Preliminary study of magnetic properties and magnetic mineralogy of the limestone samples from Egyptian Pyramids and Giza Pyramids Plateau

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Abstract: The limestone samples of the Eocene to Pliocene age from the pyramids and quarries have been studied by the magnetic methods. It has been revealed that the arsenic containing scorodite mineral is a dominant carrier of magnetic properties in the studied materials. This mineral is of a diamagnetic state in its original form. During its heating the transformation takes place accompanied by the change of magnetic susceptibility. Over the 300 °C the transformation of scorodite into maghemite and hematite took place and so the increase of magnetic susceptibility and the intensity of partial thermoremanent magnetization were detected.

Key words: scorodite, its transformation to Fe oxide

1. Introduction

The samples taken from the Egyptian Pyramids and those from the limestone quarries of the Giza Pyramids Plateau have been studied. We have applied some magnetic methods which may help solving the problem concerning building technology of the Egyptian Pyramids. According to a generally accepted idea the individual blocks of the pyramids were carved, transported from the nearest limestone quarries and hoisted on the necessary place of the pyramid. French chemist, Joseph Davidovits, rejected such an idea and proposed that the building method had involved the moulding on site: blocks were made of a kind of concrete whose basic binding

179

compound was natron, extracted from the site very close to Giza (*Demortier, 2009*). The age of the limestones in the Giza area is of the Eocene to the Pliocene (*Aigner, 1993*). Apart from other magnetic methods, the change of the partial thermoremanent magnetization (PTRM) and magnetic susceptibility of the samples with temperature was applied. The methodic procedure and the results of the study are presented below. First, however, we shall describe our experience from the shaping of the samples.

Shaping of the samples

The larger pieces of samples were collected either from the limestone quarries or from the respective Egyptian Pyramids. The orientation of samples in the field was identified using the compass. The samples were shaped to the cylindrical form in the laboratory. During the mechanical treatment of the samples, namely during their cutting by the saw or drilling of the samples, a typical smell of garlic was released. Such peculiarity was detected in the case of samples from all 20 localities. According to the analyses presented by *Demortier (2009)* there was detected a presence of some ore mineral *SCORODITE*, except other components. This mineral ore containing arsenic was added to produce sodium arsenate acting as an activating ingredient that could have been used in various concentrations to control the speed of the hydraulic setting (*Demortier, 2009*).

The arsenic containing compounds

According to *Betechtin (1955)* the scorodite mineral ($Fe[AsO_4].2H_2O$; in Greek scorodon = garlic) is a source of the garlic's smell in the respective samples. There have been known various As minerals which have occurred in the ore fields of hydrothermal activity on the Earth: pure arsenic with the Sb, Ag, Fe, Ni, Bi and V metals. Realgar – AsS; Auripigmentum – As_2S_3; Lölingtite – $FeAs_2$; Arsenopyrite – FeAsS; Allemontite – AsSb; Arsenolite – As_2O_3; Claudetite – As_2O_3; Enargite – Cu_3AsS_4; Farmakosiderite – $Fe_3[AsO_4]_3[OH]_6.6H_2O$; Ferrisymplezite – $Fe_3[AsO_4]_2[OH]_3.6H_2O$; Tenantite – Cu_3AsS_3. The mentioned arsenic containing minerals are the carriers of a garlic's smell, after mechanical or temperature treatment. These minerals have frequently occurred along with presence of sulphides of metals and sometimes with the Au and Ag ores, in the hydrothermal occurrences.

180

Table 1. The stepwise inducing of PTRM of the limestone samples in the magnetic field of intensity $B = 48\mu$ T, and the parallel detection of the change of magnetic susceptibility (κ) at temperatures from 25 to 650 °C

T, °C / Sample	25	50	100	150	200	250	300	350	400	450	500	550	600	650
1-3 P	65	99	60	87	115	232	330	503	736	1083	1668	3121	4638	5482
	-1.9	-1.9	-1.8	-2.3	-1.8	-2.2	-2.9	-2.7	-1.6	-0.5	-0.6	0.24	-0.3	4.2
2-5 P	44	64	67	67	61	67	81	104	166	250	385	736	1103	1146
	-1.8	-1.6	-1.8	-2.3	-2.2	-2.0	-2.7	-3.3	-2.8	-3.4	-2.0	-3.0	0.1	-0.9
3-7 P	34	45	25	37	85	164	328	502	549	852	2277	6990	24204	46337
	11	10	11.6	11	11	8.5	8.6	9	9	10	13	23	294	867
4-6 Q	56	79	103	112	154	205	301	316	478	955	2191	5350	8248	10589
	1.8	1.9	1.8	2	0.1	0.2	-0.8	-0.9	-2.2	-0.3	1.6	8.5	59	49
5-6 P	59	48	45	79	86	104	151	222	343	502	721	1454	2070	2138
	-4.6	-4.4	-4.6	-4.1	-4.2	-4.5	-5	-5.7	-5.7	-4.8	-3	-3.5	0.8	3.9
6-2 P	323	410	403	432	467	480	584	709	1144	1701	5146	7447	10927	13402
	13	13	12.9	12.7	12	9.2	6.8	8.1	8.5	9.5	13.5	14	30	114
7-4 P	14	40	70	80	128	161	255	364	529	1012	2581	4593	13001	11011
	-6.3	-7	-7.2	-7.7	-7.8	-7.9	-8.6	-7.8	-6.3	-4.4	-3	3.8	175	178
8-5 P	1462	1432	1455	1494	1502	1458	1494	1572	1756	2191	4188	10768	33484	38946
	5.1	5.4	4.8	4.4	4.7	1.7	-0.2	0.2	0.7	2.3	5.9	13.3	31	49
9-5 P	50	47	76	53	81	80	135	199	314	648	2961	10600	286041	38734
	0.7	0.5	0.1	-1.0	-1.0	-1.8	-3.2	-2.5	-2	0.1	6.8	20	48	77
10-4 P	9	28	49	36	41	36	41	60	67	92	213	353	441	603
	1.8	1.8	1.0	1.4	1.6	0.5	-1.2	-1.8	-1.4	-0.5	-1.4	-0.2	1.8	37
11-3 P	45	35	21	21	53	139	270	474	695	1143	2294	5032	8081	9952
	3.1	2.4	3.7	3	2.6	2.1	2.8	4.2	4.5	6.3	6.3	113	214	104
12-3 P	25	39	27	28	61	159	276	517	496	674	1712	5593	9419	11576
	7.6	6.5	7.3	7.8	6.9	5.6	5.2	6.5	7.8	1.6	14.8	94	221	104
13-9Q	50	44	67	60	101	95	178	277	421	717	2297	5345	11249	13741
	-1.9	-2.0	-2.4	-3.1	-2.5	-3.6	-4.7	-3.6	-1.6	0.7	1.8	5.2	87	198
14-10Q	683	675	19	39	100	178	256	325	657	1533	2213	10843	16461	19538
	11.6	10.8	12	11.8	11.3	10.4	9.4	9.2	9.9	13.8	14.9	72	266	104
15-2 Q	217	335	323	294	316	208	248	300	522	1170	4064	17808	40661	49277
	10.3	9.8	9.0	9.5	8.8	8.3	6.1	5.3	7.5	11	17	28	48	233
16-5 Q	36	60	33	46	72	221	252	347	557	757	2537	5182	15030	19508
	3.2	2.3	3.2	3.0	2.2	1.6	1.6	2.1	3.9	4.7	5.9	8.8	113	249
17-6 Q	38	114	58	52	95	210	248	378	613	941	1716	4754	12538	15086
	2.7	1.9	2.7	2.7	2.2	1.7	1.2	2.4	3.9	5.5	6.4	9.8	242	162
18-5 Q	126	127	131	113	120	135	207	295	359	621	845	3736	7510	12095
	-6.1	-6.4	-6.1	-6.5	-6.7	-7.3	-7.4	-6.6	-6.1	-3.1	-1.5	12	50	230
19-5 Q	44	31	77	67	89	139	147	193	282	374	643	2683	5115	8358
	-3.5	-4.4	-4.1	-3.4	-4.0	-4.1	-3.9	-5.3	-4.4	-2.3	0.3	12	75	291
20-6 Q	31	35	16	21	46	116	131	184	355	556	925	2519	5041	8365
	-7.6	-7.6	-7.6	-7.6	-7.6	-8.1	-8.1	-7.8	-7.2	-5.5	-1.6	8.2	40	152

P–pyramide; Q–quarry; PTRM–partial thermoremanent magnetization in mA/m (the first line-up for each sample); B–magnetic induction of ambient magnetic field; bulk magnetic susceptibility (κ in 10^{-6} SI Units, the second line for each sample); 25, 50, 100, 150,– 600, 650 – temperature intervals in °C, at which inducing of PTRM of samples during 30 minutes was performed.

The stepwise inducing of partial thermoremanent magnetization (PTRM) of the limestone samples in the magnetic field of intensity $B = 48\mu\,\mathrm{T}$ at the temperatures from 25 to 650 °C

This part deals with the results of the thermal inducing of partial thermoremanent magnetization (PTRM) and the change of magnetic susceptibility (κ) with temperature of the limestone samples in the magnetic field of intensity $B = 48\mu\,\mathrm{T}$ at the temperatures from 25 to 650 °C.

The procedure was as follows: The sample of the cylindrical shape was placed on the holder (it is in the middle centre of the non-magnetic oven during heating of the samples) in an oriented direction with respect to the magnetizing field. The sample was heated in the non-magnetic oven to a specified temperature, e.g. 100 °C. The sample was heated at each temperature over 30 min. interval. After heating the sample was cooled to the laboratory temperature. During the whole procedure the PTRM was induced and the PTRM and magnetic susceptibility (κ) were measured at the laboratory temperature. These procedures were performed in the presence of magnetic field of $B = 48\mu\,\mathrm{T}$. The results have been summarized in Table 1, and some selected results are pictured in Figs. 1–7.

2. Evaluation of the results

The basic data are shown in Table 1. From the basic bulk magnetic susceptibility (κ) it is evident that there are no ferro- or ferri- magnetic materials in the samples from the pyramids and from those of the limestone outcrops. The samples from 4 locations in pyramids and from 4 limestone quarries have shown the very low negative values κ and from 7 locations in pyramids and from 6 limestone quarries have shown mostly low positive values of κ. These values correspond to a presence of the diamagnetic to paramagnetic material in the samples. The Q coefficients computed for the samples with a positive κ at 25 °C are in the range of 0.8 to 18.0 (an anomalous value Q =72, for the non oriented samples of the location 8 from the pyramide). Despite the very low κ values, the natural remanent magnetization (NRM) is relatively well preserved. The directions of the original NRM are mostly remagnetized at the temperatures of 150 to 250 °C, where the PTRM has acquired a direction of the inducing laboratory field (see Figs. 1–7). The

Chefren's Pyramide1-11;NRM=
122 mA/m

Chefren's Pyramide2-5;NRM=
44 mA/m
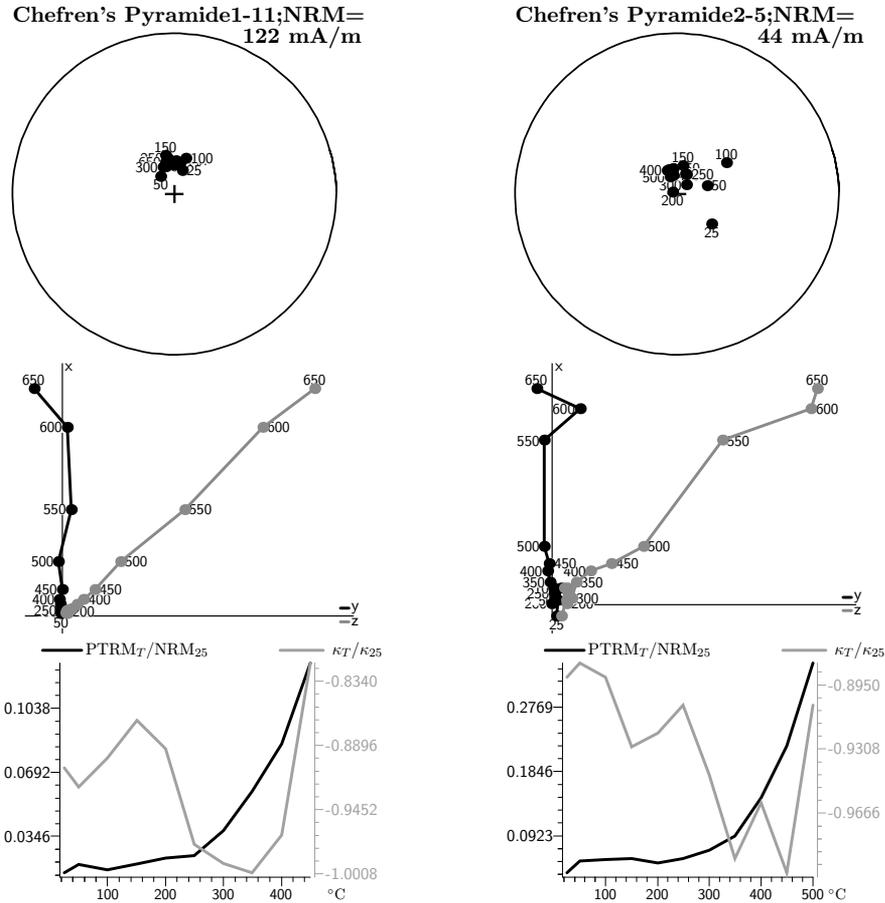
Fig. 1. Thermal inducing of PTRM of the samples, stepwise heating to 650 °C; Zijderveld diagrams and stereographic projections; ● (●) means positive (negative) polarity of RM; κ_T (κ_{25}) – magnetic susceptibility at T (at 25 °C, respectively); $PTRM_T$ – partial thermoremanent magnetization at T, NRM_{25} – natural remanent magnetization at 25 °C.

original NRM of the samples from the limestone quarries should be of the depositional-detrital origin. In some sediments there may be also a chemical – CRM origin of magnetization having been produced some time after deposition, during chemical alteration in the process of consolidation (*Stacey and Banerjee, 1974*).

Cheops's Pyramide6-2;NRM= 323 mA/m

Egyptian Pyramide10-4;NRM= 9 mA/m
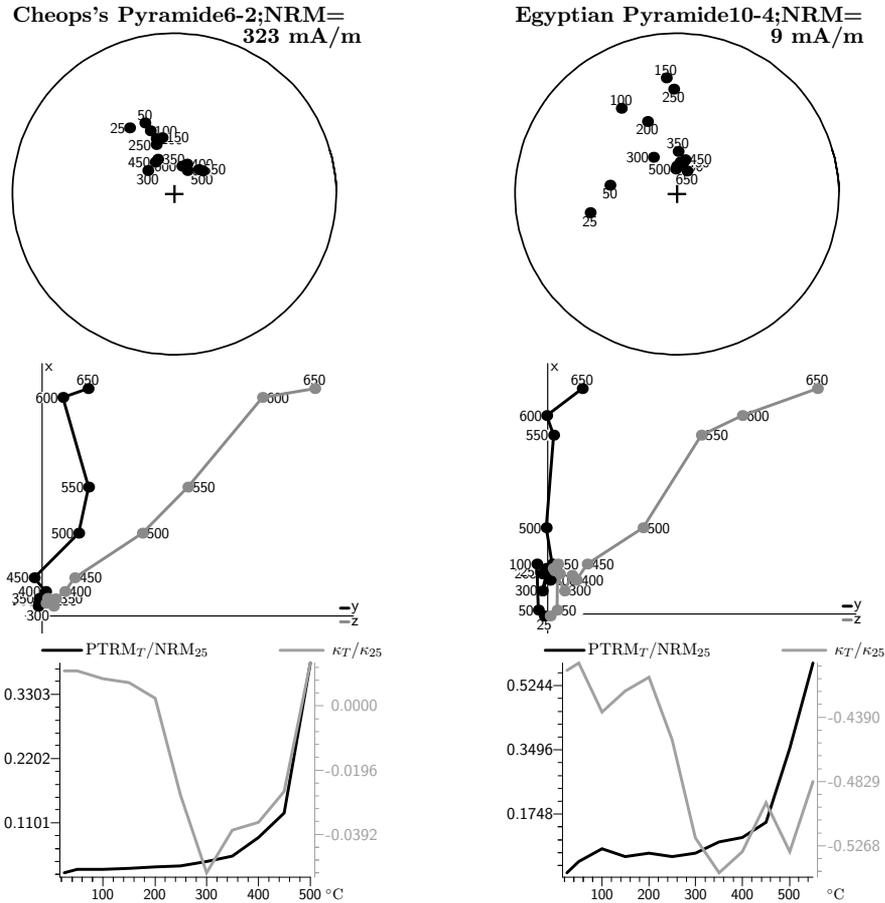
Fig. 2. Thermal inducing of PTRM of the samples, stepwise heating to 650 °C. Zijderveld diagrams and stereographic projections; for other explanations see in Fig. 1.

What is a carrier of magnetism in the limestone samples?

So far no specific analyses of the samples have been done. We can rely on the results published by *Demortier (2009)*. The author presented the analyses, according to which some ore mineral **scorodite** was detected, except for other components in some samples from the pyramids. This mineral is of chemical composition $Fe[AsO_4].2H_2O$. It has Fe^{3+} ions and it contains also 2 molecules of water. This mineral is supposed to be of paramagnetic

184
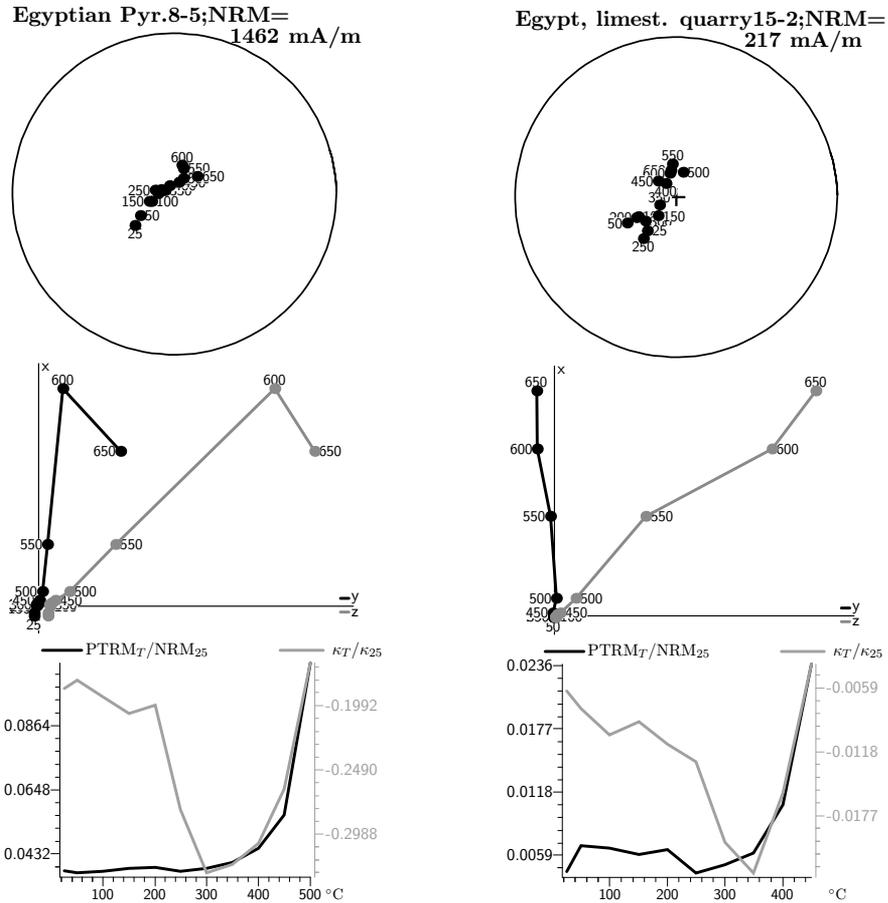
Fig. 3. Thermal inducing of PTRM of the samples, stepwise heating to 650 °C; Zijderveld diagrams and stereographic projections; for other explanations see in Fig. 1.

behaviour. It is unstable material, namely in oxidizing conditions.

We see from Figs. 1–7 that there is a typical behaviour of κ and PTRM of all samples during their stepwise heating. The κ mostly gradually decreases to the temperature interval ranging from 300 to 400 °C, but at higher temperatures the κ gradually increases. The highest value of κ was achieved mostly at 650 °C. After repeating the temperature treatment with the samples which underwent the first magnetizing cycle, no further increase of κ

185

Chefren's Pyramide3-7;NRM=
34 mA/m

Cheops's Pyramide5-6;NRM=
59 mA/m
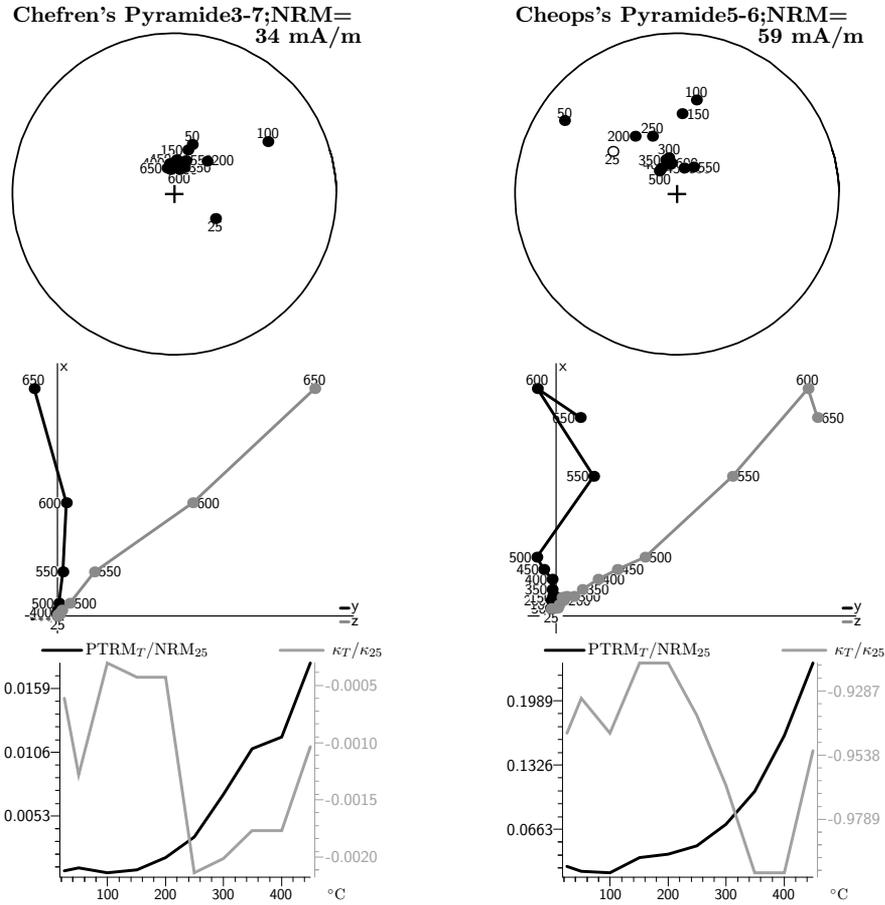
Fig. 4. Thermal inducing of PTRM of the samples, stepwise heating to 650 °C; Zijderveld diagrams and stereographic projections; for other explanations see in Fig. 1.

was observed. The behaviour of PTRM curve does not correspond to that of behaviour of κ curve. An increase of PTRM has started around of 200 °C and then increased mostly to high values. The Q coefficients computed for the individual samples at 650 °C have reached the values in the range of 13.4 to 200. It means that the PTRM is more sensitive compared with the change of κ during temperature treatment.

   A macroscopic observation of the colour of the samples indicates that

186
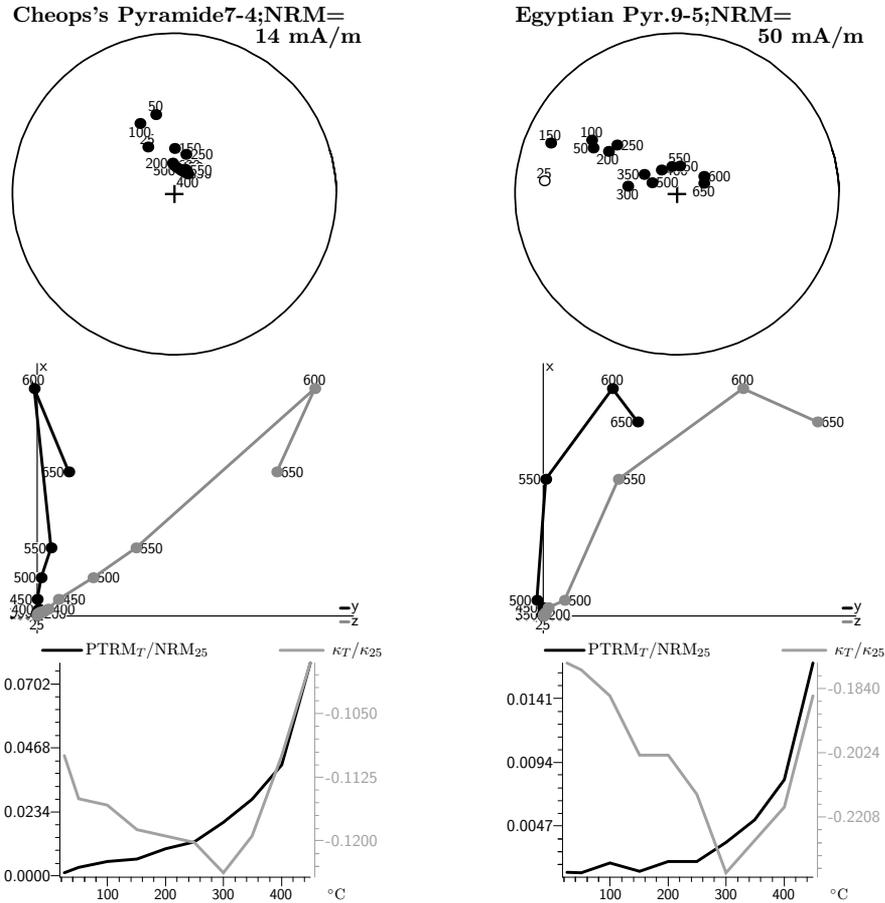
Fig. 5. Thermal inducing of PTRM of the samples, stepwise heating to 650 °C; Zijderveld diagrams and stereographic projections; for other explanations see in Fig. 1.

the samples were getting reddish over 200 °C during temperature treatment. This was also a serious signal of the chemical changes of minerals in the samples. It has been described above that the directions of PTRM of samples were concentrated to a direction of laboratory field mostly at temperatures from 250 to 650 °C. The soft reddish colour of samples after 200 °C, the increased κ, enriched values of PTRM, high values of Q after temperature treatment at 650 °C and the directions of PTRM have shown that there was

Egypt,limest.quarry13-9;NRM=
50 mA/m

Egypt,limest.quarry16-5;NRM=
36 mA/m
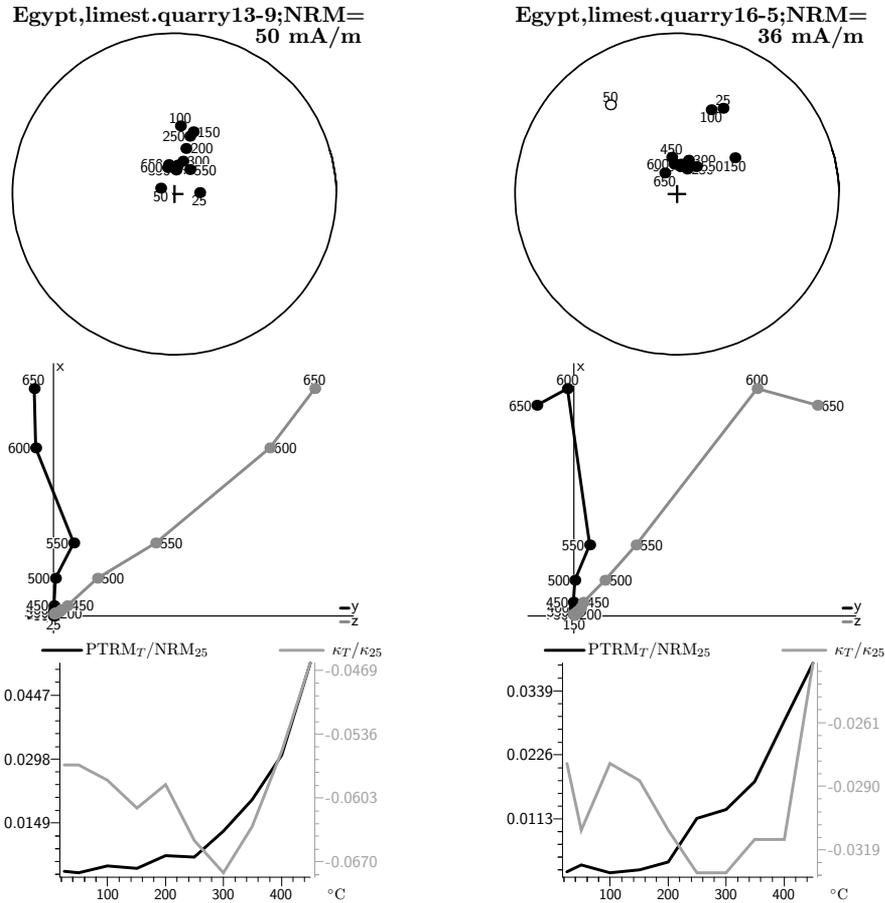
Fig. 6. Thermal inducing of PTRM of the samples, stepwise heating to 650 °C; Zijderveld diagrams and stereographic projections; for other explanations see in Fig. 1.

created a new secondary magnetic mineral from the original scorodite (the arsenic bearing compounds, including scorodite are of a diamagnetic origin, according to *Vonsovskij (1971)*). During the heating a transformation of scorodite into the maghemite and hematite is in following way: around 150 to 200 °C the scorodite $Fe[AsO_4].2H_2O$ releases firstly $2H_2O$ and then Fe^{3+} ion. This process is accompanied with a decrease of magnetic susceptibility of samples. The mineral acquires gradually more diamagnetic behaviour.
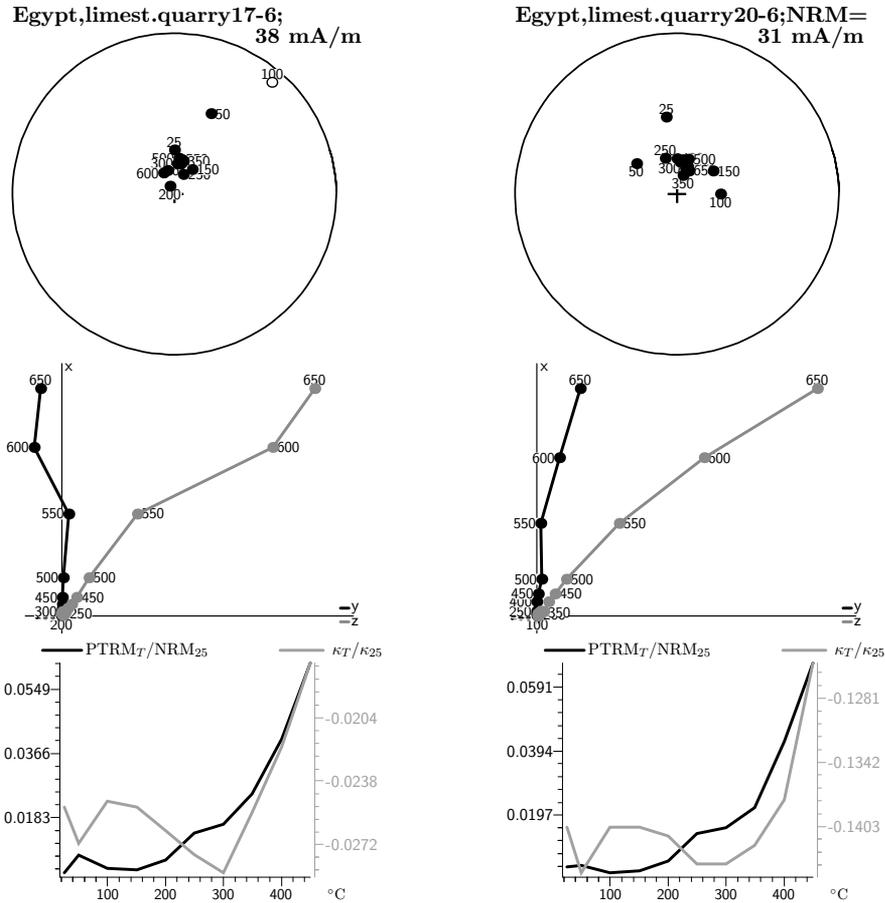
Fig. 7. Thermal inducing of PTRM of the samples, stepwise heating to 650 °C; Zijderveld diagrams and stereographic projections; for other explanations see in Fig. 1.

At the same time, there is a creation of Fe_2O_3 oxide with typical magnetic behaviour which acquires the high value of magnetic susceptibility and the PTRM intensity, at about 650 °C. AsO_4 from coordinating sphere of original scorodite was disintegrated in oxidizing conditions and completely vanished (according to *Betechtin (1955)* arsenic vanishes at about 360 °C. It is unstable in oxidizing conditions).

The behaviour of the thermomagnetic curves in Figs. 1–7 have reflected

189

the changes in the original material in favour of the secondary transformed hematite. Despite the investigated samples have shown very similar behaviour in Figs. 1–7, from Table 1 it is evident that the values of magnetic susceptibilities and PTRM intensities at 650 °C featured very different concentrations of the original material. While the samples 1–3, 2–5, 5–6, from the pyramids have reached κ in the range -0.9 to 3.9×10^{-6} SI Units and PTRM in the range 1146 to 5482 mA/m at 650 °C, for other samples κ are in the range 37 to 867×10^{-6} and PTRM intensities in the range 8358 to 49277 mA/m. One could deduce that during the moulding technique with a grinded limestone material, pouring the water and so on, there would be expected a leakage of the original material and decrease in its concentration, but in the compact limestones from quarries it should be completely preserved.

The above presented results are preliminary ones. They will be complemented in the near future.

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