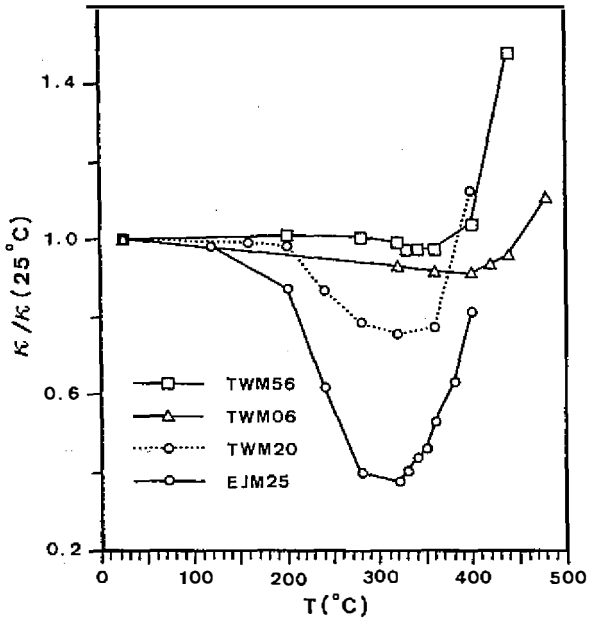

Fig. 9 Variations of normalized bulk magnetic susceptibility on heating for magnetite-, pyrrhotite- and greigite-bearing specimens. Symbols are the same as Fig. 2.

Bulk magnetic susceptibility has also shown remarkable drop in greigite-bearing rocks below 320°C, while there are minor and almost noneffective susceptibility changes in pyrrhotite- and magnetite-bearing rocks below 400°C, respectively.

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臺灣西南部曾文溪及二仁溪剖面沈積岩的磁學特性

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摘要

本研究是以四種基本磁性參數(κ, SIRM, Hcr, MDF_{SIRM})及兩種熱磁方法尋求曾文溪及二仁溪剖面中具有磁鐵礦、磁硫鐵礦及等軸磁硫鐵礦（暫譯）之岩石磁學特性。儘管岩石所含的磁性礦物相異，但等溫殘磁磁化的獲取行爲卻非常相似。由於其磁性礦物之含量與粒徑不同，因而使帶有等軸磁硫鐵礦、磁硫鐵礦及磁鐵礦之岩石各具有高、中、低的相對磁性參數值(κ, SIRM, Hcr, MDF_{SIRM})。在氮氣中的熱磁分析進一步顯示每種磁性礦物有獨特的熱磁曲線，並且在加溫時等軸磁硫鐵礦可轉變成磁硫鐵礦及磁鐵礦；磁硫鐵礦變成磁鐵礦；而磁鐵礦則變成赤鐵礦。此外，具有等軸磁硫鐵礦之岩石在加熱至320°C之前，其磁感率呈明顯的下降；而具有磁硫鐵礦及磁鐵礦之岩石在加熱至400°C之前，其磁感率各自有“輕微”和“幾乎不受影響”之變化效應。