# Self-reversal of chemical remanent magnetization during multiphase oxidation of synthetic titanomagnetites

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[1] The possibility of self-reversal of chemical remanent magnetization (CRM) is studied on synthetic samples of titanomagnetites ($Fe_{3-x}Ti_xO_4$, where $x = 0.2$, 0.4, and 0.6) that experienced multiphase oxidation in air at $500°C$ in the presence of the constant magnetic field $H = 0.1$ mT. The self-reversal was fixed in accordance with anomalous behavior of CRM curves of cooling (in $H = 0$) and heating (in $H = 0$). Complete self-reversal of CRM is discovered experimentally in samples of oxidized titanomagnetites with initial $x = 0.6$, while partial self-reversal is revealed in other titanomagnetite compositions. Superposition of a partial thermoremanent magnetization (pTRM) on CRM showed that heating curves of CRM+pTRM are also anomalous, but negative values of the total magnetization are not observed. *INDEX TERMS:* 1500 Geomagnetism and Paleomagnetism; 1518 Geomagnetism and Paleomagnetism: Magnetic fabrics and anisotropy; 1519 Geomagnetism and Paleomagnetism: Magnetic mineralogy and petrology; 1533 Geomagnetism and Paleomagnetism: Remagnetization; 1540 Geomagnetism and Paleomagnetism: Rock and mineral magnetism; 3630 Mineralogy and Petrology: Experimental mineralogy and petrology; *KEYWORDS:* titanomagnetite, oxidation, magnetization, self-reversal.

## Introduction

[2] Self-reversal (the phenomenon orienting the remanent magnetization in the direction antiparallel to an external magnetizing field) is a phenomenon alternative to an Earth's magnetic field reversal as a factor responsible for a reversed direction of the natural remanent magnetization (NRM or $J_n$) in rocks. In this respect, an anomalous magnetization of rocks can lead to an incorrect idea concerning the variation of the ancient geomagnetic field and, therefore, an inadequate interpretation of magnetic data obtained from paleomagnetic studies of the rocks. This is why elucidation of the origin of the NRM self-reversal effect and prediction of this phenomenon are of great importance for solving paleomagnetic problems (in particular, the identification of spurious paleomagnetic zones).

[3] L. Néel [*Néel, 1951*] was the first who substantiated theoretically the possibility of self-reversal with reference to thermoremanent magnetization (TRM or $J_{rt}$) and this undoubtedly stimulated the interest of paleomagnetologists in experimental studies of this phenomenon. Cases of the

TRM self-reversal were discovered in rocks the ore fraction of which is represented by assemblages of minerals belonging to isomorphous series and to series of the solid solutions hematite–ilmenite (hemoilmenite) [*Bina et al., 1999; Hoffmann and Fehr, 1996; Kennedy, 1981; Minibaev et al., 1965; Nagata, 1961; Ozima and Funaki, 2001; Ozima et al., 2003; Prevot et al., 2001*] and magnetite–ulvospinel (titanomagnetite) [*Gapeev and Gribov, 2008a; Havard and Lewis, 1965; Heller, 1980; Hoffman, 1982; Ozima and Larson, 1968; Trukhin et al., 2004; Westcott-Lewis and Parry, 1971; Zhilyaeva et al., 1970, 1971*]. The effect of anomalous behavior in $J_{rt}(T)$ was also observed on synthesized analogues of these ferromagnets [*Bugaev et al., 1972; Gapeev and Gribov, 2002b; Havard and Lewis, 1965; Lewis, 1968; Petersen and Bleil, 1973; Tucker and O'Reilly, 1980; Trukhin et al., 1997; Zvegintsev and Grankin, 1973*]. However, until recently the self-reversal effect has been very poorly studied in relation to the chemical remanent magnetization (CRM or $J_{rc}$) directly associated with magnetomineralogical alterations of rock-forming minerals at various stages of their evolution. In general, one may state that CRM is the least studied and difficultly diagnosable NRM component mainly because many stages of its formation (particularly at relatively low temperatures of reactions) cannot be reproduced on usual laboratory time scales.

[4] At present, the reality of the total CRM self-reversal has been demonstrated experimentally only in two case studies of the maghemite-hematite transformation [*Hedley, 1968;*

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*McClelland and Goss,* 1993]. Under the conditions of lacking information on the recurrence extent of the CRM self-reversal, it is desirable and appropriate to conduct long-term laboratory experiments designed for the creation of CRM and examination of its possible self-reversal using real types of reactions involving magnetomineralogical alterations of main magnetic carries in rocks.

[5] This work continues our cycle of investigations devoted to experimental simulation of titanomagnetite multiphase transformation processes (oxidation and decomposition) and the study of properties of related remanence types in newly formed phases. In particular, experiments on multiphase oxidation of synthetic titanomagnetites ($Fe_{2.4}Ti_{0.6}O_4$) revealed the possibility of the partial TRM self-reversal in two temperature intervals. The partial TRM self-reversal in a high temperature interval is realized in the spinel phase due to magnetostatic interaction between regions of different magnetic properties because of both heterogeneity of composition and the presence of stresses, while a low temperature self-reversal (near the Curie point $T_C$ of newly formed hemoilmenite) is caused by interphase interaction between spinel and rhombohedral phases: during cooling of titanomagnetites subjected to multiphase oxidation, lamellae of the low temperature hemoilmenite phase are thermally magnetized in the demagnetizing field of the higher temperature spinel phase (however, in discussing the effect, we did not exclude the possibility of anomalous TRM behavior in the rhombohedral phase itself, as concentration and continental heterogeneities develop during its decomposition). We should also emphasize here that in both cases, as was shown experimentally [*Gapeev and Gribov,* 2002b], the magnetic rigidity of the component directed along an external magnetizing field is always higher compared to the reverse-polarity TRM component. On the other hand, our earlier studies of CRM properties [*Gapeev and Gribov,* 1996, 1997, 1998, 1999, 2002a] also showed that, regardless of the titanomagnetite transformation, the acquired CRM is always magnetically more rigid than the corresponding pTRM. On the whole, these patterns suggest that the CRM self-reversal effect is even more pronounced during cooling of titanomagnetites, at the earlier stages of their multiphase oxidation, when the anomalous behavior of TRM (and pTRM) is best expressed [*Gapeev and Gribov,* 2002b]. Results of direct experimental verification of this suggestion are described in the present work.

## Results of Experiments

[6] Experiments were performed on 10- to 13-mg monolithic pieces of synthetic titanomagnetites ($Fe_{3-x}Ti_xO_4$, where $x = 0.2$, 0.4, and 0.6) placed in the central part of a 1-cm kaolin cube. Note that, precisely on duplicates of these samples, partial TRM self-reversal was fixed experimentally in products of multiphase oxidation of titanomagnetites of the given compositions [*Gapeev and Gribov,* 2002b].

[7] In the present study, initial samples were preliminarily demagnetized by an alternating magnetic field ($\widetilde{H} = 0.2$ T) along three mutually orthogonal directions, after which they

were subjected to multiphase oxidation in air in a thermomagnetometer at $500°C$ over exposure times ($t$) of 2 min to 240 h in a constant magnetic field. The CRM forming in these experiments was measured at the temperature of its acquisition in the thermomagnetometer shields with an uncompensated field being no more than 10 nT. After removal of the field, the samples were cooled in the same shields to room temperature ($T_0 = 20°C$). A self-reversal was determined from anomalous behavior in curves of cooling (at $H = 0$) and subsequent heating (also in the absence of the field) of laboratory CRMs.

[8] *Behavior of $J_{rc}(500°C, t)$.* Figures 1a, 1b, and 1c (curves 1) plot the time dependence of $J_{rc}$ at $500°C$ measured on titanomagnetite samples of the respective compositions $x = 0.2$, 0.4, and 0.6. All plots clearly display the following pattern in the behavior of $J_{rc}(500°C, t)$: an initial rise in the CRM to its maximum value, its subsequent decrease and, with the further exposure of the sample at the given temperature, a new rise followed by a decrease if the annealing time is sufficiently long. As was shown in [*Gapeev and Gribov,* 2002b; *Gribov,* 2004], such a behavior pattern of $J_{rc}(500°C, t)$ clearly reflects titanomagnetite alteration stages: the initial rise in CRM is associated with single-phase oxidation (commonly at cracks and edges of grains) of the initial titanomagnetite, and the subsequent changes in $J_{rc}(500°C, t)$ are related to the transformation of the spinel phase from a metastable (cation-deficient) into a two-phase state through nucleation and growth of hemoilmenite lamellae coherently associated with the titanomagnetite matrix.

[9] *Behavior of $J_{rc}(20°C, t)$.* In the given layout of experiment, a positive magnetization (i.e. directed along the external magnetizing field $H_0$) could have been acquired in the process of titanomagnetite annealing only by spinel phase regions that had a blocking temperature ($T_b$) above $500°C$. The fraction of such regions for low-Ti titanomagnetites is larger compared to high-Ti varieties, and the fraction of ferrimagnetic phases with $T_c < 500°$ C, on the contrary, smaller. Therefore, one might expect that, upon the transition from the $x = 0.2$ titanomagnetite to the $x = 0.6$ titanomagnetite, the effects of anomalous behavior of $J_{rc}(T)$ during cooling of a sample in the absence of a field will only be enhanced due to an increase in the relative amount of the negative (with respect to $\vec{H}_0$) component of $J_{rc}(T_0)$ produced by lower temperature ferrimagnetic regions of a grain. As is evident from Figure 1 (curves 2) and Figure 2 (curves 1), the given experiment completely confirmed this suggestion. Actually, samples of titanomagnetites with $x = 0.2$ and 0.4 that experienced multiphase oxidation in the field $H_0$ exhibit partial self-reversal of $J_{rc}(T, H = 0)$ during their subsequent cooling, while the $J_{rc}(T, H = 0)$ self-reversal is complete in samples of the $x = 0.6$ initial titanomagnetite (i.e. the values $J_{rc}(20°C)$ were always negative (Figure 1c, curve 2), although the anomalous behavior of $J_{rc}(T, H = 0)$ does not become more pronounced with an increase in $J_{rc}(500°C)$ (Figure 3)). It is natural to suppose that, during titanomagnetite oxidation at different temperatures, the relation between grain regions magnetized along the field and in the opposite direction will change, as will also be true of the relation between normally and reversely magnetized components of $J_{rc}(20°C)$.
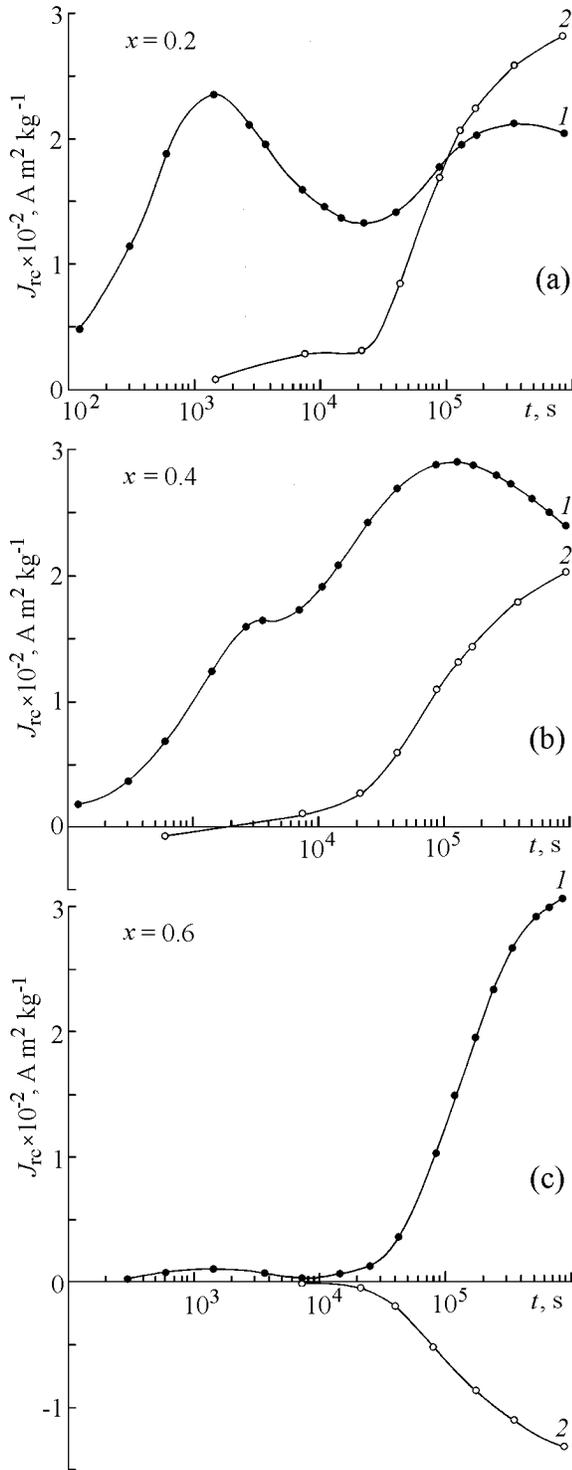
**Figure 1.** Time behavior of $J_{rc}(500°C)$ (1) and $J_{rc}(20°C)$ (2) for titanomagnetite samples of different initial compositions ($x$) subjected to isothermal annealing in air at 500°C in a field of 0.1 mT (the time is plotted on a logarithmic scale).
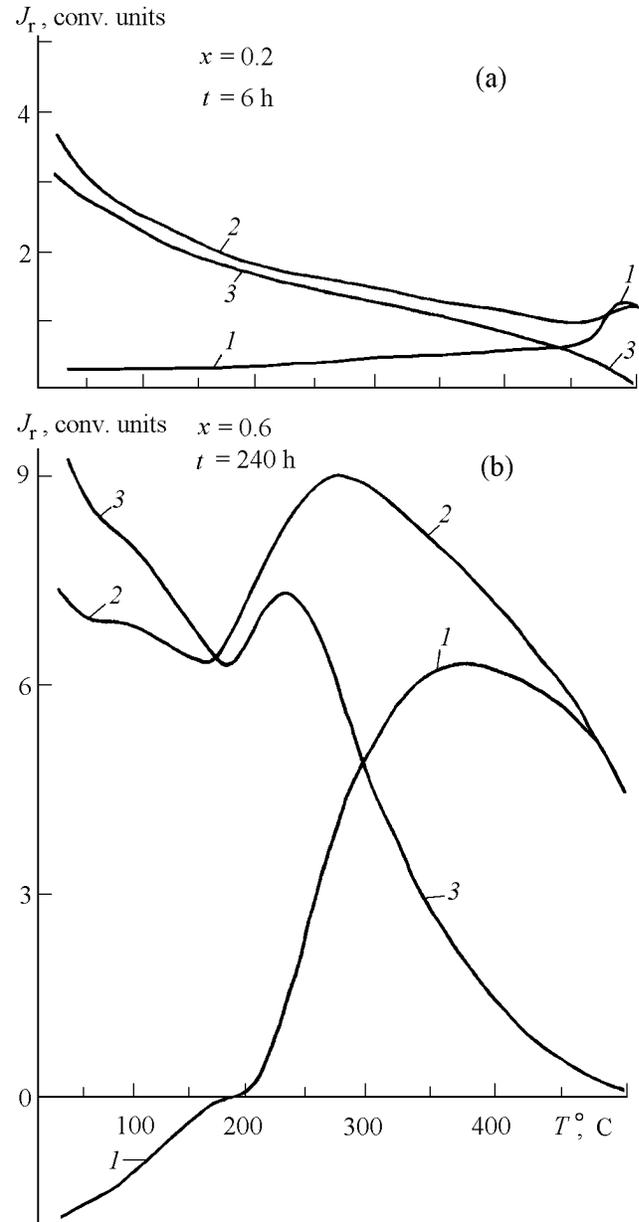


**Figure 2.** Thermal demagnetization of the components $J_{rc}$ (1), $J_{rc+rpt}$ (2), and $J_{rpt}$ (3) acquired in a field of 0.1 mT by titanomagnetite samples of different initial compositions ($x$) that underwent multiphase oxidation at 500°C during the time $t$: (a) $x = 0.2$, $t = 6$ h; (b) $x = 0.6$, $t = 240$ h.

[10] *Properties of $J_{rc}(20°C, t)$.* Figure 4 shows that distinctions are also observed in AF demagnetization curves of $J_{rc}(\widetilde{H}, 20°C)$ (created in multiphase-oxidized (at 500°C) titanomagnetite samples with different initial compositions cooled to room temperature in the absence of a field). Thus, in the case of low-Ti samples, the destruction of a viscous component during $\widetilde{H}$-demagnetization is followed by demagnetization of the negative low-temperature component $J_{rc}(20°C)$, bringing about a rise in $J_{rc}(\widetilde{H})$ observed in Fig-
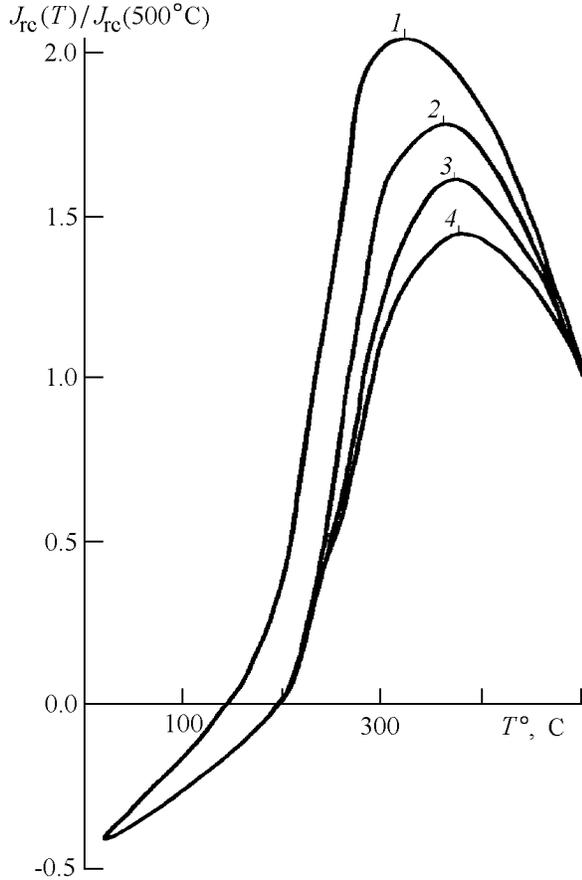
**Figure 3.** Thermal demagnetization of components $J_{rc}$ acquired by titanomagnetite samples (initial $x=0.6$) that underwent multiphase oxidation at $500°C$ in a field of 0.1 mT during time intervals of (1) 48, (2) 96, (3) 168, and (4) 240 h.

**Table 1.**

| Annealing time, h | $\widetilde{H}_m[J_{rc}(20°C)]$, mT | | | $\widetilde{H}_m[J_{rpt}(500°C, 20°C)]$, mT | | |
|---|---|---|---|---|---|---|
| 0.5 | – | – | – | 4.2 | 4.8 | – |
| 2 | 5.0 | 29.6 | – | 4.2 | 5.3 | 4.7 |
| 6 | 5.6 | 34.0 | 1 | 3.0 | 5.2 | 7.8 |
| 12 | 11.4 | 30.0 | – | 3.0 | 8.2 | 10.1 |
| 24 | 17.3 | 29.0 | 7 | 7.1 | 10.3 | 11.4 |
| 48 | 20.0 | 27.2 | – | 10.4 | 11.3 | 12.0 |
| 96 | 21.2 | 28.0 | 6 | 11.6 | 10.9 | 11.8 |
| 240 | 21.4 | 27.0 | 14 | 12.0 | 10.5 | 10.8 |
| Initial composition, $x$ | 0.2 | 0.4 | 0.6 | 0.2 | 0.4 | 0.6 |

ure 4a (curves 1–4). The further $\widetilde{H}$-demagnetization of the sample leads to the destruction of magnetically more rigid positive component and, as a result, to a drop in CRM. On the other hand, an increase in the time of oxidation of the given titanomagnetites leads to a gradual disappearance of ferrimagnetic regions with $T_c < 500°C$ in a grain. Accordingly, the amplitude of $J_{rc}(\widetilde{H}, 20°C)$ gradually decreases, and only a decrease in $J_{rc}(\widetilde{H}, 20°C)$ (curve 5 in Figure 4a) is observed at $t > 11$ h; therefore, this decrease can be related to the fact that the state with $T_b > 500°C$ is attained in all grain regions.

[11] In the case of high-Ti samples, with a predominant contribution to $J_{rc}(20°C)$ of the softer reversely magnetized low-temperature component, AF demagnetization of the latter gradually decreases its value, up to the transition into the region of negative values (Figure 4b).

[12] We should note that median fields $(\widetilde{H}_m)$ of AF destruction of $J_{rc}(20°C)$ in both examples considered above will reflect the rigidity of its different components. Thus, in the first case (low-Ti samples), the value $\widetilde{H}_m$ reflects the magnetic rigidity of the $J_{rc}(20°C)$ component magnetized along the field $H_0$, whereas in the second case (high-Ti sam-

ples) the corresponding component is directed in the opposite direction. Results of $\widetilde{H}_m$ measurements presented in the Table 1 show that the rigidity of the along-field component is two to three times higher than the rigidity of the reversely magnetized component of CRM. For comparison, the table also presents the values of median fields of AF destruction of pTRMs acquired in the field $H_0$ during cooling of multiphase-oxidized samples (of course, with the use of duplicates) from $500°C$ to $20°C$, i.e. in the temperature interval in which the reversely magnetized component $J_{rc}(20°C)$ forms. As expected, the resulting values of $\widetilde{H}_m$ for pTRM and the low-temperature CRM component are commensurate.

[13] Further experiments were intended to asses the role of CRM in the self-reversal of the resulting magnetization $J_{rc+rpt}$. For this purpose, samples of multiphase-oxidized titanomagnetites that acquired $J_{rc}(500°C, H_0)$ during annealing were cooled to room temperature in the magnetic field $H_0$. Results are presented in Figure 2 (curves 2). It is well seen that the superposition of the partial remanence $J_{rc}(500°C, 20°C; H_0)$ on $J_{rc}(500°C)$ make the heating curve $J_{rc+rpt}(T, H = 0)$ anomalous as well, but negative values of the total magnetization are not observed.

[14] Figure 5 presents transformations of the curves $J_{rc+rpt}(T)$ for multiphase-oxidized (at $500°C$ during 168 h in a field of 0.1 mT) titanomagnetite samples (initially with $x = 0.6$) under different conditions of their subsequent cooling also in a field of 0.1 mT. The anomalous behavior of $J_{rc+rpt}(T)$ is clearly seen to become more pronounced with a decrease in the cooling rate. It is natural to suppose that this is associated with an increase in the contribution of the magnetically soft viscous component of the resulting magnetization.

[15] To gain constraints on the relative contributions of magnetically rigid and soft components to the remanence $J_{rc+rpt}(T_0)$, we constructed the dependences $J_{rc+rpt}(\widetilde{H}, T_0)$ shown in Figure 6. The curves $J_{rc+rpt}(\widetilde{H})$ are seen to be multicomponent curves. With a decrease in the cooling rate of samples, the fraction of the magnetically soft component of $J_{rc+rpt}$ reflecting an increasing contribution just of its vis-
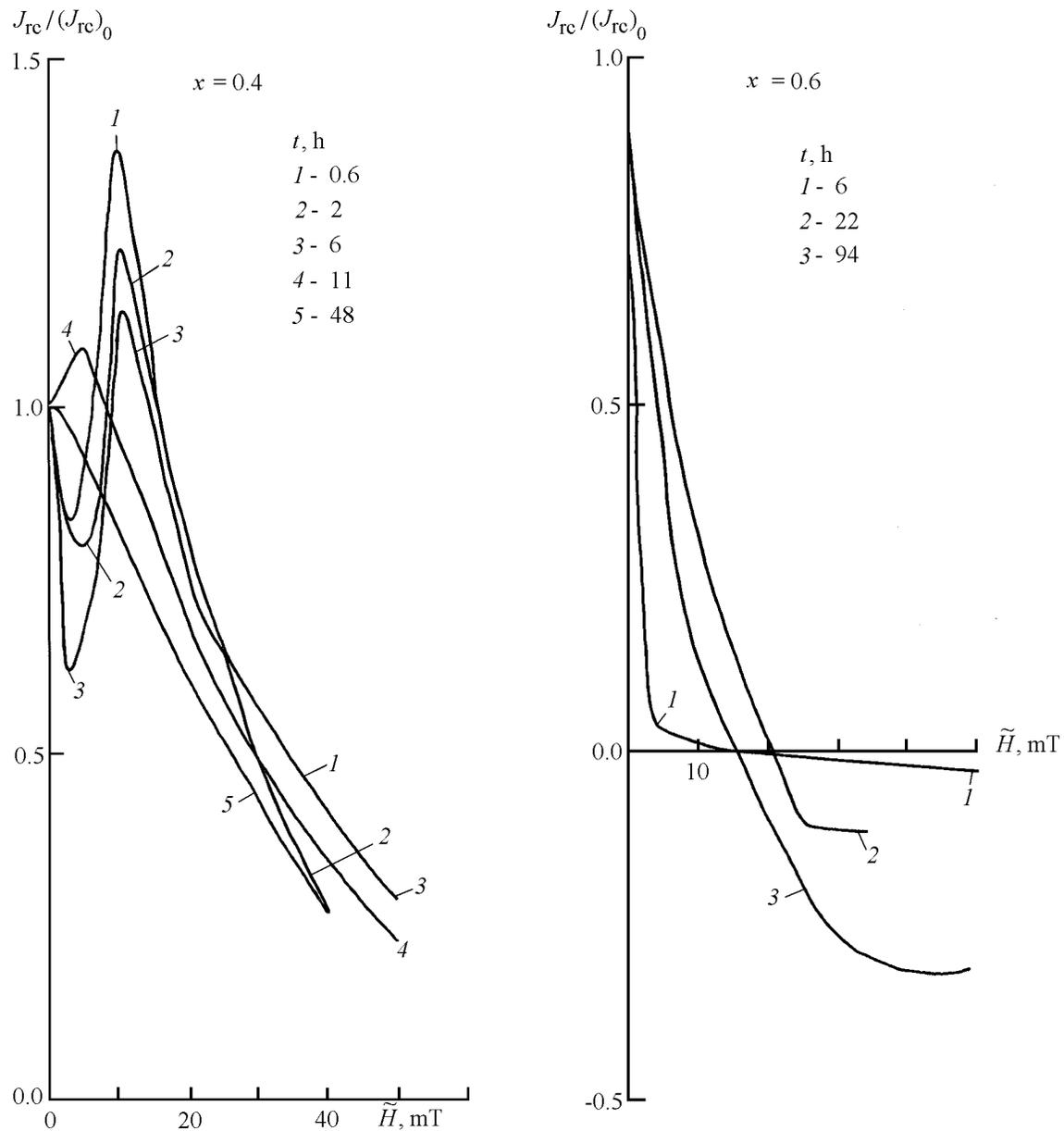
**Figure 4.** Demagnetization of the component $J_{rc}(20°\text{C})$ by an alternating magnetic field $(\widetilde{H})$ for titanomagnetite samples with different initial compositions $(x)$ and different times $(t)$ of their exposure in air at $500°\text{C}$ in a field of $0.1$ mT.

cous component increases. However, the measured integral value $\widetilde{H}_m$ is always evidence for the contribution of magnetically rigid regions in a grain.

## Discussion

[16] As noted above, partial self-reversal of TRM was already observed during multiphase oxidation of the given titanomagnetite samples [*Gapeev and Gribov,* 2002b]. Based on the evident dependence of this effect on the level of internal elastic stresses, the authors stated that, in this case, the cooling-related anomalous behavior of the thermal curve $J_{rt}(T)$ can be accounted for by magnetostatic interaction between sample volumes differing in the level of stresses accommodating elastic deformations of the crystal lattice in the junction plane of phases that form during oxidation, the rhombohedral phase segregated as a network of ilmenite lamellae and the spinel phase forming a system of cells whose composition continuously varies from almost pure magnetite in the direct neighborhood of a lamella to
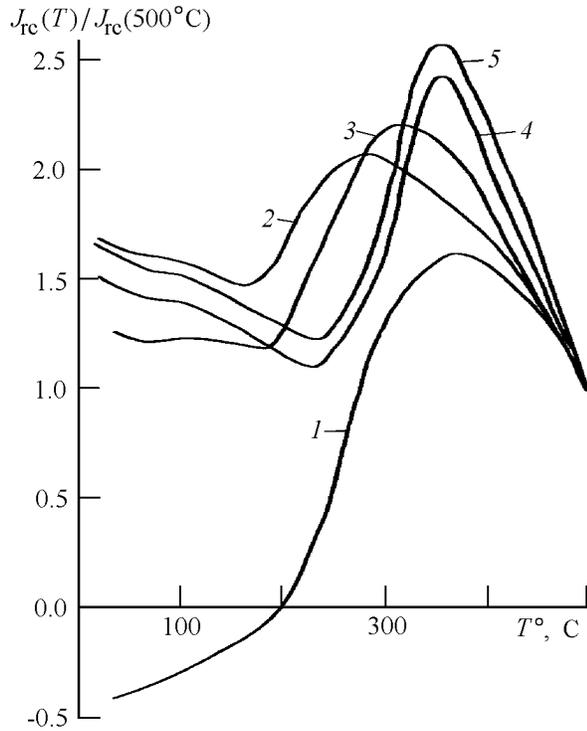
**Figure 5.** Temperature dependence of various types of remanence under different conditions of cooling of titanomagnetite samples (initial $x = 0.6$) that underwent multiphase oxidation at $500°C$ in a field of 0.1 mT during 168 h: (1) CRM, cooling rate of $1000°C/h$ at $H = 0$; (2–5) CRM+pTRM, cooling rates of (2) 1000, (3) 100, (4) 10, and (5) $1°C/h$ in a field of 0.1 mT.

**Figure 6.** Curves of AF ($\widetilde{H}$) demagnetization of various types of remanence created in titanomagnetite samples (initial $x = 0.6$) under different conditions of their cooling that underwent multiphase oxidation at $500°C$ in a field of 0.1 mT during 168 h: (2–4) CRM+pTRM, cooling rates of (2) 100, (3) 10, and (4) $1°C/h$; (1) pTRM created in sample 4 (after demagnetization of its components CRM+pTRM by the field $\widetilde{H}$) during its cooling from $500°C$ at a rate of $1000°C/h$ in a field of 0.1 mT.

the initial titanomagnetite far from it. A possible method of this phenomenon of TRM self-reversal is considered in detail in [*Shcherbakov et al.,* 1976] and is based on the well-known fact of diffuse ferri-paramagnet phase transition in structurally heterogeneous ferrimagnets [*Belov,* 1959]. According to [*Shcherbakov et al.,* 1976], we have a situation in which stressed (deformed) and unstressed volumes of ferrimagnetic material play a role of physically different phases (cf. [*Verhoogen,* 1959]) that have somewhat different Curie points. In this case, the cooling-related self-reversal of TRM can be effected due to magnetostatic interaction (magnetically les rigid unstressed regions of the ferrimagnet are magnetized in the demagnetizing field of higher-temperature stressed regions) and different temperature behavior of spontaneous magnetizations ($J_S$) of the given regions (we emphasize that, according to [*Shcherbakov et al.,* 1976], smaller $J_S(T_0)$ correspond to higher $T_C$, which is very favorable for the validity of self-reversal conditions).

[17] In the framework of the given model, the total TRM experiencing partial self-reversal during thermal demagnetization of the multiphase-oxidized titanomagnetite fraction of the samples under study [*Gapeev and Gribov,* 2002b] can be tentatively subdivided into three components: (a) a magnetically rigid or high-temperature positive component whose

carriers are spinel regions adjacent to lamellae, (b) negative component acquired under the action of component (a) and related to titanomagnetite volumes directly contacting with stressed zones, and (c) positive magnetically soft component whose carriers are unstressed titanomagnetite regions that are not involved in the self-reversal process.

[18] A gradual attenuation and disappearance of a self-reversal in the spinel phase recorded in laboratory experiments [*Gapeev and Gribov,* 2002b] is evidently related to a decrease in the volume of this phase during the subsequent oxidation of a sample. In this case, the volume of the rhombohedral phase increases and a self-reversal should be expected in it due to the interaction between stressed and unstressed volumes. This is a characteristic feature of the magnetostatic self-reversal of TRM of both, low and high temperature types [*Gapeev and Gribov,* 2002b] during multiphase oxidation of titanomagnetite samples in which a self-reversal is discovered in the present work in curves of cooling and subsequent thermal demagnetization $J_{rc}(T, H = 0)$.

[19] In principle, the aforediscussed causes of the TRM self-reversal can be similar to those of the CRM self-reversal. Actually, in the case of CRM formation, the Curie points of forming material move away from the chemical reaction temperature $T_r$ (on the geological time scales, even at relatively low temperatures of the medium), with the mag-

netic moments of the ferrimagnet being blocked along the direction of the external magnetizing field. In other words, with a fixed temperature of reaction, the Curie point of the newly formed material, rather than the cooling temperature (as in the case of the pTRM formation), is a variable value. This means that chemical magnetization accompanied by a rise in $T_C$ with fixed $T_r$ is actually similar to the thermal magnetization in the analogous temperature interval $T_C - T_r$ of stable ferrimagnets. On the other hand, as was expected, anomalous behavior of curves of cooling and heating $J_{rc}(T, H = 0)$ was fixed in our experiments just at the stage of titanomagnetite multiphase oxidation associated with development of significant elastic stresses in a sample (cf. [*Gapeev and Gribov,* 2002b]). In accordance with these facts, it seems therefore quite natural to apply the aforementioned considerations to the treatment of the CRM self-reversal with appropriate modifications: during chemical magnetization, the magnetization order in an oxidized structurally heterogeneous grain will be determined not by Curie points but by the succession of blocking temperatures of segregations (in the form of different phases or even separate pseudo-single-domain regions of a higher coercivity). Although in the present experiments we failed to obtain negative values of the resulting magnetization, it is important that the values of $J_{rc+rpt}$ measured at 20°C were invariably smaller than the corresponding values of the "pure" magnetization $J_{rpt}(500°C, 20°C)$ (cf. Figures 2b and 5). Evidently, the complete CRM self-reversal requires that the earlier forming resulting $J_{rc}$ must be less intense and less rigid than the magnetization forming later.

[20] The physical mechanism of the self-reversal of remanent magnetizations (both TRM and CRM) considered here and previously proposed for the explanation of a particular case of the TRM self-reversal in titanomagnetites at the stage of coarsening of titanomagnetite spinodal decomposition structures [*Melnikov and Khisina,* 1976] appears to be of a more general significance. This is also indicated by results derived from studies of the TRM and CRM self-reversals at the spinodal stage (the formation stage of a modulated structure) of the natural titanomagnetite decomposition process [*Gapeev and Gribov,* 2008a], (A. K. Gapeev and S. K. Gribov, in press, 2008b).

[21] In particular, as was established in [*Gapeev and Gribov,* 2008a], a stressed state can be preserved in a ferrimagnetic fraction representing finely disperse magnetite-ulvospinel segregations of the primary solid solution in ~250-Ma trap formations. Moreover, it is significant that samples of this doleritic collection generally had one or two antipodal components of the NRM vector. However, thermal treatment of the given natural samples revealed a partial self-reversal of TRM only in samples at the stage of semicoherent interfaces between segregations and, accordingly, at a relatively low level of stresses in the structure. In this case, laboratory-induced homogenization led to diffusional redistribution of cations and, as a result, generation of additional stresses in a nonequilibrium solid solution; i.e. conditions required for the realization of TRM self-reversal were created in the experiment. On the other hand, samples stabilized under in situ conditions at earlier decomposition stages (before the coherence loss in segregations) did not reveal the

$J_{rt}$ self-reversal effect because of rapid homogenization during thermal treatment of the samples. Therefore, we have every reason to suggest that the $J_n$ self-reversal property can be lost at some stages of the titanomagnetite evolution but the reversed remanent magnetization is preserved. As a result, an undiscovered neglected self-reversal will lead to incorrect paleomagnetic and geochronological correlations, inadequate geostructural interpretations, and so on.

[22] Thus, the comparison of results obtained in [*Gapeev and Gribov,* 2002b, 2008a; *Melnikov and Khisina,* 1976]; (A. K. Gapeev and S. K. Gribov, in press, 2008b) and in this study shows that the presence of concentration heterogeneities of the short-range layering type and the presence of significant inhomogeneous deformations of a crystal lattice are, on the one hand, features characteristic of products of titanomagnetite decomposition of both spinodal and oxidative types and, on the other hand, factors responsible for anomalous behavior of TRM and CRM.

[23] Results of this work imply that the NRM self-reversal effect is not a unique case but a general phenomenon caused by physicochemical processes post-crystallization transformations (decomposition and oxidation) of a primary titanomagnetite solid solution.

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