水－岩相互作用和成矿物质来源

梁祥济 著

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WATER-ROCK INTERACTION AND THE SOURCE OF METALLOGENIC MATERIAL

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This book is dedicated to the

30th International Geological Congress

梁祥济

Liang Xiangji

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内容简介

本书是“八·五”期间水—岩相互作用实验的最新成果，也是作者20多年来从事实验地球化学、矿物和岩石研究的系统总结。实验是在柿竹园、德兴、大冶、石碌、黄岗、红砬子和小汤山等全国17个典型的钨、锡、钼、铋、铁、铜、铅、锌、金、银矿区和泉区野外地质工作基础上进行的。通过这些矿区不同类型的围岩与含钾、钠卤化物为主的水溶液，在不同的温度和压力、不同成分的溶液及其pH值和不同氧化—还原环境等条件下，经历了120～576小时持续相互作用的实验(5000多次)，结果表明这些矿区(泉区)的成矿和成泉物质来源于各自的围岩——侵入岩、火山岩、变质岩、混合岩和沉积岩。

本书地质基础扎实，实验技术先进，内容丰富，资料翔实，数据可靠，观点新颖，论述深入，文图并茂。它打破了一切金属成矿元素来自深部岩浆的旧框框，提出了成矿物质来自围岩的新认识，揭示了这些成矿和成泉元素活化、迁移和沉淀的机理，从而为建立成矿新理论、开拓油田、开发热田和人造温泉提供了实验和理论的依据，开辟了新的途径。

本书可供地球化学、矿床学、水文地质学、石油地质学和物理化学等专业的科研、实验、生产人员以及有关院校师生参考。

WATER—ROCK INTERACTION AND THE SOURCE OF METALLOGENIC MATERIAL

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ABSTRACT

3/4 water and 1/4 rock coexist on the earth's surface. The surface water get into underground to mantle through break, fault and crevice. And the water of forming fluid in mantle come back to the surface after undergoing crustal movement, volcanic eruption, magmatism and metamorphism. And then the water get into atmosphere. The water in the atmosphere are condensed into cloud, which changes into rain and then drops on the surface of the earth. In this large cyclic process, the interaction of the water with encountering rocks occurs endlessly, profitable elements and harmful materials are activated and extracted from the rocks. It provides the requsite condition for the existence of mankind. But it also bears coming unfortunate natural calamity to mankind. Therefore, water-rock interaction is the most important subject under discussion of the modern earth science. It also has a direct bearing on the mineralization. The experiments with modern advanced techniques and methods are the most important way on studying the relation between water-rock interaction with ore-forming. Consequently, combined 17 typical mines (spring area) of China, author performed the experiments of water-rock interaction for studying the ore-forming mechanism of the activation, migration and precipitation of ore-forming elements in rock. The results of the experiments no doubt have important theoretical significances and applied values.

On the basis of field geological works of 17 typical mines for Fe, W, Sn, Mo, Bi, Cu, Pb, Zn, Au, Ag and spring area in Honge-Panzhihua of Sichuan, Makeng of Fujian, Daye of Hubei, Jianshan of Guangdong, Chihai and Yamansu of Xinjiang, Huanggang of Inner Mongolia, Shliu of Hainan, Mengjiagang, Tianbaoshan and Ximashan of Heilongjang, Tongboshan of Henan, Ningwu, Shizhuyuqn of Hunan, Dexing of Jiangxi, Honglazi of Liaoning and Xiaotangshan of Beijing etc. in China, author has engaged in the experiments of water-rock interaction in 5000 times according to the geological bases of experiment. The results of the experiment proved the ore-forming materials of these matallic

mineral deposits, specially hydrothermal-metasomatic mineral deposit and hot, mineral springs principally stemed from their wallrocks. Simultaneously, author studied the forms and conditions of the activation, migration and precipitation for their ore-forming elements and brought to light their ore-forming mechanism.

Geological base of experiment

Author selected above-mentioned 17 typical, researching high level, and representative good mineral deposits as studying subjects of water-rock interaction. The deposits have following common geological base of experiment:

(1) All telluric tectonic places of 17 ore (spring) areas are located in fold systems. In the fold systems, structures movemented strongly, magma intrude and effused frequently, some breaks and folds were appeared. The mineral deposits in these ore area practically are located in break zone, fold zone and contact zone or near those places. And these zones are the passageway of ore-forming and spring-forming material migration and the place of accommodation ore (see Fig. 2-1 Geological and structural map of the distribution for experimental samples).

(2) The average groundback values of ore-forming elements in ore body wallrocks of 16 mineral deposits of them are over a few-several thousand times than the crustal Clark values. For example of Procombrian System metamorphic rocks in Shizhuyuan mine, W is over 21.5 times than the crustal Clark value, Sn is over 1.32 times than that, Mo is over 19.2 times than that and Bi is over 5177.8 times than that. The ore-forming elements in the near ore wallrocks of some ore body loss in large area. For instance of Dexing porphyry copper deposit, Cu, Mo contents in the layers of the ore body outside distributed in ring-like negative (low) values round the ore bodies.

(3) Some wallrocks near ore bodies of the mineral deposits appear alteration and discoloration. For example of Makeng diabase, its colour universally changes from lightgreen-green to greyish white-light green; The potash feldspathization in the wallrocks of Dexing porphyry copper deposit is very strong; The wallrocks of Honglazi gold deposit universally arise the pyritization, silicification and carbonatization; The wallrocks (andesite) of Haunggang iron deposit generally are subjected to skarnation. Obviously, these are the marks of material metasomatism in water-rock interaction.

(4) The data of measuring temperature, measuring pressure, composition, saltiness and pH value for mineral inclusions in these mines are more complete. For instance, in the mineral inclusions of Honglazi mine, $Na^{+}=0.330$ ug/g; $K^{+}=1.230$ ug/g; $Mg^{2+}=0.330$ ug/g; $F^{-}=0.510$ ug/g; $Cl^{-}=0.118$ ug/g; $SO_4^{2-}=0.923$ ug/g. Spring area has rich materials of hydrogeology. For example of Xiaotangshan hot spring, its measuring data on ground water quality, water temperature, hydrochemical type, acid-alkalinity and hydrothermal reservoir are all in readiness comparably.

(5) Some mineral deposits are related to volcanic pneumato-hydrothermal solutions and volcanic rocks closely. As the wallrocks of these deposits-volcanic and subvolcanic rocks, which distributes within a large region, they are alterating strongly and with groundback values high of ore-forming elements. The ore-forming solution is the acid solution, rich in volcanic gas-hydrothermal compositions. For example, the Huanggang, Yamansu, Chihai, Ningwu and Honglazi mines have these characteristics. In volcanic gas, $H_2O=43.2\sim97.7$, $Cl=0.1\sim0.4$, $F=0.0\sim3.3$, secondary composition are H_2, S, P etc. (capacity %). NaCl is 7.2%, KCl is 6.0% in over 70% volcanic liquid phase, $pH=1.77\sim2.92$ in volcanic boiling water. And $pH=0.5\sim3.0$ in volcanic hot spring.

(6) The studying results of sulfur isotope ($\delta^{34}S$), lead isotope (208pb/204pb, 207pb/204pb, 206pb/204pb) and oxygen isotope ($\delta^{18}O$) in these mines show that sulfur mainly originated from the wallrocks, lead sourced from upper crust, $\delta^{18}O$ values $\leqslant 10‰$. As Shizhuyuan mine, $\delta^{34}S=-10.93\sim9.92‰$, it explained that sulfur participated from wallrocks. All 208pb/204pb, 207pb/204pb and 206pb/204pb of 17 galena lead isotopes have fallen in the region of upper crust. $\delta^{18}O=-8.9\sim10.7‰$ in fluid.

Techniques and methods of the experimentation

All experiments were performed in the Bridgman sealed pressure vessels with gold liners and pure titanium pressure vessels of acid-resisting and alkali-resiting ($Tc-_9$). The volumes of these pressure vessels are $13.40\sim14.00$ ml. Experimental samples were collected from the no alteration and no weathering wallrocks of the ore bodies and the hot spring above-mentioned 17 mines (spring area) -intrusive rock, volcanic rock, metamorphic rock, migmatite and sedimentary rock. For guaranteeing the sample being typicality and reflecting geological reality abjectively, author collected a fresh rock fragment in $2\sim3$ cm size of an interval of $5\sim10$ metres between each two fragments along stratigraphic tendency beyond $2\sim5$ km usually when collecting samples of metamorphic rock, sedimentary rock, volcanic rock and migmatite, and then big samples of $20\sim25$ kg were composed. Using the same method, author collected a fresh rock fragment in $2\sim3$ cm size through the phase zones of centre, interim and edge to the samples of intrusive rock and subvolcanic rock, and then big samples of $15\sim20$ kg were formed. After that, these samples were smacked for $1\sim3$ mm size separatly and then mixed thoroughly, and use the part as experimental samples. The others continuity were smacked and abraded through 200 sifting screen, mixed thoroughly again, and then sent to analysis for petrological chemistry, ore-forming elements and trace elements etc.

According to the compositions of volatile components, mineral inclusions and volcanic gas-hydrothermal solutions and the characteristics of the wallrock alteration above-mentioned 17 ore (spring) areas, the reactive solutions were compounds of chemical reagents and taking the aqueous solution of Na, K chloride (fluoride) as dominant factor. Their pH

=1.0~11.0. Reactive solution of the spring area in them is quadratic distilled water.

The temperatures for the experiment were 50~650℃, which were controlled by the automatic precise controls (JWK-703) and were measured by standard Le Chatelier thermocouples (LB-3). The instrumental error is ±2℃. The pressures of the experiment were $100\times10^5\sim1400\times10^5$ Pa, which were computed in the light of the P-V-T function of Kennedy G. C. or were demonstrated directly on pressure gauges. The durations of experiment were 120~576 hours. The oxidation reduction environments of the experiment were controlled by the mineral pairs of buffer. The solid and liquid products of the experiment were analysed by water chemistry, ore-forming element chemistry and petrological chemistry, and were studied under the microscope and scanning electron microscope as well as by X-ray.

Experimental results and their significance on the geology

1. The characteristics of solid remain samples after the experiment

Usualy in the solid remain samples after the experiment, they changed into lighter color, the qualities of the material were porous. the textures and structures were destroyed to different extent. On the surfaces of the grain of some solid remain samples appeared a lot of small holes by observing under scanning electron microscope. Some can catch sight of altered minerals. For example, the dolomites and the andesines in the granite from Xiaotangshan spring area, their surfaces appeared various laminated marks, small pits of face type, crevice and solution caves. At the same time, the clay minerals of kaolinite, montmorillonite and zeolite etc. were precipitated and formed on the other. Just as well, the minerals of diopside and tremolite etc. also appeared on the diabase surface of Makeng.

The specific gravity of solid remain samples after the experiment dropped. Their hardnesses were lowered. The weightlessness in 200 mg original sample were generally 0.10~20.50 mg. The results of petrological chemical analysis showed that rock weightlessness were led to activating metal ore-forming elements from it to lessen their content. At the same time, the increases of Na_2O and K_2O contents were appeared universally. These phenomenon tallied with field deposit geology, colour fading of wallrock alteration and characteristics of patash feldspathization and albitization.

2. The migrational modes of ore-forming elements

The ore-forming elements of Fe, W, Sn, Mo, Bi, Cu, Pb, Zn, Au and Ag extracted from different rocks by simulating compound aqueous solutions of distinct natures were mainly appeared in chlorine (fluorine) complex, chlorine (flucrine) -oxygen complex, chlorine (fluorine) -hydroxyl complex, hydroxyl complex and sodium sulphate. Secondly, they were existed in complex anionic group, halogeride, sulphide and simple substance ion. And the complexes, complex anionic groups etc. of the metal ore-forming elements stably migrated under the suitable conditions of temperature, pressure, acid-alkalinity of the solution and oxidation reduction.

The spring-forming elements in distilled water extracted from dolomite and granite were existed in simple substance negative and positive ions as well as acid radicle principally. Secondly, they were appeared in oxide mode. In the same way, the negative and positive ions, acid radicles and oxides stably activated in fixed region of temperature, pressure and favourable environment. The aqueous solutions with ore-forming and spring-forming elements after the experiment of water-rock interaction were mainly the true colorless and transparent solution. There are a small number of turbid solution. The colloid solution was very few, and the colloid core was SiO_2.

3. Thermodynamic properties for ore-forming element

Author combined the data and results of the experiment, performed in the study of thermodynamic properties of active and migrative equilibrium reaction for ore-forming elements especially Fe and Au, Ag. For example of Fe, according to the equilibrium reaction formula of experimental products in Shilu and Mengjiagang mines, $\log f_{O_2}$-T-P and Eh-pH-T diagrams of the formations of metasomatic rocks and iron deposits were drawn by the computing of thermodynamic parameters. The diagrams showed the different cause of formation Shilu deposit took hematite as the dominant factor and Mengjiagang deposit took magnetite as the dominant factor mainly for the reason of the former f_{O_2} and Eh high than the latter. For instance of Au and Ag, on the geological characteritics of Honglazi gold deposit, wallrock alteration and six equilibrium reaction formulas of experiment, their thermodynamic properties in reactive process were studied systematically. The diagrams of T-f_{O_2}-pH, Eh-pH-T-f_{H_2O}, logK-T and $\log f_{S_2}$-T for their ore-forming were drawn. It brought to light the chemical-physical conditions and the inter relation for Au, Ag activation, migration and precipitation.

4. Experimental results and their significances

The experimental results show:

(1) Under the pressures of $500\times10^5\sim700\times10^5$ Pa and the temperatures of 350～650℃, simulating compounding post-magmatic hydrothermal, volcanic pneumato-hydrothermal and underground brine hydrothermal solutions extracted $T_{Fe}=5.60\sim3881.89$ mg/l, majority were 50～2000mg/l from intrusive rock and volcanic rock, they made up ore-forming solutions. For example of the experiments of Makeng mine, under the conditions for the temperatures of 200～650℃ and the pressures of $200\times10^5\sim600\times10^5$ Pa, by 7～28 days and nights of experimental duration, interaction of acid hydrothermal solutions with diabase activated $T_{Fe}=69.65\sim3532.33$ mg/l from diabase. The data are equal to 21.17～326.28 kg/m^3 of iron from diabase. The diabase can provide T_{Fe}150～200 kg/m^3 if on the computing of pH=2.5 acid hydrothermal solution at 168 hours of duration of experiment under the temperatures of 300～500℃ and the pressures of $300\times10^5\sim500\times10^5$ Pa. The fade diabase in Makeng mine is distributed very widly, it can provide 1.5～2.0

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hundred million Ton/km^2.

(2) Under the temperatures of 350～550℃ and the pressures of 500×10^5～600×10^5 Pa, through 120 hours duration of experiment, interaction of simulating compounding metamorphic, migmatizatic and underground brine hydrothermal solutions with metamorphic rocks activated T_{Fe} 10.49～784.77 mg/l from sericite-quartz schist, biotite-quartz schist and diopsid-amphibole-plagioclase granulite. They made up ore-forming solution. These metamorphic rocks in Shilu, Mengjiagang, Ximashan, Tianbaoshan and Tongboshan mines are dispersed widly and with great thickness. The iron quantity from the rocks is very considerable. There are enough irons for providing respective iron deposits. This has proved that mudstone, muddy rock and mud shale of predecessors for sericite-quartz schist and biotite-quartz schist are the source beds of Shilu and Mengjiagang iron deposits, the plagiodase-amphibale schist and dark granulite of predecessors for migmatite and gneiss migmatite are the source beds of Ximashan, Tianbaoshan and Tongboshan iron deposits separately.

(3) Under the temperatures of 250～550℃ and the pressures of 300×10^5～1000×10^5 Pa and through 120～480 hours of experimental duration, interaction of simulating compounding underground brine hydrothermal solution with the W, Sn, Mo, Bi-bearing metamorphic rocks of Procambrian System-the wallrocks of tungsten and polymetallic deposit in Shizhuyuan extracted from the rocks: W quantity is 0.032～1.250 μg/ml, which accounted for 0.97～41.90% of W total value in original rocks; Sn quantity is 0.312～1.520 μg/ml, which made up 18.72～70.15% of Sn total value in original rocks; Mo quantity is 0.800～9.000 μg/ml, which amounted to 12.56～91.37% of Mo total value in original rocks and Bi quantity is 0.30～78.43 ng/ml, which ranked 0.23～76.47% of Bi total value in original rocks. This factor has showed that interaction of the underground hydrothermal solution with the wallrocks-metamorphic rocks activated W, Sn, Mo and Bi from the rocks to migrate to the contact zone of granite and the place nearby. And then, the W, Sn, Mo and Bi metals again centered and formed mineral deposits in favourable structural location at the sudden changing of temperature, pressure, acid-alkalinity of the solution and oxdation reduction environment.

(4) Under the temperatures of 250～400℃ and the pressures of 200×10^5～500×10^5 Pa and through 120～480 hours of experimental duration, interaction of simulating compounding underground brine hydrothermal solutions of pH=1.5～4.5 with the wallrocks of Dexing porphyry copper deposit -Cu, Mo, Pb and Zn- bearing metamorphic rocks of pro-Sinian System extracted from the rocks: Cu quantity is 29.01～1654.24 ng/ml, the highest of them comes up to 22685.00 ng/ml; Mo quantity is 200.00～2720.00 ng/ml, the highest of them achieves 9990.00 ng/ml; Pb quantity is 34.90～106.20 ng/ml, the highest of them reaches 271.90 ng/ml; Zn quantity is 20.00～800.00 ng/ml, the highest attains 3200.00 ng/ml. They come up to the standards of ore-forming solution completely. On rough computing of experimental data, the 115 km^2 metamorphic rocks of Cu

etc. deficits around granodiorite-porphyry can provide Cu 4986 ten thousand Ton, which is enough to form the Cu reserves for Dexing porphyry copper deposit. It shows that the metallogenic materials of Cu, Mo tec. in the copper deposit stemed from the metamorphic rocks of pro-Sinian System.

(5) Under the temperatures of 150~450℃ and the pressures of 300×10^5~900×10^5 Pa and through 120~480 hours of experimental duration, interaction of simulating compounding volcanic pneumato-hydrothermal solution of pH=2.0~11.0 with the wallrocks of Honglazi gold deposit Au-and Ag-bearing volcanic rocks (rhyolite-porphyry, andesite, tuff, breccia etc.) activated from the rocks: Au quantity is 0.046×10^{-6}~0.120×10^{-6}, which made up 17.27~98.33% of Au total value in original rocks. The average value is equal to 0.03 g/T; Ag quantity is 0.37×10^{-6}~39.52×10^{-6}, which ranked 33.33~99.05% of Ag total value in original rocks. The average value is equal to 20 g/T. Au 103×10^7 ton and Ag 3.05×10^8 ton were activated from Mesozoic Era volcanic basin of Yi County (long 200 km, width 39 km, the total thickness of the layers is 4311 m.). On the computing of Hong lazi basin (long 60 km, width 27 km), the rocks can provide Au 69.9 ton and Ag 4.64×10^4 ton. It shows rhyolite-porphyry and andesite are the main layers of source for metallogenic material.

(6) Under the temperatures of 200~50℃ and the pressures of 500×10^5~200×10^5 Pa and through 168~672 hours of experimental duration, interaction of the distilled water for compounding pH=5.5~8.5 with the bedrock of delegating Xiaotangshan spring areas - 90% dolomite and 10% granite activated from the rocks that the contents of main ions and oxides for K^+, Na^+, Ca^{2+}, Mg^{2+}, Al^{3+}, ΣFe, HCO_3^-, NO_3^-, SO_4^{2-}, F^-, Cl^- and SiO_2 etc., dissoloved solids and the water quality class tallied with the facts of hot, mineral springs in Xiaotangshan area. It shows the underground hydrothermal water and hot, mineral springs in Xiaotangshan area are formed at above-mentioned physico-chemical conditions. It provided the theoretical bases for the founding of man-made warm spring and the developing of geothermal field.

CONCLUSTON

On the basis of field geological works in aforementioned 17 mines (spring area), author systematically performed the experiments of water-rock interaction at the temperatures of 50~650℃ and the pressures of 100×10^5~1400×10^5 Pa, and combined with studies of the thermodynamic properties for the phase equilitrium. The results prove that:

(1) The metallogenic materials in the iron deposit of contact metasomatic type mainly stem from the intrusive rocks; The metallogenic materials in the iron deposits of volcanic pneumato-hydrothermal metasomatic type originate chiefly from volcanic and subvalcanic ricks. The metallogenic materials in the iron deposits of sedimentary-metamorphic metasomatism and migmatization principally come from the sedimentary rocks and metamorphic

rocks.

(2) The wallrocks of the porphyry copper deposit in Dexing, Jiangxi -Cu and Mo etc. -bearing metamorphic rocks of pro-Sinian System are their beds of ore source.

(3) The wallrocks of tungsten polymetallic deposits in Shizhuyuan, Hunan -W, Sn, Mo and Bi -bearing metamorphic rocks of Procambrian System are their beds of ore source.

(4) The Au, Ag of the gold deposit in Honglazi, Liaoning principally stem from rhyolite-porphyry and andesite.

(5) The spring-forming materials in Xiaotangshan spring area, Beijing, chiefly originate from the dolomite of Sinian System, and secondly come from the crystalline rocks of granite etc..

At the same time with that, author brought to light the mechanism on the activation, migration and precipitation for the ore-forming elements of Fe, W, Sn, Mo, Bi, Cu, Pb, Zn, Au, Ag and spring-forming materials of the 12 hydrochemistry ions for K, Na, etc. as well as oxides. And then pointed out the passage of quality and exchange of heat quantity in various reactions of water-rock interaction are the most important factor in all physicochemical conditions.

序

水—岩相互作用或水—岩反应研究在矿产资源、环境科学、灾害防治及全球变化诸领域中均占十分关键的地位。经过近年来的实践、深思和讨论、提高，上述意见已逐渐在地球科学界取得了共识。同样，水—岩相互作用实验研究也是二十年来国内外实验矿物、岩石、地球化学学科中的热门课题，是地球科学前沿问题之一。回忆二、三十年前大量涉及水、矿物、岩石之间的实验，多侧重一定温、压条件下的平衡态研究，开展水—岩相互作用实验在一定程度上突破了平衡态研究，进入更符合地质实际的动态及非平衡态实验。

本专著，即《水—岩相互作用和成矿物质来源》是梁祥济教授及其同事二十年从事水—岩相互作用实验的系统总结。在工作中梁教授开拓了新的思路，采取了新的技术路线，即取材于我国多种金属矿床及热泉的实际资料，进行细致的野外观察，剖析各种现象与事实，并从中构筑水—岩相互作用实验依据和设计实验方案。经过多种实验后将实验结果与地质观察相结合，力求得出恰如其分的合理解释。这与国外目前流行的海水—玄武岩相互作用实验可以互补互济，确有异曲同工之妙，而梁教授的工作与中国的矿床实际密切结合，更属别具一格。

从二十年来所进行的大量水—岩相互作用实验，梁教授得出的结论是：成矿金属可来源于含矿围岩。在经过水—岩相互作用后，围岩中所含诸种金属可被活化、转移，并在构造有利部位富集而形成矿床。用作者的语言，“这为打破一切成矿物质由深部岩浆带上来的旧观念，建立成矿物质多来源，尤其是为热液矿床成矿物质从围岩中来的新概念提供了依据。”

看来，围岩为成矿提供一定物质来源，这一论点除有其实验依据外，在一些矿床也可观察到地球化学依据，如在靠近矿体一定范围内出现的围岩中成矿金属的负异常等。成矿物质可来自远处（如深部岩浆、上地慢），也可来自近处（如含矿围岩）。正如生油物质可来自近处的生油岩，也可来自远处的生油岩（数十公里甚至上百公里之外），也可来自深部一样。当然，就某个具体矿床或油气田而论，成矿或成油物质可能具单一来源。

水—岩相互作用是地球科学中一个有着丰富内容和广阔天地的崭新研究领域。相信在梁教授之后会出现更多的实验工作、设想与应用。

涂光炽

1995.5.7

目　　录

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