Kuroko Deposit Modeling Based on Magmatic Hydrothermal Theory

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Abstract: Three lines of evidence which support the magmatic hydrothermal origin of kuroko deposits are evaluated.

(1) Mineralization of kuroko deposits took place around 15 Ma, at the same time as several other geologic episodes including a major shift in the tectonic stress field, a peak of bimodal volcanic activity, and a maximum degree of subsidence. The style of eruption of rhyolite changed from lava flows, acid hyaloclastites, and lithic tuff breccias before mineralization to extensive post-ore pumiceous pyroclastic flows. In an analogy with the Yellowstone caldera, absence of pre-ore basalt directly below the known kuroko deposits suggests that contemporaneous acidic magma existed at a depth of a few kilometers with the potential to release magmatic fluid. Basalt lava is often seen in the hanging-wall sequence of kuroko deposits; it extruded after the acidic pluton had solidified subsequent to mineralization. The post-ore pumiceous pyroclastics could have been produced by violent submarine eruptions similar to those related to the formation of Valles type calderas from resurgent acidic magma. Apparently such an abrupt release of material from the magma chamber was not suitable for conditions to develop and form kuroko deposits. (2) Based on oxygen and hydrogen isotopic study, sea water cannot be the sole source of the kuroko ore solution. Metals cannot be leached from footwall volcanic rocks, because an outward flow of hydrothermal fluids within the volcanic units is deduced from wall-rock alteration study. In addition, the notable homogeneity of isotopic compositions of ore minerals rules out the possibility that the ore metals were largely derived from basement rocks by leaching. (3) The rhyolitic magma which was contemporaneous with ore deposition had a large potential to result in separation of a metal-rich aqueous solution during its crystallization. This aqueous phase would have been a very efficient ore-forming solution, even after dilution by convecting sea water. This magmatic hydrothermal model of kuroko deposits is more consistent with the observed geological features than other models.

Statement of the Problem

Like many other volcanogenic massive sulfide deposits (hereafter termed massive sulfides), Japanese kuroko deposits are regarded strata-bound or time-bound in a broad sense. The reason for the exclusive occurrence of kuroko deposits within a volcano-sedimentary sequence of the Hokuroku district (Fig. 1)

Fig. 1 Distribution of pumice, basalt, and mudstone type kuroko deposits (See text for explanation of each type of deposits). The numbers correspond to the deposits shown in Fig. 3. The first two types of deposits tend to occur in the eastern part of the region, whereas the mudstone type kuroko deposits tend to occur in the Japan Sea side of the Honshu Island, where the intensity of acidic volcanism was weak. The kuroko deposits cluster in Hokuroku (deposits 1, 2, 5, 6, 7) and Aizu (3, 4) districts. The Green Tuff region is shaded.

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around 15 Ma is a fundamental question that must be answered through characterization of the entire geologic setting which was relevant to the formation of the deposits (KAJIWARA and HIRAYAMA, 1983). In other words, the proposed model must include an explanation for the reason why the mineralization was limited to a short period of time, despite the fact that volcanism continued throughout the entire Late Cenozoic period. However, an effort to solve this problem has not been attempted except by a few workers such as KAJIWARA and HIRAYAMA (1983), who suggested the biogenic origin of kuroko deposits from “Primitive Undifferentiated Metalliferous Organic Sediments” (KAJIWARA, 1982). Therefore, this paper is an attempt to build an alternative genetic model of kuroko deposits based on a magmatic hydrothermal hypothesis.

### Known Geologic Events around 15 Ma

A large amount of information has been ac-
cumulated on the geologic setting of the kuroko deposits since the compilations edited by Ishihara (1974) and Ohmoto and Skinner (1983). One of the most remarkable conclusions, obtained through paleomagnetic surveys, is that there was a rapid anti-clockwise rotation of Northeast Japan around 15 Ma (Hamano and Toshia, 1985; Tsunakawa and Takeuchi, 1983); this could have resulted in the opening of the back-arc basin, the Japan Sea (Otofuji et al., 1985). Many other characteristic episodes happened around that time, including the formation of kuroko deposits (Fig. 2).

The initial phase of volcanism began about 40 Ma (Monzen stage) in a broad area of Northeast Japan (Konda and Ueda, 1980). Volcanic products during this phase were dominated by andesite with a small amount of calc-alkaline basalt and rhyolite (column F in Fig. 2). The andesitic volcanism extended from Southwest Japan around 23-24 Ma (Matsuda, 1983) to “Oyashio ancient landmass” off the Pacific coast of Northeast Japan at 20-23 Ma (Yanagisawa et al., 1980). Some of the andesitic tuff units have a welded texture and are considered to be subaerial (column E). This phase of volcanism was followed by a long period of quiescence (column F). There are no K-Ar ages for volcanics in a range between 20 and 17 Ma in Northeast Japan, based on compiled data (Konda and Ueda, 1980; Kaneoka, 1983; Tsunakawa et al., 1983). The observed time gap roughly coincides with the Daijima stage (column C) and is unlikely to be a result of sampling bias, because the samples were taken systematically.

The second phase of volcanism is characterized by its bimodal nature (Konda, 1974). Tholeiite basalt and calc-alkaline rhyolite (Togashi, 1978) were erupted during the Nishikurosawa and Onnagawa stages as a consequence of active east-west extension; this agrees with the observation by Eichelberger (1974) who correlated bimodal volcanism to extensional stress fields within the continental crust. A large amount of acidic rocks accumulated during the early stages of this phase, mainly along the “Backbone range” (axial part) of Northeast Japan (column G); these rhyolites host the kuroko deposits (column H).

The depth of the sea could have been at its greatest during the Nishikurosawa-Onnagawa stages due to the combination of a rise in sea level (column E) and volcano-tectonic subsidence (Fuji, 1974). Guber and Merrill (1983) and Matoba (1983) estimated the depth of the trough as 3400-4100 and 2000-3000 meters, respectively, from foraminiferal data. Kitazato (1983) argued, based also on the foraminiferal data, that there were north-south trending “Kuroko deep” (eastern trough) and the “Akita Oil Field deep” (western trough) in Northeast Japan and they had an average depth of 1000 and 1500 meters, respectively. The reason for the large discrepancy in the opinions of paleontologists about the depth estimate of the sea is not obvious. The minimum depth required to prevent the boiling of kuroko ore solution at 350°C is calculated to be about 1800 meters according to the analyses of NaCl and CO₂ in fluid inclusions (Pisutha-Arnond and Ohmoto, 1983).

During the Nishikurosawa stage, the paleogeography of the Japan arc consisted of an archipelago with a tidal marsh surrounding those islands under tropical climate (Tsuchi ed., 1981). Tropical or sub-tropical foraminiferal fauna of the Nishikurosawa stage was replaced by cold current types of the Onnagawa stage in a short period of time (Maiya and Inoue, 1981).

Based on the available information, volcanism was reactivated when the regional stress field changed at about 22 and 15 Ma. These shifts lasted 1 to 2 million years, though the associated volcanism had a duration of 2 to 3 million years (Tsunakawa and Takeuchi, 1983). Masuda (1984) ascribed these episodes to the oscillatory movement of the Pacific plate; this movement is recorded in the periodic change in rotation of loci of volcanic centers along the Hawaii-Emperor chain (Jackson et al., 1975).

Therefore, the formation of kuroko deposits can be correlated with several geologic events, including; 1) a major shift in the global tectonic stress field, 2) active bimodal volcanism dominated by rhyolite and a sub-
ordinate amount of basalt, 3) active rifting and rise of sea level to form deep trough structure, and, 4) a climatic change and/or an abrupt change in the flow of sea currents. Some of these phenomena may have played an important role to form and preserve the kuroko deposits on the sea floor.

Ohmoto et al. (1983b) stressed that the formation of all the kuroko deposits are not strictly contemporaneous, even within the Hokuroku district, based on K-Ar dating of the surrounding volcanic rocks. Nakajima (1986) came to a similar conclusion on the basis of detailed geological mapping and correlation. This evidence is particularly important because it can be used to specify which geologic event(s), among the four candidates listed above, actually triggered the formation of the kuroko deposits.

Change in the Mode of Volcanism around the Kuroko Horizon

Altered andesite of the Menaichizawa Formation of the Hokuroku district is usually correlated with the Monzen and/or Daijima stages since part of the andesite covers coal measures of the Monzen stage (Column I; Fig. 2). Considering the break of volcanic activity during the Daijima stage, the andesite is likely to be of Monzen age. The lateral extent of the andesite coincides well with that of overlying acidic volcanic rocks in the western Hokuroku district (Horikoshi, 1983). On the contrary, no such andesite is known in eastern half of the district. Less than half of the kuroko deposits in Japan have such prior andesite in association; therefore, it is unlikely that the andesitic volcanism was essential as a source of heat or metals in the formation of the kuroko deposits.

The Nishikurosawa Formation of this district is characterized by the Hotakizawa basalt flow and subsequent voluminous rhyolitic activity. Hotakizawa basalt fills presumed depressions and shows a positive correlation with the thickness of mudstone (M3) in the base of the Nishikurosawa Formation (Horikoshi, 1983). It is noteworthy, however, that the basalt has never been found directly below the known kuroko deposits. In other words, despite the close spatial association of middle Miocene basalt and kuroko on a regional scale (Horikoshi, 1976), the distribution of kuroko deposits and footwall basalt do not overlap on a local scale.

Three units of rhyolitic tuff breccias are recognized within the Nishikurosawa Formation in the western Hokuroku district (Takahashi and Sug, 1974). Poorly vesiculated essential lithic fragments are predominant in these units, which often grade both laterally and vertically into auto-brecciated lava and acid hyaloclastite. Therefore, such units are designated as lithic tuff breccias in this paper. On the other hand, the mode of occurrence of the acidic rocks of the Onnagawa Stage (Shigenai Formation in Column I of Fig. 2) shows a clear contrast with that of the underlying units. Extensive pumiceous pyroclastic deposits characterize the volcanism of the stage (Urabe, 1979). Only few lava flows near Hanaoka are known, in spite of the fact that these acidic rocks have a similar chemical composition to the footwall rhyolites (Hashimoto and Fujita, 1983). This is an important observation which gathered little previous attention. In an outcrop about 3 km northwest of the Fukazawa mine, mudballs up to 40 cm large commonly occur near the base of a graded pumice bed about 10 meters thick. Pisolites occur sometimes in this ash fall deposits of the Onnagawa stage (M. Utada, personal comm.), leading Utada to consider that the depth of the “Kuroko deep” might have become shallower considerably during the Onnagawa stage. The relative upheaval of the volcanic center may be the reason for the violent submarine pyroclastic eruption.

A detailed examination of the volcanic succession indicates that the timing of kuroko formation was not unique. Two to three ore horizons are recognized in several places such as the Uwamuki deposits, Kosaka mine (Urabe and Sato, 1978), the Hanaoka-Shakanai area (Ito et al., 1974; Kumita et al., 1982), the Hanawa mine (Ohtagaki et al., 1974b), and the Yokota mine (Hirabayashi, 1974). The mineralization seems to have been triggered
not by an unique time-bound phenomenon but by the eruption of lithic tuff breccia or the extrusion of rhyolite lava from nearby individual volcanic centers (Horikoshi, 1969), since most of the orebodies rest exclusively on these volcanic rocks without any intercalated epiclastics.

Three Types of Kuroko Deposits
Classified by Hanging-wall Rocks

Simplified geologic sections for 13 kuroko deposits, both within and outside the
Table 1 Classification of Kuroko deposits on the basis of the dominant rocks in hanging wall sequence. Explanation of these three types is given in text. Localities of the mines listed are shown in Figure 1. Large gypsum deposits tend to be associated with mudstone type deposits.

<table>
<thead>
<tr>
<th>Types</th>
<th>Hanging-wall Rocks</th>
<th>Average ore reserves (ton)</th>
<th>Massive Black ore (Pb-Zn)</th>
<th>Sulfide Yellow ore (Cu-Fe)</th>
<th>Breccia dikes</th>
<th>Examples (deposits)</th>
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<tr>
<td>Pumice</td>
<td>pumice tuff</td>
<td>$10^7$</td>
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<td>frequent</td>
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<td>Hanawa</td>
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<tr>
<td>Basalt</td>
<td>pumice tuff</td>
<td>$10^6 - 10^7$</td>
<td>large</td>
<td>small</td>
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<td></td>
<td>Akita-Yoshino</td>
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<tr>
<td>Mudstone</td>
<td>mudstone</td>
<td>$10^5 - 10^6$</td>
<td>small</td>
<td>small or lacking</td>
<td>rare</td>
<td></td>
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Hokuroku district (Fig. 1), are shown in Figure 3. The footwall acidic rocks are either lava or lithic tuff breccia, both of which often grade into auto-brecciated lava or hyaloclastite. In contrast to the relatively monotonous occurrence of acidic rocks beneath the kuroko deposits, the hanging-wall rocks show a wide variation in composition. It is possible, therefore, to classify the deposits into three types based on the dominant rock facies in the hanging-wall sequence. The overlying acidic rocks, if any, are usually well vesiculated pumiceous pyroclastic flow deposits as was discussed before. These pumiceous pyroclastics are observed only in the Hokuroku and Aizu districts, which are the first and second largest kuroko mining areas in Japan. This group of kuroko is designated here as the pumice type. The general sequence for this type is footwall lithic tuff breccia, kuroko, mudstone, pumiceous pyroclastics, and basalt flow, in ascending order (Fig. 3).

In some cases, basaltic activity precedes the deposition of any pumiceous pyroclastics. Therefore, kuroko deposits which have a thick basalt flow over the orebody constitute the second group and is named the basalt type. The Furutobe and Fukazawa deposits, both within the Hokuroku district, and the Takara deposit, about 100 km west of Tokyo (Fig. 1), belong to this type. The last type of kuroko deposition is characterized by a thick mudstone bed above the deposit. Volcanic activity seems to be extinct in these areas except for a small amount of basalt or dolerite of the Onnagawa stage. These mudstone type kuroko deposits are usually small and tend to occur in the western part of the kuroko belt or in the Oil Field belt, where rhyolitic volcanism was less intense than in the areas of the other two types of deposits (Fig. 1).

Several other features of the kuroko deposits distinguish these three types (Table 1). For example, the volume of Yellow Ore (chalcopyrite-pyrite ore) in the deposit is large in the pumice type, intermediate in the basalt type and usually small or lacking in the mudstone type deposits. The difference in the copper content among these three types may have been related to the difference in the temperature of the corresponding hydrothermal system (as observed at the East Pacific Rise 21°N hydrothermal field; e.g. Goldfarb et al., 1983). Abundant chalcopyrite in the first two types probably indicates a higher temperature of the ore-forming system than that of the mudstone type deposits. (Readers may refer to the discussion of Ohmoto et al., 1983, about temperature dependence of the precipitation of metals.) This is consistent with the observation that breccia dikes are common in the pumice and the basalt type deposits and are very rare in the mudstone type. It is likely that both the intensity of igneous activity and, in turn,
the temperature of the accompanying hydrothermal processes decrease from pumice, through basalt to mudstone types.

**Constraints on Kuroko Volcanism: Evidence from the Yellowstone Caldera**

The resurgent caldera within the Yellowstone Park, western United States, is regarded as an example of the hot-spot type volcanism under extensional tectonic stress (IYER, 1979). Volcanism has comprised three cycles of activity with an average duration of 0.7 million years. The cycle of rhyolitic activity always begins with subaerial tholeiite basalt lava flows and a small amount of rhyolitic products. This is followed by voluminous ash flow eruption of rhyolitic composition. The basaltic activity is limited to the peripheral area during the build up of the acidic magma (EATON et al., 1975). Acidic magma is believed to be generated by whole-melting of the lower crust from the heat of basaltic magma from the mantle. Once an acidic magma body is generated, the basaltic melt cannot penetrate the pluton because of density differences.

The absence of basalt directly below the kuroko deposits suggests that acidic plutons existed deep under the area during the Nishikurosawa stage, by analogy with the Yellowstone caldera. Hanging wall pumiceous deposits are volcanic ejecta from a catastrophic Valles-type caldera eruption which must have occurred during Onnagawa time. Post-ore basalt or dolerite dikes often cut the orebody in Hanaoka and Fukazawa mines; judging from their distribution, the feeder zone of the basalt must have been very close to the kuroko deposits. The presence of hanging wall basalt closely associated with kuroko deposits suggests that mineralization ceased once the acidic pluton had solidified. In other words, the extrusion of the basalt may indicate the termination of the kuroko mineralization, which was caused by the end of the molten magma indispensable for mineralization. The common occurrence of granodiorite fragments in breccia dikes which cut the kuroko ore bodies of the pumice type is strong evidence for the existence of underlying acidic pluton (URABE and SATO, 1978).

The genetic relationship between caldera formation and kuroko mineralization has been repeatedly inferred by many authors (HODGSON and LYDON, 1977; OHMOTO, 1978; KOUDA and KOIDE, 1978; OHMOTO et al., 1983b; TAKAHASHI, 1983; and HORIKOSHI, 1983). However, none of them agree in details, such as size, location, and time of formation of the presumed calderas. The diversity of opinions indicates the difficulty of reconstructing the caldera structure, if any, from existing bore hole data. The observed lines of evidence include: (1) Volcanic ejecta of the footwall units have their lateral distribution confined within estimated caldera structures (e.g. KOGA and TAKAHASHI, 1985; ISHIKAWA et al., 1976). Or, vice versa, the caldera structure was deduced from the circular distribution of the volcanics. (2) Volcanic depressions existed, at least in part, prior to the kuroko deposition (e.g. HORIKOSHI, 1983). (3) There are no voluminous ash flow deposits in the footwall units.

An explanation of these observations is presented in Fig. 4. The initial phase of the volcano-plutonic complex prior to kuroko mineralization was an accumulation of lava or lithic tuff breccia above large acidic plutons, thus building the "Kuroko volcano" (cf. evidence (1); as was pointed out by ISHIKAWA et al., 1976). Because of the volume loss from the magma, the area of the kuroko volcano subsided to form a volcanic depression (cf. evidence (2)). Volatile components, accumulated at the top of the magma chamber, were released along fractures as magmatic fluid. This fluid was involved in the kuroko ore solution. The acidic pluton solidified and was penetrated by basaltic magma, after mineralization. Resurgent acidic magma was generated (in the case of the pumice and basalt types) and then erupted possibly due to the heat of the post-ore basalt during the Onnagawa stage.

**Wallrock Alteration and Source of Metals**

Recent investigations on wallrock alteration around the kuroko deposits (UTADA, 1980; ISHIKAWA et al., 1980; HASHIGUCHI et al., 1981;...
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(1) Eruption of basalt & crustal melting.

(2) Formation of Kuroko above pluton.

(3) Rhyolitic pyroclastic flow.

(4) Basalt through solidified pluton.

Fig. 4 Schematic illustrations of the evolution of the volcano-plutonic system beneath the kuroko deposits.

(1) Basaltic activity had started due to active extension related to the opening of the Japan Sea around 15 Ma (Otofuji et al., 1985). Lower crustal rocks were melted from the heat of the basalt. (2) Formation of the kuroko deposits above the bimodal magma chamber at around 15 Ma, shortly after the effusion of acidic lava and tuff breccia. (3) Catastrophic pyroclastic eruption occurred resulting in the formation of extensive pumiceous deposits during the Onnagawa stage. This may have been due to the relative upheaval and decompression of the high level pluton. (4) Basaltic magma penetrated through solidified acidic magma after pyroclastic eruption. Kuroko mineralization ceased, though some vein deposits formed subsequently.

Utada et al., 1983; Urabe et al., 1983) all demonstrated that kuroko deposits in the Hokuroku area have well-developed alteration halos. Date et al. (1983) revealed that relative depletion of sodium and calcium and relative enrichment of magnesium and potassium in host rocks can be used as useful criteria in the exploration for the kuroko deposits. Urabe et al. (1983) showed that other types of volcanogenic massive sulfide deposits also had alteration halos of similar diameter, about 2 to 4 kilometers. As a matter of course, massive sulfides tend to occur in areas of high heat flow near the center of contemporaneous volcanism. The outer limit of the massive sulfide alteration halo is therefore more or less obscured by the diagenetic alteration which is common in such areas.

There are several characteristics which are common among massive sulfide alteration. First of all, a well developed alteration zoning exists around a deposit and the number of minerals within each alteration zone increases outwards. The activity of hydrogen ion relative to alkalis and alkaline earth elements always decreases outwards, i.e. the pH increase (Urabe et al., 1983). These observations are explained by a process where a relatively acidic solution not in equilibrium with the surrounding rocks is introduced to the center of the alteration halo to form a water-dominated hydrothermal system. The observed zonation is a "fossil" of the disequilibrium between the solution and the rocks. The system becomes rock-dominated at the margin and the physico-chemical conditions of water-rock in-
teraction cannot be distinguished from those of diagenesis.

High temperature natural solutions are enriched in potassium relative to sodium (Fournier and Truesdell, 1973), with potassium being added to the mineral assemblage as the temperature of the solution decreases. Therefore, a potassium enrichment can occur by injection of high temperature solution to the system. Fluid inclusions in minerals from deeper levels of the stockwork ore commonly have maximum salinity twice that of seawater (Urabe and Sato, 1978; Bryndzia et al., 1983). The quartz-sericite-sulfide assemblage observed in the stockwork ore could be the products of such saline and high temperature solutions. This potassium-enriched core is usually surrounded by sericite-chlorite-quartz rock (Urabe et al., 1983). This magnesium enrichment could be the result of shallow mixing of seawater with the high temperature fluid, since heated seawater precipitates magnesium instantaneously (Seyfried and Bischoff, 1977).

The oxygen isotopic composition of the surrounding rocks is a good indicator of hydrothermal alteration (Urabe et al., 1983; Matsu-hisa and Utada, 1983; Green et al., 1983). The whole-rock $\delta^{18}O$ values always show a remarkable increase outwards from the mineralized center of the studied kuroko deposits. The whole-rock $\delta^{18}O$ value of the altered rock is a function of alteration temperature, isotopic compositions of rocks and fluid, water-rock ratio (Spoonier and Fyfe, 1973), kinetic rate of oxygen exchange reaction (Cathles, 1983), and the alteration mineral assemblages. However, the principal factor which controlled the isotopic composition of the altered rocks was the temperature (Urabe et al., 1983; Matsu-hisa and Utada, 1983). Therefore, the oxygen isotopic zoning around the kuroko deposits is a fossil of the temperature drop from the center towards the margin of the hydrothermal system. The lateral thermal gradient within the Tertiary volcanic rocks is calculated to be 1°C per 10 meters at the Uwamuki deposits, Kosaka mine (Urabe et al., 1983). Such a steep thermal gradient is characteristic of the discharge zone in present day geothermal systems (cf. White, 1970). Thus, the highly permeable footwall tuff breccia of the Uwamuki deposits is regarded as the discharge zone (Urabe et al., 1983). A similar conclusion was reached by Green et al. (1983) for the Fukazawa mine.

In other words, the main vector of the fluid flow within the footwall volcanic rocks was from the center of the discharge zone to the margin of the geothermal system (model B in Fig. 5). Therefore, the footwall volcanics are not a possible source of metals through leaching by convective seawater. The well-developed pyritized halo around the kuroko deposits of the Hanaoka area (Nakahama and Sasaki, 1985) and relative enrichment of copper and zinc, rather than depletion, in volcanics around the Fukazawa deposit (Ishikawa et al., 1976; Green et al., 1983) support this conclusion. The lateral migration of kuroko solutions would have added the excess base metal and sulfur to the rocks. The possible area left to consider as a source of metals through leaching (model A in Fig. 5) consists of basement rocks. However, this possibility is eliminated in the following section.

Constraints from Isotope Geochemistry

Kuroko deposits have an abundance of isotopic data. However, these data are not adequate to arrive at an unique conclusion about the source of the ore elements (e.g. Franklin
Fig. 6 Summary of isotopic compositions of kuroko ore fluid and minerals such as sulfate (strontium) and sulfide (sulfur and lead). Compiled from OHMOTO et al. (1983a), PISUTHA-ARNOND and OHMOTO (1983), HATTORI and SAKAI (1979), HATTORI and MUEHLENBACHS (1980), NAKAJIMA and SASAKI (1985), FARRELL and HOLLAND (1983), SATO et al. (1981), SASAKI et al. (1982), FEHN et al. (1983), and other sources cited in these works. Open triangles denote the isotopic composition of the relevant elements in seawater. See Appendix (Fig. A-1) for the hydrogen and oxygen isotopic composition of the kuroko solution. The carefully analysed lead isotope data by SASAKI et al. (1981) are superimposed by closed square on the data by FEHN et al. (1983).

The most noticeable characteristic of the kuroko lies in the remarkable homogeneity in terms of their isotopic compositions. For example, sulfur, strontium and lead isotopes all have much smaller ranges than those of footwall rocks (Fig. 6). This is rather striking if we consider the fact that the kuroko solutions had travelled through various combinations of rock types before it discharged at sea bottom. The maximum variation in $^{206}\text{Pb}/^{204}\text{Pb}$ ratios of nine specimens from various kuroko deposits is 0.16 percent if we only consider the controlled analyses (SATO et al., 1981; SASAKI et al., 1982) (Fig. 6). This range is similar to the reproducibility ($2\sigma$) of the measurement. SASAKI et al. (1982) suggested that the kuroko lead must already be homogeneous within the source, since kuroko deposits more than a few hundred kilometers apart have nearly identical lead isotopic compositions.

The variation range of isotopic compositions of the hydrothermal deposits of the East Pacific Rise at 21°N are comparable to those of the kuroko deposits, in spite of the fact that the rocks underlying the East Pacific Rise deposits are exclusively ridge basalt. For example, the $\delta^{34}\text{S}$ value of the sulfides from 21°N ranges from +1.3 to +4.1 per mil (STYRT et al., 1981). The $\delta^{18}\text{O}$ value of anhydrite in the East Pacific Rise deposits shows a larger scatter (0.70580–0.70846; ALBAREDE et al., 1981) than that of the kuroko deposits. Any genetic model of the kuroko deposits, therefore, must explain the homogeneous isotopic compositions.

Before considering the origin of the kuroko solutes, the nature of the kuroko ore solution must be discussed. There are three possible sources of kuroko solution, namely; (1) meteoric water, (2) magmatic fluid or deep-circulating water which was once in equilibrium with molten magma, and (3) seawater or its evolved equivalent after interaction with host rocks. The contribution of (1) has been inferred by several geologists to explain the presence of some low salinity fluid inclusions (e.g. MARUTANI and TAKENOUCHI, 1978) as well as the lateral variation of metal ratios in the deposits of the western Hokuriku district (HORIKOSHI and SHIKAZONO, 1978). However, the incorporation of meteoric water is not plausible due to the large water depth of the kuroko deep (OHMOTO et al., 1983b).

The $\delta D$ value determined for the kuroko solution is less than $-10$ per mil (SMOW; Fig. 6). OHMOTO and RYE (1974) tried to explain this range as being due to the interaction of seawater with volcanic rocks. However, their model requires that the water content of rocks should remain unchanged after water-
rock interaction (HATTORI and SAKAI, 1979). Apparently, this assumption is unrealistic since the altered volcanics always have a much higher water content than their unaltered equivalents. Since the $\delta D$ value of sea water always increases through ordinary water-rock interaction at hydrothermal temperatures, sea water cannot be the sole source of the observed kuroko solution (OHMOTO et al., 1983a).

OHMOTO et al. (1983a) rejected the idea of a magmatic origin for kuroko solutes on the basis of sulfur isotope data. Their two reasons were; (1) fluids derived from acidic magmas are likely to contain 0.1 molal of total dissolved sulfur, whereas the kuroko fluids appear to contain between 0.01 to 0.001 molal of total sulfur, and (2) magmatic fluids which have average $\delta^{34}S$ value of +5 per mil are unlikely to form sulfides of the observed isotopic composition (about 5 per mil) at temperatures lower than 300°C. The first objection is easily discounted by the dilution of the magmatic fluid by a factor of ten as a result of the late incursion of oxygenated seawater into the upper portion of the mineralizing system. Regarding the second point, OHMOTO et al. (1983a) computed the $\delta^{34}S$ value of $H_2S$ to be $+5 \pm 3$ per mil at 250°C, assuming that the average $\delta^{34}S$ composition of fluids to be $+23$ per mil and the ratio of $(SO_4^{2-}/H_2S)$ equal to one. These assumptions are not warranted, since deposition of the kuroko occurred on the sea floor where a steep gradient in redox state is inferred. Only a slight change in the $SO_4^{2-}/H_2S$ ratio would result in large shifts in the sulfur isotopic composition of the kuroko deposits. Hence, such a delicately balanced model is unlikely to result in the notable homogeneity of sulfur isotopic composition of the kuroko (Fig. 6).

OHMOTO et al. (1983a) proposed a new model, designated as the anhydrite buffer model, because biogenic or inorganic reduction of seawater sulfate to obtain hydrogen sulfide (OHMOTO and RYE, 1974) will not explain the uniform and positive $\delta^{34}S$ value of ore sulfides. According to their hypothesis, anhydrite precipitated in volcanic rocks during diagenesis or in the early stage of hydrothermal activity and was then partly reduced at 150-300°C to generate sulfide ion by a delicate balance in the redox state. However, the observed narrow range in sulfur isotopic composition of kuroko anhydrite (WATANABE and SAKAI, 1983) cannot be explained by the partial reduction of sulfate from anhydrite. This is probably the reason why OHMOTO et al. (1983a) also considered the leaching of igneous sulfur fixed as pyrite in the rocks to explain the relative enrichment of $^{34}S$ of the kuroko sulfide. However, the sulfur content of the fresh volcanics is very low (NAKAYAMA and SASAKI, 1985) and the contribution of the “igneous” pyrite from the surrounding rocks would be negligible.

In contrast, the magmatic hydrothermal theory of kuroko deposits (URABE and SATO, 1978; URABE, 1985) accounts for all of the isotopic data; (1) the homogeneous isotopic composition of kuroko minerals is a result of the magma being a well-mixed source of metals, (2) the low $^{87}Sr/^{86}Sr$ ratio in barite (FARRELL and HOLLAND, 1983) is due to the less radiogenic nature of the lower crust where the acidic magma was generated, (3) the oxygen and hydrogen isotope trend of kuroko fluids is due to a physical mixing of magmatic fluid with sea water (HATTORI and MUEHLENBACHS, 1980), (4) the coincidence of the isotopic compositions of ore sulfur and igneous sulfur reflects the origin of the ore sulfur (ISHIHARA and SASAKI, 1978), and (5) the pattern of wall rock alteration and the structure of the inferred hydrothermal system (URABE et al., 1983) is controlled by a deep magmatic source.

Support from Experiments on Hydrous Magma

Recent experimental results on the partition of base metals between coexisting acidic silicate melt and aqueous fluid indicate that the metals are strongly partitioned towards the fluid phase if the melt is; (1) rich in chlorine, (2) aluminous, and (3) under low confining pressure (i.e. less than 1 kbar) (URABE, 1984, 1985, 1987). An acidic magma body related to the formation of the kuroko deposits will satisfy these requirements.
The common occurrence of intensely altered clasts of granitoid in breccia dikes near the pumice type and basalt type kuroko deposits indicates that a high level pluton existed during the Nishikurosawa stage. A heat budget calculation also suggests that there was a large pluton beneath the known kuroko deposits (Urabe and Sato, 1978). However, these plutons are not exposed at the surface so their detailed chemistry is not known. The few granitoid plutons which are exposed within the Hokuroku basin have K-Ar ages of between 9 and 10.3 Ma (Ohmoto et al., 1983b) and are correlated to the Onnagawa stage.

It is also very difficult to sample unaltered acidic volcanic rocks in the district. Of 1400 bulk chemical analyses of the volcanics, grouped into horizons D1 and D2 (Onnagawa) and D3 and D4 (Nishikurosawa) (MITI, 1983), only 35 specimens have loss on ignition of less than 1.0 per cent. Since there are no petrographic descriptions available for these samples, the loss on ignition is the least ambiguous criterion to select "unaltered" volcanic rocks. Calculated C.I.P.W. Norms of these "unaltered" acidic rocks show a dependency on their stratigraphic position (Fig. 7). Dacites and rhyolites of the D3 and D4 horizons are classified as corundum normative or meta-aluminous, whereas those of D1 and D2 are slightly alkaline compared with the footwall acidic rocks. Therefore, it is likely that the acidic magma related to the kuroko mineralization was slightly aluminous and favorable for the generation of a metal-rich magmatic fluid. On the other hand, the alkaline nature of some of the post-ore acidic magma would have been disadvantageous for the production of magmatic hydrothermal deposits.

Chlorine in volcanic rocks and magma is readily lost during vapor loss from magma, solidification, and hydrothermal alteration (Anderson, 1974). Therefore, it is not easy to estimate the original chlorine content in magma from the analysis of volcanic rocks. The chlorine content of unaltered high-silica rhyolite breccia in a breccia dike in the Shigenai Formation (Onnagawa stage) at Akibayama, Odate city, is only 30 ppm, which is an order of magnitude lower than the average for rhyolite (Wedepohl, 1974). Unfortunately, there are no footwall rhyolites fresh enough to provide a reliable value for chlorine. However, two lines of indirect evidence suggest that the chlorine content of the pluton below the kuroko deposits was high. The first is the high salinity of fluid inclusions in minerals from the lower part of the stockwork ore (so-called Siliceous Ore) (Marutani and Takenouchi, 1978). The most plausible reason for this high chloride content is a magmatic contribution to the ore solution. Secondly, rhyolitic melt in contact with basaltic magma tends to be strongly enriched in chlorine (Anderson, 1974).

On the contrary, Pisutha-Arnond and Ohmoto (1983) proposed the idea of salinity increase due to seawater-rock interaction, with water being extracted from solution to form smectite at lower temperatures. However, such a saline water would have $\delta^{18}$O and $\deltaD$
compositions quite distinct from those estimated for the kuroko solution (Fig. 18 of Pisutha-Arnon and Ohmoto, 1983). Therefore, another fluid component is needed to explain the estimated range in composition of the kuroko solution through a mixing process (see Appendix). A hypothetical smectite-plagioclase rock R was adapted (Fig. 18 of Pisutha-Arnon and Ohmoto, 1983) as the candidate which is in equilibrium with such a solution. Obviously, the isotopic composition of the rock R ($\delta^{18}$O = 7 per mil and $\delta^D = -50$ per mil) coincides with that of acidic magma.

**Summary and Conclusion**

The conclusions of this study may be briefly stated.

1. Mineralization of the kuroko deposits was associated with a peak of acidic igneous activity during the opening of the Japan Sea around 15 Ma b.p.

2. Comparison with the volcanic succession in the Yellowstone caldera suggests that a molten acidic pluton existed at depth during the formation of the kuroko deposits.

3. The mode of eruption of submarine volcanics prior to kuroko mineralization is distinct from that after mineralization. Footwall rhyolite is characterized by lava flows and lithic tuff breccias, whereas the hanging-wall volcanics are dominated by pumiceous tuff. Such a change, which occurred over a short period of time throughout the Hokuroku district, was apparently the cause of the strata-bound nature of the kuroko deposits.

4. The kuroko horizon is not unique, in a strict sense, but occurred as multiple events over a few million years. The geologic phenomenon which triggered mineralization was a specific type of volcanic eruption which produced the lava and lithic tuff breccia. This event was followed by a gradual release of aqueous magmatic fluid. This fluid became an essential component of the kuroko ore solution.

5. The normative composition of the acidic magma shifted from meta-aluminous before the kuroko mineralization to alkaline afterward. Potentially high metal contents in the magmatic fluid released from the meta-aluminous magma may have been able to precipitate the kuroko deposits even after dilution by deeply circulated seawater.

6. A critical evaluation of wallrock alteration around kuroko deposits rules out the possibility that the Tertiary volcanic rocks were the source of the ore elements through a leaching process. The notable homogeneity of ore lead isotopic composition indicates that the lead could not have been extracted from basement rocks. Therefore, the hypothesis of leaching as a source of metals has many disadvantages.

7. This magmatic hydrothermal hypothesis of kuroko genesis is generally consistent with all observed geological and chemical features, and in particular explains the homogenous isotopic data and the relation of mineralization to a specific style and composition of volcanic activity.

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Kuroko Deposit Modeling Based on Magmatic Hydrothermal Theory


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Appendix

Fig. A-1. The hydrogen and oxygen isotopic compositions of kuroko ore solution. Those of seawater, meteoric water and magmatic fluid (Taylor, 1979) are also superimposed as star, arrow and dotted area, respectively. The square A denotes the range of isotopic composition of kuroko solution calculated from δD of sericite and δ18O of quartz using experimentally determined isotope fractionation factor (Hattori and Muehlenbachs, 1980). The square A' shows; calculated δ18O range based on the isotopic composition of quartz of period III (yellow ore stage) and measured range in δD value of fluid inclusions in quartz and sulfides, after Pisutha-Arnond and Ohmoto (1983). The square B represents the measured compositions of fluid inclusions by Pisutha-Arnond and Ohmoto (1983). They interpreted the results as the consequence of dilution of original kuroko solution by meteoric water (trapped as low-temperature secondary inclusions) after the area became subaerial. So they extrapolated the values to obtain primary solution which might have seawater salinity. A fairly large compositional range shown as square C was obtained through this procedure. If we assume that the isotope exchange equilibrium was established among fluid, sericite and quartz during the formation of kuroko deposits, the squares A and/or A' are the most reasonable as the compositional range of the primary kuroko ore solution. Apparently, these ranges are easily explained by mixing between seawater and magmatic fluid.

The line DF denotes estimated isotopic compositions of diagenetic fluid evolved from seawater by low temperature processes (Pisutha-Arnond and Ohmoto, 1983; Fig. 18). Note that the δD value of evolved seawater increases during water-rock interaction and seawater itself cannot be the sole source of ore solution. The line DS represents the isotopic composition of smectite in equilibrium with the diagenetic fluid, after Pisutha-Arnond and Ohmoto (1983). The composition of hypothetical rock R is also superimposed (see text for explanation).
マグマ性熱水鉱床説による黒鉱のモデル化

浦辺 徹郎

要旨：黒鉱鉱床は酸性マグマから放出された熱水を重要条件とする鉱床と生成した。言い換えれば、それが現在までに得られている地質学的、地球化学的データを最も良く説明することを明らかにした。

黒鉱鉱床は広域応力場の急変、流紋岩質火成活動のビーグ、急激な沈降といった重要な地質条件の推移期に生成した。鉱床生成前の火山噴火の様式を多くの黒鉱地帯で比較すると、下盤側に溶岩、発泡度の低い角礫凝灰岩、ペイアラクトアイトが見られるのに対し、上盤側の流紋岩質火山活動は大規模な火砕流を発生するものに変化しているが分る。この噴火様式の変化の起った時期は地域によりずれており、このため黒鉱の層準は厳密には同一ではない。

黒鉱鉱床の分布は広く見れば中新世の玄武岩活動の地域と一致していることが指摘されている。しかし細かく見ると黒鉱直下には玄武岩溶岩が見られない。このことはイエローストンカルデラにおける火成活動を想起させる。ここでは玄武岩の熱により地殻下部が融解し、流紋岩質プルトンが形成されている。といったごのようなマグマが発生すると、玄武岩鉱マグマは比重差のためその中に上昇できず、玄武岩活動はマグマ溜り周辺に限定される。この機構を黒鉱の場合に当てはめると、黒鉱生成中には地下に酸性マグマ溜りが存在しており、生成後それはいったん固結することが明らかである。というのも黒鉱の直上には、しばしば玄武岩溶岩が見られるからである。

更に上位の軽石火砕流はイエローストンの例にもあるようにリサージェント（再生プルトンの噴火によるものと考えられる。このような爆発的な噴火は、マグマ中の水溶液成分を逸散させてしまうので、もはや熱水鉱床は形成され得なくなる。

母岩の変質のゾーニングその他の検討より、鉱床下盤火山岩層中では熱水は主として炭化の中心から外側へ向って流れたことが推定される。このことより、火山岩類より鉱床構成元素を溶脱してきた鉱床を作るというモデルは妥当ではない。一方鉱床中の鉱、ストロンチウム、イオウの同位体組成に共通する重要な特徴は、いずれも非常に均質であるということである。つまりそれらの元素の起源は、事前に混合された地質体中であると言え、種々地質的な条件を持つ基盤岩類からのものとは考えづらい。

黒鉱鉱床の水素・酸素同位体組成は、単純な海水・岩石反応によって説明し得ないものである。一方マグマ水の寄与を考え、鉱床がそれと海水との混合物であるとすれば説明できる。実際花崗岩モルトと水溶液間の金属元素の分配実験によると、モルトがアルミナ質で圧力が低ければ、水溶液中の金属元素濃度は非常に高くなることが示されている。黒鉱鉱床下盤の火成岩類はその条件を満たしており、このことからも黒鉱鉱床のマグマ性熱水起源説は間接的に支持される。