

Ore-Forming Fluids

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Ore-forming (hydrothermal) fluids, consisting largely of H₂O, CO₂, and NaCl, formed most of Earth's ore deposits. The fluids exist as largely unconfined systems in meteoric, seawater, and basinal settings, or locally and intermittently confined systems in magmatic, metamorphic, and basinal settings, and they are driven largely by differences in temperature, elevation or density. Temperatures are highest (~600°C) in magmatic and lowest in basinal and meteoric (~100°C) systems. Salinities well above that of seawater are reached by boiling, evaporation, and evaporite dissolution, largely in magmatic and basinal systems. Today, research is focused on establishing the concentrations of metals in these fluids, the volume and duration of hydrothermal flow, and the links between ore systems and larger, regional fluid systems.

KEYWORDS: ore deposit, hydrothermal fluid, fluid, water

INTRODUCTION

Ore deposits are formed by geologic processes ranging from weathering to magmatic crystallization, but most form from hot, aqueous fluids. These hydrothermal fluids are the main agents for the movement of thermal energy and mass transfer in the crust. The wide variety of textures in hydrothermal ores (Fig. 1) provides evidence of the many settings in which hydrothermal fluids operate.

ORE-FORMING FLUIDS

Temperature and Composition

The composition and physical state of ore-forming fluids can be inferred from studies of active systems, such as the Au–As deposits of the Waiotapu geothermal system in New Zealand and the Cu–Zn–Pb “black smokers” along the mid-ocean ridges (Von Damm et al. 1997; Brown and Simmons 2003). Additional information can be obtained from extinct hydrothermal systems exposed in ore deposits, where minerals that fill open spaces or that replaced and altered wallrocks reflect the composition of their parent hydrothermal fluid (Barnes 1997). Fluid inclusions are especially useful sources of information on extinct hydrothermal systems because they are microsamples of these ancient fluids (Roedder and Bodnar 1997). Daughter minerals in some inclusions provide evidence of salinities and compositions of the paleofluids, and inclusions consisting largely of vapor demonstrate that they boiled (Fig. 2). Heating and freezing measurements on fluid inclusions indicate the temperature at which the fluid was trapped

and its salinity, and analyses of inclusion contents provide information on compositions of the fluids. Isotopic analyses of hydrothermal minerals and inclusion fluids serve as geothermometers and tracers of the source of the fluid and its dissolved constituents (Farmer and DePaulo 1997; Taylor 1997; Ohmoto and Goldhaber 1997).

Active hydrothermal systems near the present surface operate at temperatures of less than about 350°C; on the other hand, ancient systems exposed in ore deposits reached at least 600°C. This large difference reflects the deeper magmatic, basinal, and metamorphic settings of some ore deposits, which produced higher temperatures and salinities. For instance, near the surface the only widespread saline water is seawater with a salinity of about 3 wt%, and evaporation is the only common process that increases salinity. At depth, however, salinity can be increased by dissolution of evaporites, boiling, phase separation, and hydration of wallrock (Bodnar et al. 1985; Hanor 1987; Yardley and Graham 2002).

Inclusion fluids in ore deposits consist largely of H₂O with locally abundant CO₂ and smaller amounts of H₂S, CH₄, and N₂. Dissolved cations include Na, K, Ca, Mg, Fe, and Si, and the dominant anion is Cl, with smaller amounts of HCO₃⁻ and SO₄²⁻ (Roedder and Bodnar 1997). Less information is available on concentrations of ore elements such as Cu, Zn, Pb, and Au in inclusion fluids. Early efforts comparing the volume of metal-bearing daughter minerals to that of their host fluid inclusions indicated concentrations of thousands of parts per million for Cu in inclusion fluids from porphyry Cu deposits, and more recent analyses of single fluid inclusions confirm these high concentrations (Sawkins and Scherckenbach 1981; Heinrich et al. 1999; Vanko et al. 2001; Baker et al. 2004).

Experimental studies show that ore metals are carried in hydrothermal fluids as complex ions (Wood and Samson 1998). Most metals are cations, and the most common anions in these complexes are Cl⁻ for base metals and HS⁻ for Au. The generally lower concentrations of organic anions and lower stabilities of organic complexes limit them to special roles such as the transport of mercury by oil (Fein and Williams-Jones 1997).

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State of Ore-Forming Fluids

Boiling is a common feature of active geothermal systems and is clearly indicated by co-existing liquid- and vapor-rich inclusions in ore deposits. Formation of a vapor phase has a strong impact on the hydrothermal system. Expansion of the newly formed vapor can open fractures to create new porosity, but this can also block porosity creating new flow patterns (Burnham 1997; Hayba and Ingebritsen 1997). Boiling is also important to ore deposition because CO_2 and H_2S are partitioned into the vapor causing deposition of metal from some chloride complexes and destabilization of bisulfide complexes such as $\text{Au}(\text{HS})_2^-$ (Drummond and Ohmoto 1985).

Efforts to delineate regions of the crust in which vapors could form are commonly based on phase equilibria in the $\text{H}_2\text{O}-\text{CO}_2$ and $\text{H}_2\text{O}-\text{NaCl}$ systems, in which vapor and liquid coexist in very different P-T ranges (Fig. 2C). In the $\text{H}_2\text{O}-\text{CO}_2$ -NaCl system, vapor-liquid conditions persist to pressures typical of deep crustal levels (Schmidt and Bodnar 2000). Additions of small amounts of K, Ca, and other dissolved solids cause relatively small changes in the $\text{H}_2\text{O}-\text{NaCl}$ system, but small amounts of CH_4 and other non-condensable gases greatly expand the two-phase region in the $\text{H}_2\text{O}-\text{CO}_2$ system (Hanor 1980; Roedder and Bodnar 1997).

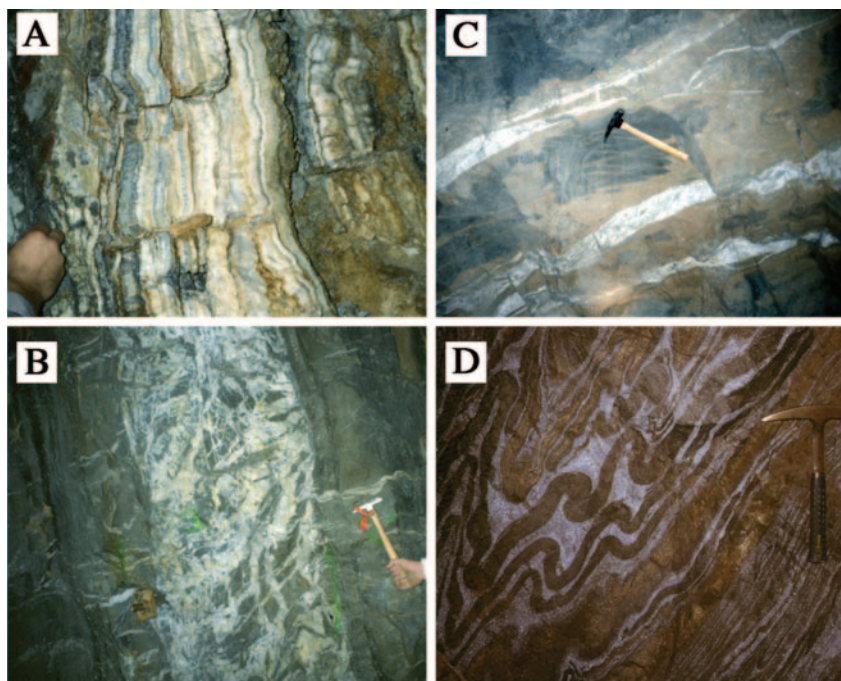


FIGURE 1 Minerals deposited from ore-forming fluids. **A.** Crustiform quartz-adularia vein, Hishikari epithermal gold deposit, Japan. **B.** Near-vertical, massive quartz-carbonate vein cut by smaller horizontal quartz veins indicating over-pressured fluid, Pamour greenstone gold deposit, Canada. **C.** Massive quartz vein with Fe-rich carbonate wallrock alteration, Mt. Charlotte greenstone gold deposit, Australia. **D.** Folded sedimentary layers of galena (grey) and sphalerite (brown), Sullivan sedimentary exhalative (sedex) Pb-Zn deposit, Canada.

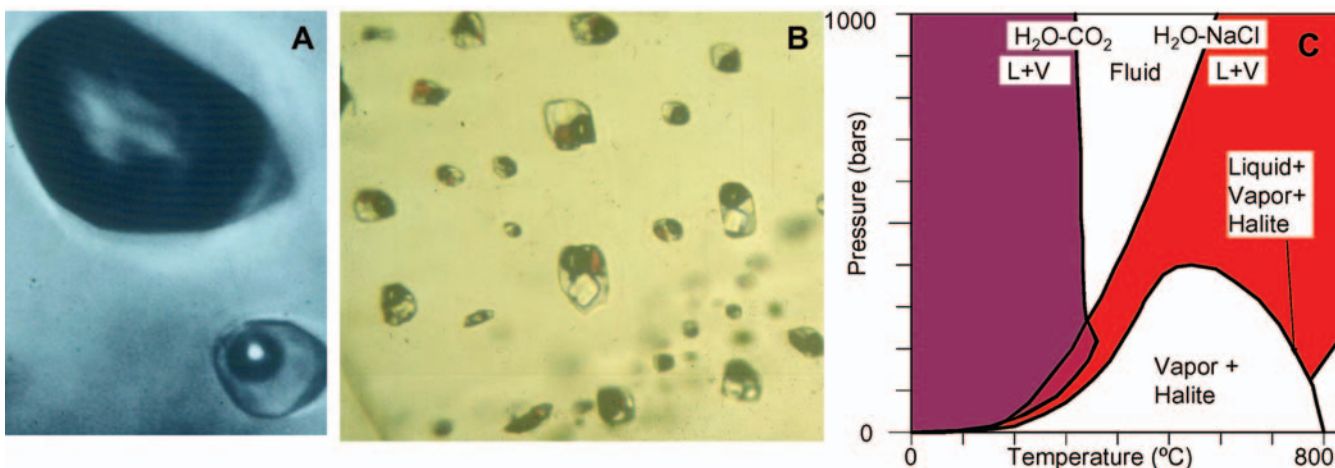


FIGURE 2 **A.** Coexisting liquid-rich and vapor-rich inclusions, Naica Pb-Zn-Ag carbonate-replacement deposit, Mexico. **B.** High-salinity liquid-rich inclusions, Granisle porphyry Cu-Au deposit, Canada. **C.** Critical curves showing the general location of two-phase fields for $\text{H}_2\text{O}-\text{NaCl}$ (red) and $\text{H}_2\text{O}-\text{CO}_2$ (purple) fluids depending on the fraction of NaCl or CO_2 in the fluid (generalized from Bodnar et al. 1985; Diamond 1994).

Circulation

Although hydrothermal flow is a large-scale, pervasive process, the fact that hydraulic conductivity of rocks varies over 13 orders of magnitude means that most fluids will move through highly permeable zones (Cathles 1997). Without this asymmetry of flow, there would probably be no ore deposits.

Hydrothermal systems can be divided into two main groups similar to those applied to groundwater. *Unconfined systems* commonly operate near hydrostatic pressure and are driven largely by topographically high recharge or differences in temperature or density of the fluid (Fig. 3). *Confined systems*, are largely isolated from the surface and are driven by magmatic or metamorphic fluids or by tectonic or sedimentary processes that isolate fluid reservoirs. When the fluid pressure in confined systems exceeds confining pressure, or when faults breach the reservoir, fluid is

expelled into surrounding unconfined zones (Sibson and Rowland 2003; Blundell et al. 2003). Unconfined systems usually form in cool, brittle rocks, whereas confined systems form in hot or ductile rocks.

The lifetime of hydrothermal systems is controlled by the factor that drives flow, the availability of fluid, and porosity. Unconfined and confined systems differ in the scale of fluid that they can release because unconfined systems can be recharged by external fluids, whereas confined systems can release only the fluid that they contain. Porosity is usually filled by quartz, calcite, anhydrite or related minerals,

reflecting the relatively high concentrations of major elements in hydrothermal fluids. Whereas quartz has prograde solubility at low temperature, calcite and anhydrite have retrograde solubility under most hydrothermal conditions (Fournier 1983a,b; Rimstidt 1997). Thus, calcite and anhydrite might deposit in the downwelling, recharge limb of unconfined systems where temperature increases, and quartz would deposit in the upwelling, outflow zone where temperature decreases (Cathles 1983) (**Fig. 3**).

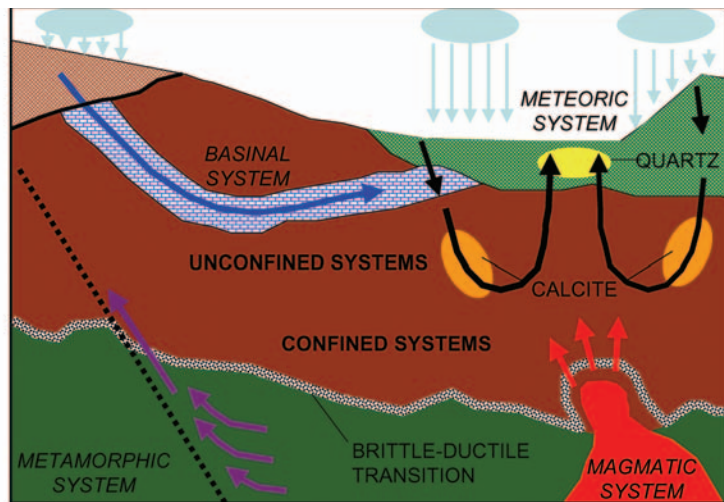


FIGURE 3 Typical hydrothermal systems. Confined magmatic and metamorphic systems are shown below a brittle-ductile transition and a partly confined aquifer is shown in the basinal system. In the unconfined meteoric system, quartz is deposited from cooling water flowing out the top of the system and calcite is deposited from heating recharge water flowing into the sides of the system. Similar flow and depositional patterns would be observed in unconfined seawater hydrothermal systems.

GEOLOGIC SETTING

Ore-forming hydrothermal systems are common in meteoric, seawater, basinal, magmatic, and metamorphic environments where water is abundant and a process is available to drive fluid flow. Although these systems can be described separately, many geological environments are transitional, and waters from different systems and sources can mix. In addition, changing geological and tectonic patterns through Earth's history have produced parallel changes in hydrothermal environments through time (Barley and Groves, 1992).

Meteoric hydrothermal systems form in the upper kilometer or so of subaerial crust where precipitation percolates downward through brittle rock, is heated, and rises along more focused zones (Fig. 3). Where these unconfined systems reach the surface, they form thermal springs with deposits of carbonate travertine or silica sinter. Many parts of meteoric flow systems can form ore. Downward moving recharge waters can oxidize sulfide ore deposits forming oxide Cu and Zn deposits and enriched Cu and Ag deposits (Chavez 2000; Hitzman et al. 2003). Slightly deeper recharge waters form roll-front uranium deposits (Nash et al. 1981). The hottest meteoric systems in volcanic terranes, which are heated by magmatic activity, are associated with most active geothermal systems, as well as epithermal Au-Ag deposits of the low-sulfidation type (Cooke and Simmons 2000). Fluids in these systems are nearly neutral, with relatively low S contents, and have temperatures of about 200 to 350°C and salinities of less than about 3 wt% NaCl. H and O isotope analyses of waters from these systems show H-isotope compositions similar to that of prob-

able meteoric recharge water and an "oxygen shift", with O-isotope compositions that are greatly changed by reaction with O-rich and H-poor wallrocks (Taylor 1997) (**Fig. 4**).

Seawater hydrothermal systems form in submarine crust, usually along mid-ocean and back-arc ridges, but also along faults associated with calderas and rift basins (Ohmoto and Skinner 1983; Scott 1997; Roberts et al. 2003). Ridge and caldera systems are hosted largely by volcanic rocks and form volcanic-hosted massive sulfide (VMS) deposits enriched in Fe, Cu, and Zn, whereas rift margin systems are hosted by sedimentary rocks and form sedimentary exhalative (sedex) deposits enriched in Zn and Pb. Seawater hydrothermal systems are largely unconfined and are driven by mafic magma chambers, dikes, and sills (Cathles 1983). Active "black smokers" and ancient VMS fluids have temperatures of about 200 to almost 400°C and salinities close to seawater, whereas limited data from sedex ore deposits suggest lower temperatures and possibly higher salinities (Fig. 4). The temperature difference reflects the smaller volumes of igneous rock driving sedex systems, but the cause of their higher salinities is not clear. VMS fluids are relatively acid and carry H₂S and metals together. Some sedex fluids have similar compositions, but other, usually cooler, sedex fluids contain dissolved sulfate rather than H₂S, and deposit sulfides where this sulfate is reduced or where the fluids vent into anoxic basins containing reduced sulfur (Goodfellow et al. 1993; Scott 1997; Cooke et al. 2000). In theory, these systems should form chimneys and mounds where the venting fluid is less dense than seawater, and lens-shaped ponds where it is more dense, although the high salinities necessary to produce bottom-seeking brines at these elevated temperatures might require special settings such as evaporite-contaminated rifts (Scott 1997; Solomon and Quesada 2003).

Basinal hydrothermal systems include connate water, as well as water from diagenetic reactions and meteoric recharge (Kharaka and Hanor 2003). Hydrocarbon fluids are also important parts of basinal systems, but usually play a minor role in the formation of ores. Sedimentary basins form unconfined systems driven by near-surface evaporative brines or topographically high recharge, and confined systems where reservoirs are isolated by burial or deformation (Cathles and Smith 1983; Garven 1985; Bethke 1986; Jones et al. 2002). The most common ores formed by basinal fluids are Mississippi Valley-type (MVT) deposits, which contain Zn and Pb (Leach and Sangster 1993). MVT fluids have temperatures of 100 to 200°C and salinities of 10 to 30 wt%, which result from evaporation and dissolution of evaporites (Kesler et al. 1995). Their pH is not adequate to permit transport of metals and reduced S together, requiring reduction of sulfate in the fluid or mixing with a second fluid to deposit sulfides (Sverjensky 1984; Anderson and Garven 1987). Generally similar basinal fluids with different metal contents form unconformity-type uranium and Kupferschiefer-type Cu deposits (Kotzer and Kyser 1992; Blundell et al. 2003).

Magmatic hydrothermal systems consisting of fluids released from crystallizing magma are most common around cupolas or other shallow felsic intrusions (Holland 1972; Burnham 1997). The systems are probably supplied in part by an underlying magma chamber, are confined by a ductile zone along the margin of the magma, and are surrounded by an unconfined hydrothermal system containing water of meteoric or seawater origin (Norton 1978; Shinohara et al. 1995; Fournier 1999). Abrupt release of magmatic fluid results in highly fractured, stockwork zones that host porphyry-type Cu, Mo, and Au deposits (Gustafson and Hunt 1975; Carten et al. 1988; Muntean

and Einaudi 2001). Skarn deposits form where magmatic fluid intersects carbonate rocks. Fluid inclusions in many of these deposits homogenize at 350 to 700°C, have salinities of 10 to 70 wt%, and were saturated with halite locally (Wilson et al. 1980; Bodnar and Beane 1980; Reynolds and Beane 1985; Landtwing et al. 2002; Redmond et al. 2004). Vapor, which forms abundant fluid inclusions in most high-temperature porphyry copper deposits, can be expelled into the surrounding meteoric system (Henley and McNabb 1978). High-sulfidation epithermal Au–Ag deposits, which form at temperatures of about 200 to 300°C from S-rich, acid fluids, are thought to result from this process (Vennemann et al. 1993; Hedenquist et al. 1998).

Metamorphic hydrothermal systems result from expulsion of water and other fluids from rocks that undergo prograde metamorphism, usually in tectonically stacked zones or subduction zones (Peacock 1989; Ferry 1995; Breeding and Ague 2002). These fluids consist of mixtures of H₂O and CO₂, the most common fluids released during metamor-

phic reactions (Powell et al. 1991). Metamorphic hydrothermal systems are largely confined below the brittle-ductile transition and release fluid only when they exceed confining pressure (Sibson et al. 1988). Greenstone or mesothermal Au deposits, which consist of quartz veins in strongly carbonatized wallrock, probably formed from metamorphic hydrothermal fluids with variable contributions of meteoric and magmatic fluid (Burrows et al. 1986; Nesbitt 1988). Fluid inclusions in these deposits homogenize at temperatures of 250 to 400°C and have a wide range of CO₂ contents, which reflect phase separation related to episodic release of overpressured fluid, as indicated by cross-cutting horizontal and vertical veins (Fig. 1) (Robert and Kelly 1987; Ridley and Diamond 2000).

Mystery hydrothermal systems, which derive fluids from as yet poorly understood sources, include those that formed the important Carlin-type Au and Fe-oxide–Cu–Au deposits. Carlin-type deposits formed from acid, low-salinity, 150 to 250°C fluids containing abundant reduced S. Different geochemical and isotopic tracers support magmatic, basalinal, meteoric, and even metamorphic origins for these fluids (Hofstra and Cline 2000). Iron-oxide–Cu–Au deposits, which form at temperatures of 200 to at least 500°C from high-salinity, oxidized fluids that produce regional sodic alteration, have been suggested to result from systems of basalinal or magmatic origin (Barton and Johnson 1996; Borrok et al. 1998; Williams et al. 2001).

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RESEARCH CHALLENGES

Ore deposits are the high-concentration end of a spectrum of enriched rocks, including distal trace-element anomalies such as the S-rich coals of the mid-continent U.S. and the As-rich waters and soils of northern Taiwan (Goldhaber et

al. 2003). Our research challenge in the coming century is to link ore deposits to their more distal kin. At both the deposit and regional scale, this requires better knowledge of exactly where the ore element resides in terms of the complex history of multiple fluid events that characterize most systems. It cannot be emphasized too strongly that knowledge of the mineralogical setting of ore elements is the key to all geochemical research. Recent recognition of gold nanoparticles in Carlin-type deposits provides an example of the type of surprises that will result from further high-resolution imaging of ores and rocks (Palenik et al. 2004).

We must also learn more about the fluids that formed deposits and that moved through surrounding rocks, particularly by means of microanalysis of minerals and fluid inclusions. Preliminary results using these methods suggest that low-level trace-element contents can be used to identify specific growth zones in minerals such as arsenian pyrite (Reich et al. 2003), and that elements such as Cu, As, and Au are partitioned differently into vapor or liquid during

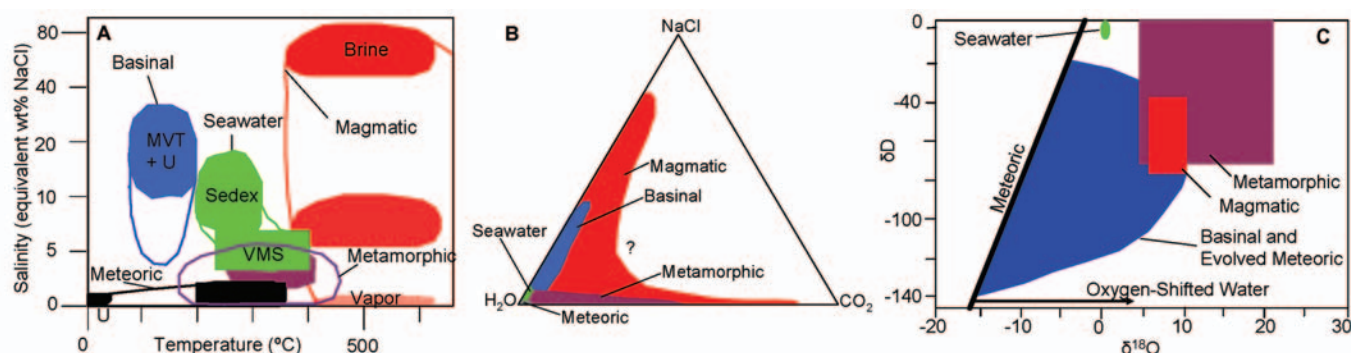


FIGURE 4 Chemical compositions of typical hydrothermal fluids grouped according to type of hydrothermal system and deposit, generalized from references cited in text. **A.** Temperature-salinity (dark zones inside lines show approximate T-salinity range for ore-forming fluids; two red fields in magmatic fluid zone represent brine and vapor in boiling systems). **B.** NaCl–H₂O–CO₂ (meteoric systems form a field around the H₂O apex that is too small to show clearly). **C.** δD–δ¹⁸O. Deposit types are explained further in the text (Sedex = sedimentary exhalative; VMS = volcanogenic massive sulfide; MVT = Mississippi Valley-type; U = Rollfront uranium).

boiling (Heinrich et al. 1999; Vanko et al. 2001; Baker et al. 2004). Parallel experimental studies are required to define phase equilibria in relevant systems, ore metal solubilities, and activity models in complex fluids with multiple ligands, all of which are needed for the next generation of coupled reaction-flow modeling of hydrothermal systems (Garven and Raffensperger 1997). Finally, we need quantitative information on the age and duration of hydrothermal systems (Cathles 1997), and on the source of ore metals and the fluids that carry them.

At some point in the 21st century, these efforts will result in a new generation of geological maps and sections that delineate crustal hydrothermal systems, just as today's maps delineate rock units. These maps will be the result of careful study of ores and rocks aided by satellite, geophysical, and other remote observations. They will help us understand the role of crustal fluids in the complex geochemistry of Earth's crust and will be the basis for new efforts in fields ranging from medical geology to the search for deeply buried ore deposits that will supply future generations. ■

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