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Rare Earth Elements Geochemistry of Laowan Gold Deposit in Henan Province: Trace to Source of Ore-Forming Materials

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Abstract: The compositions of REE in quartz and pyrite from the main stage of the Laowan gold deposit in Henan Province and that in quartz from Laowan granite were determined by inductively coupled plasma-mass-spectrometry (ICP-MS) to trace the source of ore-forming materials. Meanwhile, the REE compositions of the deposit ore, granite and metamorphic wall rock were also considered for comparative studies in detail. The range of $\sum$REE of quartz and pyrite from the deposit ores is $4.18 \times 10^{-6} \sim 30.91 \times 10^{-6}$, the average of $\sum$REE is 13.39×10^{-6}, and the average of $\sum$REE of quartz in the Laowan granite is 6.68×10^{-6}. There is no distinct difference of REE parameters between the deposit ore quartz and granite quartz. The quartz in gold deposit has the same REE particular parameters as quartzes from Laowan granite, such as δ_{Eu}, δ_{Ce}, $(La/Yb)_N$ and $(La/Sm)_N$, partition degree of LREE to HREE, especially, the chondrite-normalized REE patterns, but no similarity to those from metamorphic wall rock, which shows that ore-forming hydrothermal fluid is mainly the fluid coming from the Laowan granite magma, rather than metamorphic fluid. Meanwhile, comparison studies on REE features between minerals from the deposit ores and related geological bodies in the deposit show that REE characteristics of minerals can serve as an indicator of ore-forming fluid properties and sources, while the REE characteristics of the bulk samples (such as deposit ores, granites and wall rocks) can not trace the source of the ore-forming materials exactly.

Key words: Laowan gold deposit; REE geochemistry; source of the ore-forming material; ore-forming fluid; rare earths

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ICP-MS is generally regarded as powerful element analysis technique with advantages of high sensitivity, low background, and its detection limitation is $1 \times 10^{-3} \sim 1 \times 10^{-2}$ ng·ml^{-1} for most elements at present. So it can be used to determine content of trace elements and super-trace elements[1]. The fluid inclusions in deposits have capability to trace the source of ore-forming fluid, however, the fluid inclusions are generally tiny and have low concentration of rare-earth elements within them, which restricts their application in deposit area. For a long time, the REE compositions of deposit ores and related geological bodies have been used to trace the source of ore-forming materials. But different kinds of minerals have different REE partition of distribution coefficients, deposit ores have complicated mineral compositions, and rare-earth elements of different source were mixed in bulk deposit ore samples, which makes it difficult to trace the

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source of ore-forming materials accuratery. ICP-MS technique makes it possible to study the low concentration of rare earth elements in minerals fluid inclusions. Now this technology has been widely used to trace source of ore-forming materials and rock-forming materials in economic geology and petrology[2~6]. In this paper, the REE compositions of quartz and pyrite from deposit ores and granite quartz in the Laowan gold deposit region were determined by ICP-MS to trace the source of ore-forming materials, particularly ore-forming fluid of Laowan gold deposit with divergence of genesis now[7~9].

1 Geological Characteristics of Deposit

The Laowan lies in Tongbai county of Henan Province, and tectonically in Tongbai structural zone, transition region of Qinling-Dabie complex orogen. The Laowan gold deposit is hosted in NWW Laowan ductility shear zone. The Laowan gold deposit lies in south of Songpa fault zone adjoining to Qinling formation and north of Laowan-Hongyihe fault, near the Laowan granite.

In Laowan gold deposit area, the exposed strata are Guishan Group, which underwent several times of strong tectonic movement, metamorphism and deformation. The strata mainly consists of mica-quartz schist and hornblende schist, and its schistosity trend parallels with regional tectonic spreading direction (NWW). The ore bodies occur in mica-quartz schist, and wall rocks of ore body top and bottom are hornblende schist. Bedded and similarly bedded ore bodies contain 90% ore reserve. Vein ore bodies extensively distribute in deposit area too, generally crossing bedded ore bodies and the strata. The wall rock alterations related to mineralization in the deposit are extensively developed and dominated by silicification, pyritization, sericitization-quartzitzation and chloritization. The sericitization-quartzitzation is most closely related to gold mineralization. There is no distinct boundary between ore bodies and wall rock, only gradual zone from wall rock to ore bodies. During fluid-wall rock interaction, feldspar and mica in wall rock are usually replaced by sericite with quartz. According to lithological characteristics of the wall rock, there are two ore types: mica-quartz schist gold ore and hornblende schist gold ore. Based on ores characteristics, minerals assemblages and minerals relations of the gold deposit, main mineralization periods can be classified into stage Ⅰ, stage Ⅱ, stage Ⅲ. Among these stages, stage Ⅱ is the main gold mineralization stage. The main gangue minerals are quartz, sericite, and main ore minerals are pyrite, galena and chalcopyrite minor in quantity in gold ore of Laowan gold deposit.

2 Samples Treatment and Analysis Technique

For this study, 10 samples were collected from Shangshanghe ore portion in Laowan deposit area. Be-

Table 1 REE compositions of minerals in Laowan gold deposit area(10^{-6})

Bodies	Laowan gold deposit					Laowan granite				
Sample No.	SH22	Ⅱ2	CP2	Wan102	SH21	Ⅱ1	D2	Wan101	1Q	3Q
Mineral	Pyrite	Pyrite	Pyrite	Pyrite	Quartz	Quartz	Quartz	Quartz	Quartz	Quartz
La	6.02	1.01	1.78	5.11	0.76	0.80	2.22	2.67	1.64	0.85
Ce	10.8	1.95	3.08	9.60	1.36	1.34	4.11	4.77	3.23	1.56
Pr	1.21	0.27	0.39	1.12	0.39	0.21	0.56	0.56	0.42	0.22
Nd	4.23	0.89	1.45	3.98	0.80	0.71	1.78	2.06	1.52	0.63
Sm	0.87	0.18	0.25	0.73	0.10	0.12	0.33	0.37	0.22	0.12
Eu	0.12	0.03	0.05	0.17	0.02	0.03	0.06	0.05	0.06	0.02
Gd	0.46	0.14	0.23	0.076	0.08	0.10	0.25	0.30	0.22	0.08
Tb	0.10	0.02	0.03	0.17	0.05	0.01	0.07	0.06	0.03	0.01
Dy	0.33	0.15	0.15	1.12	0.09	0.08	0.22	0.30	0.13	0.09
Ho	0.08	0.04	0.05	0.29	0.03	0.02	0.05	0.07	0.02	0.01
Er	0.18	0.09	0.10	0.91	0.06	0.05	0.12	0.18	0.03	0.05
Tm	0.05	0.03	0.02	0.17	0.03	0.01	0.01	0.02	0.02	0.02
Yb	0.19	0.10	0.09	1.01	0.05	0.02	0.10	0.16	0.07	0.06
Lu	0.02	0.04	0.01	0.19	0.01	0.04	0.03	0.02	0.03	0.03
Y	0.58	0.56	0.25	5.58	4.89	0.64	2.03	1.03	1.01	0.94
ΣREE	25.24	5.5	7.93	30.91	8.72	4.18	11.94	12.62	8.66	4.69
ΣLREE	23.25	4.33	7.00	20.71	3.43	3.21	9.06	10.48	7.09	3.4
ΣHREE	1.99	1.17	0.93	10.2	5.29	0.97	2.88	1.11	1.57	1.29
ΣLREE/ΣHREE	11.68	3.70	7.53	2.03	0.65	3.31	3.15	9.44	4.52	2.64
δ_{Eu}	0.53	0.56	0.63	0.70	0.67	0.82	0.62	0.45	0.83	0.59
δ_{Ce}	0.90	0.86	0.83	0.90	0.57	0.75	0.84	0.87	0.89	0.83
$(La/Yb)_N$	20.87	6.65	13.03	3.33	10.01	26.35	14.62	10.99	15.43	9.33
$(La/Sm)_N$	4.21	3.41	4.33	4.26	4.62	4.06	4.09	4.39	4.54	4.31
$(Gd/Yb)_N$	1.94	1.12	2.05	0.6	1.28	4.00	2.00	1.50	2.52	1.07

fore minerals were selected, ores samples were observed under optical microscope in order to confirm that quartz selected has the same metallogenic main-stage with pyrite, and fresh granite samples selected were also observed under optical microscope so as to ensure that the quartz selected has no transformation in late stage of magma evolution. Because quartz in metamorphic wall rock was too tiny and difficult to select, REE composition of quartz in metamorphic wall rocks were not measured in this experiment. In selecting minerals process, first, quartz and pyrite were sorted by heavy concentration, then selected by hand under the pairs of tube microscope. Pyrite selected was washed with distilled water three times, finally dried. The quartz was soaked for 30 min in 10% of the HCl, then washed with the distilled water several times, and finally dried for study. The REE compositions of all samples were analyzed at center of geological experiment of China, Chinese Academy of Geological Science. The results of REE analyses are listed in Table 1 and shown in Fig. 1.

3 Results and Discussion

3.1 REE geochemistry characteristics of ore-forming fluid and magmatic fluid

Based on studies on REE geochemistry of quartz in Sidaogou hydrothermal gold deposit of Liaoning ,

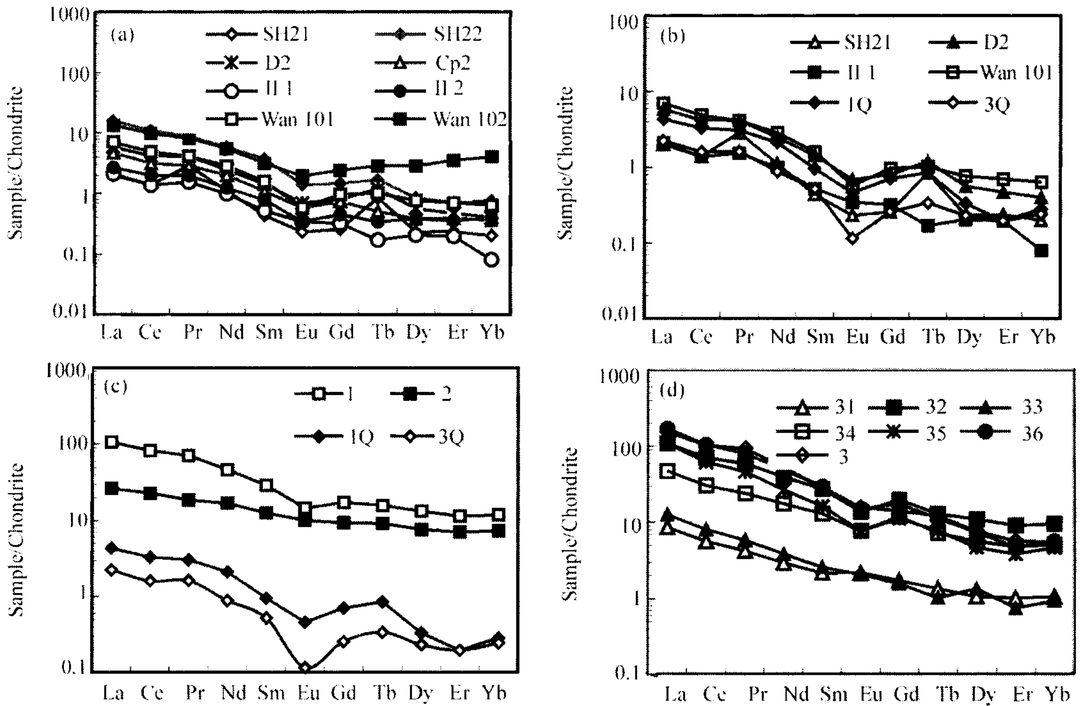

Fig. 1 Chondrite-normalized REE patterns of samples in Laowan gold deposit area
(a) Quartz and pyrite in ores; (b) Quartz from ores and granite; (c) Laowan granite quartz(1Q, 3Q) and bulk metamorphic wall rock (1, 2); (d) Two-mica quartz schist type ore (32, 34), plagioclase hornblende schist type ore (31, 33) and bulk granite(3, 35, 36); Sample No. in Fig. 1 are corresponding to those in Table 1, other data from Ref. [11]

Table 2 REE particular parameters in Laowan gold deposit area*

Bodies	Samples	$\sum$REE/10^{-6}	LREE/HREE	δ_{Eu}	δ_{Ce}	$(La/Yb)_N$	$(La/Sm)_N$
Deposit	Two-mica quartz schist type ore	142.14	3.45	0.61	0.85	10.49	6.31
	Plagioclase hornblende type ore	14.4	3.89	1.07	0.88	11.39	6.9
	Pyrite	17.40	6.24	0.61	0.87	10.97	4.05
	Quartz	9.37	4.14	0.64	0.76	15.49	4.29
Granite	Quartz	6.68	3.58	0.71	0.86	12.38	4.45
	Whole rock	212.13	17.411	0.64	0.82	5.9	9.65
Wall-rock	Two-mica quartz schist	219.96	3.69	0.63	0.93	9.18	6.06
	Plagioclase hornblende schist	78.9	1.8	0.92	1.01	3.65	3.45

* REE concentration of chondrite from Ref. [10]; all are average values in the Table; REE data of deposit ore, granite and wall rocks from Ref. [11]

Fan et al[12] suggested that rare earth elements in quartz mainly exist in fluid inclusions. So REE characteristics of quartz reflect that of ore-forming fluid in equilibrium with quartz. Then REE compositions of pyrite coexist with quartz also show REE characteristic of ore-forming fluid. Therefore REE characteristics of quartz and pyrite formed during main mineralization stage in Laowan gold deposit are representative of mineralization hydrothermal fluid REE compositions.

From Table 1, it can be seen that the total amounts of REE in quartz from Laowan gold deposit are in the range of $4.18 \times 10^{-6} \sim 12.62 \times 10^{-6}$, LREE/HREE ratios are $0.65 \sim 9.44$ and $(La/Yb)_N$ ratios are $10.0 \sim 16.35$. The partition of LREE to HEEE is high. $(La/Sm)_N$ ratios are $3.41 \sim 4.62$, which shows the weak partition within the light rare earth. There are distinct negative anomalous Eu and Ce, and the average of δ_{Eu} and δ_{Ce} are 0.64 and 0.76, respectively. The total amounts of REE in pyrite are higher than those in quartz from the same sample, so is the average value of total amounts in pyrite that may be resulted from different presence modes of REE in minerals. The REE in quartz mainly exists in quart fluid inclusions, while REE in pyrites exists in not only fluid inclusions but also crystal lattice of pyrite, where REE substitutes for Fe element in the form of isomorphism. Though there are differences of rare earth elements concentration between quartz and pyrite, the partition degree of LREE to HREE and anomaly of Eu and Ce of pyrite are similar to those of quartz. The chondrite-normalized patterns of REE in pyrite and quartz are also extremely similar, i.e. distinct right-tipping curves which can be seen in Fig. 1(a) (except for sample No. Wan102, because sericite adheres to pyrite on the surface). Obviously, there is no remarkable difference of REE curves between pyrite and quartz, which indicates that REE chondrite-normalized patterns of pyrite and quartz can act as representatives of mineralization hydrothermal fluid in Laowan gold deposit. The REE patterns of the minerals are not affected by differentia of mineral kinds and REE abundance.

The total amounts of rare-earth elements of two quartz samples from Laowan granite are 4.69×10^{-6} and 8.66×10^{-6}, LREE/HREE ratios are 2.64 and 4.52, $(La/Yb)_N$ ratios are 9.33 and 15.43, partition between LREE and HREE is also apparent. The REE chondrite-normalized patterns of quartz in Laowan granite are obviously tip to right (Fig.1(b)). The values of $(La/Sm)_N$ are 4.54 and 4.31, and the values of δ_{Eu} are 0.59 and 0.83, similar to those of quartz in gold deposit. The REE characteristics of the minerals may show those of the magmatic hydrothermal fluid in equilibrium with quartz during Laowan granite magma fractional crystallization.

3.2 Source of ore-forming materials

As discussed above, the REE features of quartz and pyrite in deposit ores may reflect the REE characteristics of ore-forming fluid in equilibrium with minerals (such as quartz and pyrite) during mineralization, and the REE compositions of quartz in granite also represent the REE compositions of the hydrothermal fluid in equilibrium with the granite magma. In Table 2, the REE feature parameters of quartz of Laowan granite are similar to those of quartz in gold deposit. In Fig.1(b) there is no obvious differentiation of REE chondrite-normalized patterns between granite quartz and gold ore quartz, which indicates that magmatic hydrothermal fluid from Laowan granite has much accordance with ore-forming fluid of Laowan gold deposit. In Fig.2(a), the plots of parameters values of quartz in granite and in gold deposit are close. The variation range of mineral plots of deposit minerals exceeds that of granite, but the values of YbN are similar in Fig.2(b), which indicates that there is close genetic relationship between magmatic fluid and gold mineralization hydrothermal fluid. It can be inferred that the latter mainly comes from the former . Like -

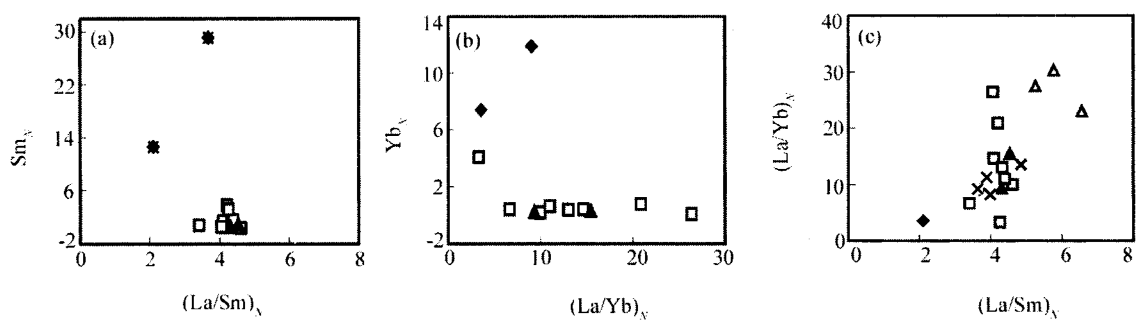

Fig.2 REE correlation figures in Laowan gold deposit
□ Quartz and pyrite in gold ore; ▲ Quartz in granite; × Bulk deposit ore; ● Bulk wall rock;
△ Bulk granite; Data of deposit ore, granite and wall rock from Ref.[11]

wise, in the Fig.2(c), the data plots of minerals from granite and deposit concentrates are still in the same range, which further prove that magma fluid dominates in ore-forming fluid. The research result is consistent with characteristics of hydrogen, oxygen and helium isotopes in Laowan deposit[9].

Though the Laowan gold deposit distribution is strictly controlled by the Laowan granite spreading, the gold deposit occurs in the metamorphic wall rock, then the metamorphic water in equilibrium with wall rocks may have certain contribution to the gold mineralization. In order to study the contribution of the metamorphic water to ore-forming fluid, REE characteristics of bulk wall rock have been served as REE characteristics of metamorphic minerals in wall rock because of lack of REE composition data of metamorphic minerals in wall rocks. Now research has proved that the total REE amounts increase during shearing and deformation of the shear zone, but REE chondrite-normalized patterns of wall rock can also reflect the characteristics of primitive rock[13]. Therefore, REE Chondrite-nomalized patterns of the metamorphic fluid in equilibrium with metamorphic rock should be similar to those of the metamorphic wall rock. In Table 2, the total REE amounts of the (two kinds) metamorphic wall rock are by far higher than those of minerals in deposit and granite, REE chondrite-normalized curves and partition degree between LREE and HREE are all different from those of the minerals from deposit and granite (shown in Table1 and Fig. 1 (a, b, c)). These suggest that the backbone of the ore-forming fluid may not be metamorphic fluid, which is in accord with the research results of metamorphic progress, metamorphic era of wall rock and metallogenetic era of Laowan gold deposit[14]. In Fig.2(a, b), the plots of REE parameters of wall rock far exceed the range of minerals from deposit and granite, which proposes that metamorphic water is not main component in ore-forming fluid. From the Fig.2(c), it can be seen that the plots of REE parameters of the deposit and granite minerals are different from those of the metamorphic wall rock, as well as those of the bulk granite samples. These demonstrate that the wall rock has minor contribution to ore-forming fluid. However, the granite data plots are not in good agreement with bulk ore and ore minerals' in the Fig.2(c). Meanwhile, bulk deposit ore has not the absolute same REE patterns as bulk granite in the Fig.1(b, d) though having similar mineral REE curves. It can be obtained that the REE feature of bulk granite and bulk deposit ore may not be an sensitive indicator of ore-forming fluid source.

At the same time, quartz and mica in metamorphic wall rock are tiny in grain size under microscope, and the inclusions among them are small and low in content. It may be a subordinate role for metamorphic hydrothermal fluid in ore-forming fluid. However, metallogenetic elements may be enriched during metamorphism and deformation of the wall rock, then metallogenetic elements are released during fluid-wall rock interaction and join the mineralization process.

4 Conclusion

The Laowan gold deposit is a large-scale gold deposit in northern of Tongbai mountain, even in north Huaiyang tectonic region. It is particularly crucial to reveal genesis of the deposit for mineral resources investigation and exploration in this region. Based on studies of REE geochemical characteristics of minerals in main mineralization stage of Laowan gold deposit and Laowan granite, it is found that the Chondrite-normalized REE patterns of quartz and pyrite in the gold deposit are similar to those of quartz in granite. Their REE geochemistry parameters are much similar to each other, while the REE Chondrite-normalized patterns of the minerals distinguishes from those of the wall rock. Therefore it can be concluded that the majority of ore-forming hydrothermal fluid should be of magmatic fluid origin from Laowan granite magma, hence metamorphic fluid may be neglected. Compared REE characteristics of ores and granite with minerals from deposit and granite, it is proposed that minerals REE geochemistry is a more effective method to trace source of ore-forming fluid than bulk samples REE compositions of ores and related geological bodies.

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Grain Growth Behavior of 5Cr21Mn9Ni4N Steel Micro-Alloyed by Rare Earth

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Abstract: Grain growth behavior of 5Cr21Mn9Ni4NRE steel was experimentally studied at various solid solution treatment temperatures and holding for different times. The experimental results show that the 5Cr21Mn9Ni4NRE steel has the feature of sharp austenite grain coarsening after solid solution treatment at the temperature above 1150 ℃. RE added in the steel has the benefit to restrain grain growth and increase grain growth activation energy. Grain growth mechanism is self-diffusion controlling process. The dispersed carbo-nitride in the steel greatly restrains grain growth. Solid solution treatment at the temperature below 1100 ℃ leads to little increase of grain size and little decrease of Rockwell hardness for mass carbo-nitride undissolved, whereas solid solution treatment at the temperature above 1150 ℃ leads to rapid grain coarsening and Rockwell hardness decrease. Grain growth exponent increases with the increasing solid solution treatment temperature.

Key words: solid solution treatment; Rockwell hardness; grain growth; grain growth exponent; rare earths

(See J. Chin. RE. Soc. (in Chin.), 2006, 24(1): 76 for full text)

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Microstructures and Phase Transition Points of As-Cast Mg-Zn-Y System Alloys

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Abstract: As-cast microstructures and their distribution of Mg-Zn-Y ternary alloy with high magnesium, low zinc and yttrium were examined using Nikon Epiphot optical microscopy (OM), RigakuD/max-3C X-ray diffraction (XRD), and JEOL JSM-6700F scanning electron microscopy (SEM) equipped with an energy-dispersive X-ray spectroscopy (EDS). In the as-cast microstructures, Yttrium and zinc tend to segregate at grain boundaries, therefore meets the compositional condition to form Mg-Zn-Y compounds at grain boundaries even with relatively low content of Y. The principal ternary phase in Mg-Zn-Y alloys can be consisted of W-phase ($Mg_3Y_2Zn_3$), Z-phase (Mg_3YZn_6) and X-phase ($Mg_{12}YZn$). Combining differential thermal analyzer (DTA) the phase transformation temperature of alloys were discussed. The eutectic melting temperature of W-phase at 520 ℃, the phase transformation temperature of Z-phase at 445 ℃ and the melting temperature of X-phase at 540 ℃ are shown.

Key words: as-cast microstructure; Mg-Zn-Y alloy; differential thermal analyzer; phase transition point; rare earths

(See J. Chin. RE. Soc. (in Chin.), 2006, 24(1): 86 for full text)