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Sulfur-poor intense acid hydrothermal alteration: A distinctive hydrothermal environment



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ABSTRACT

A fundamentally distinct, sulfide-poor variant of intense acid (advanced argillic) alteration occurs at the highest structural levels in iron oxide-rich hydrothermal systems. Understanding the mineralogy, and geochemical conditions of formation in these sulfide-poor mineral assemblages have both genetic and environmental implications. New field observations and compilation of global occurrences of low-sulfur advanced argillic alteration demonstrates that in common with the sulfide-rich variants of advanced argillic alteration, sulfide-poor examples exhibit nearly complete removal of alkalis, leaving a residuum of aluminum-silicate + quartz. In contrast, the sulfur-poor variants lack the abundant pyrite ± other sulfides, hypogene alunite, Al-leached rocks (residual “vuggy” quartz) as well as the Au-Cu-Ag ± As-rich mineralization of some sulfur-rich occurrences. Associated mineralization is dominated by magnetite and/or hematite with accessory elements such as Cu, Au, REE, and P. These observations presented here indicate there must be distinct geologic processes that result in the formation of low-sulfur advanced argillic styles of alteration.

Hydrolysis of magmatic SO₂ to sulfuric acid is the most commonly recognized mechanism for generating hypogene advanced argillic alteration, but is not requisite for its formation. Low sulfur iron-oxide copper-gold systems are known to contain abundant acid-styles of alteration (e.g. sericitic, chloritic), which locally reaches advanced argillic assemblages. A compilation of mapping in four districts in northern Chile and reconnaissance observations elsewhere show systematic zoning from near surface low-sulfide advanced argillic alteration through chlorite-sericite-albite and locally potassic alteration. The latter is commonly associated with specular hematite-chalcocopyrite mineralization. Present at deeper structural levels are higher-temperature styles of sodic-calcic (oligoclase/scapolite – actinolite) alteration associated with magnetite ± chalcocopyrite mineralization. These patterns are in contrast to the more sulfur-rich examples which generally zone to higher pyrite and locally alunite-bearing alteration.

Fluid inclusion evidence from the systems in northern Chile shows that many fluids contain 25 to >50 wt% NaCl_{eq} with appreciable Ca, Fe, and K contents with trapping temperatures >300 °C. These geological and geochemical observations are consistent with the origin of the low-sulfur advanced argillic assemblages from HCl generated by precipitation of iron oxides from iron chloride complexes from a high-salinity fluid by reactions such as $3\text{FeCl}_2 + 4\text{H}_2\text{O} = \text{Fe}_3\text{O}_4 + 6\text{HCl} + \text{H}_2$. Such HCl-rich (and relatively HSO₄⁻-poor) fluids can then account for the intense acid, Al-silicate-rich styles of alteration observed at high levels in some iron-oxide-copper-gold (IOCG) systems. The geochemical differences between the presence of sulfide-rich and sulfur-poor examples of advanced argillic alteration are important to distinguishing between system types and the acid-producing capacity of the system, including in the modern weathering environment. They have fundamental implications for effective mineral exploration in low-sulfur systems and provide yet another vector of exposed alteration in the enigmatic IOCG clan of mineral deposits. Furthermore, understanding the geochemistry and mineralogy of this distinct geologic environment has applications to understanding the acid generating capacity and deleterious heavy metals associated with advanced argillic alteration.

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1. Introduction

Intense acid (advanced argillic; Meyer and Hemley, 1967) alteration is widespread in the uppermost parts (<1 km) of calc-alkaline volcanic arcs and related hydrothermal systems, where it is commonly sulfur-rich (abundant pyrite, alunite) and contains a distinctive suite of elements of economic interest (Au, Cu, Ag) and environmental concern (As, Hg, S; Arribas, 1995; Plumlee, 1999; Simmons et al., 2005). In contrast, similar intense acid alteration assemblages form in the near surface, overlying sulfur-poor iron-oxide (Cu-Au-Co-U-REE) systems (Barton and Johnson, 2000; Kreiner, 2011; Barton, 2014). Advanced argillic assemblages associated with low-sulfide systems share many of the same characteristics to the more common variants found in sulfur-rich settings, such as an assemblage characterized by Al-Si phases (e.g. kaolinite, pyrophyllite, andalusite, dumortierite), however they are distinct in the scarcity of sulfide and sulfate minerals (Kreiner and Barton, 2011).

Early work on advanced argillic alteration has been on sulfide-rich systems (e.g. Meyer and Hemley, 1967; Hemley et al., 1980) and recent work has focused on the interpretation and understanding of these sulfur-rich examples (e.g. White, 1991; Hedenquist et al., 1998; Chang et al., 2011; Hedenquist and Taran, 2013). A smattering of examples of sulfide-poor alteration have been described previously without genetic interpretation (e.g., Sykes and Moody, 1978; Kerr, 1998; Ray and Dick, 2002; Zhou et al., 2007; Zhang et al., 2011). Barton and Johnson (1996, 2000) noted the sparse occurrence of S-poor advanced argillic alteration assemblages in IOCG systems worldwide, where they were inferred to be the S-poor equivalents of what occurs at high structural levels in porphyry copper deposits (commonly referred to as lithocaps, e.g. Sillitoe, 1995).

This study grows from broader investigations in the western USA and northern Chile examining the relationships between IOCG mineralization and their broader hydrothermal context (Barton et al., 2011; Kreiner, 2011). Seventeen examples of low-sulfur, advanced argillic alteration related to IOCG-style systems in both North and South America are reviewed here. Our study presents new geologic and geochemical observations of the low-sulfide advanced argillic IOCG occurrences and details their association with structurally deeper iron-oxide (magnetite or hematite) rich, Cu(\pm Au)-bearing styles of mineralization. These observations provide a context to understanding the zoning relationships of high-level portions of the systems to associated underlying Cu-bearing, iron-oxide rich mineralization, and deeper and distal metal-depleted feldspar-stable alteration. We then discuss the similarities and differences between the high- and low-sulfide styles of advanced argillic alteration, including comparisons of the various styles of low-sulfide advanced argillic alteration and the genetic implications. The vertical zoning patterns observed around the low-sulfide advanced argillic alteration in the Cordillera of North and South America are similar despite different geologic environments (e.g. magmas, tectonic settings, compositions of host rocks), which require a genesis independent of these features (Barton et al., 2013). Distinct, non-sulfur-bearing sources of acid (e.g. dissociation of HCl resulting from the precipitation of metals in a brine-dominated system) are required to generate the IOCG, and perhaps other styles, of low-sulfur assemblages (Kreiner, 2011).

2. Diversity of advanced argillic alteration

Meyer and Hemley (1967) defined advanced argillic alteration as replacement of feldspars and micas by one or more of the aluminum-silicate minerals, andalusite, pyrophyllite, or kaolinite.

The Al-Si phases indicate a more acid assemblage at any given temperature, when compared to feldspar or muscovite bearing assemblages in the K_2O - Al_2O_3 - H_2O - SiO_2 system. Associated minerals commonly include alunite, pyrite with accessory rutile, Al-P-S phases, dumortierite, zunyite, lazulite, and hematite.

Most studies have focused on volcanic (fumarolic)-related advanced argillic alteration and the locally associated Cu-Au-rich epithermal and porphyry deposits (Rye et al., 1992; Arribas, 1995; Hedenquist et al., 2000). In such settings, acidic alteration is characterized by abundant pyrite and alunite, and is most common at shallow depths. Advanced argillic styles of alteration form in most near-surface hydrothermal environments over a broad range of temperatures, sulfidation states, total sulfur contents, oxidation states, and silica saturation (Hemley et al., 1980). Thus, a single origin for these intense acid alteration assemblages should not be expected.

Advanced argillic alteration in many varieties of porphyry and epithermal systems, volcanogenic massive sulfide systems, and other near-surface environments (e.g. crater lakes) is commonly characterized by abundant sulfur-bearing minerals, which implies an origin from sulfuric acid-rich fluids which may result from the condensation of low-density SO_2 -rich fluids (Einaudi et al., 2003; Heinrich, 2005; Seedorff et al., 2005; Hedenquist and Taran, 2013). Systems with sulfur-rich styles of advanced argillic alteration may be associated with Cu, Au, and Ag, and other metals such as As, Hg, Bi, and Sb that readily form sulfide-complexes (Arribas, 1995; Simmons et al., 2005; Seedorff et al., 2005) and are commonly deposited in genetically-related, but often later mineralizing events associated with higher-density S-bearing fluids (e.g. Heinrich, 2005; Hedenquist and Taran, 2013). These systems show predictable zoning patterns to associated mineralization, and generally, genetically-associated tectonic or igneous processes related to the magmatic-hydrothermal systems.

The occurrence of advanced argillic alteration associated with iron oxide-bearing alteration and low total sulfide implies a distinct geologic environment that has a sulfide-poor source of acid, yielding hematite or magnetite instead of pyrite and alunite. The low-sulfur variants are genetically and spatially related to underlying iron-oxide dominated mineralization associated with low sulfidation states and total sulfur (Kreiner and Barton, 2011; Barton, 2014). Characteristic of the low-sulfur variety are metals (Cu, Ag, Au), and other elements that readily form chloride complexes but a scarcity of those more closely linked to sulfide complexing (Hitzman et al., 1992; Barton and Johnson, 1996; Barton, 2014). Low-sulfide styles of advanced argillic alteration are commonly observed in the upper levels of iron oxide(-copper-gold) (= "IOCG") systems (Barton and Johnson, 2000; Barton, 2014).

3. Sulfide-poor advanced argillic alteration

Advanced argillic alteration with low total sulfur, unrelated to weathering, has occurred globally at several locations. This study has documented 17 localities in northern Chile and the western USA (Table 1) through detailed and reconnaissance observations and geochemical analysis. Although earlier studies have reported Fe-oxide-stable, low-sulfide advanced argillic alteration (Table 1), none have described the occurrences in detail nor have they addressed the genetic implications of the distinctive mineralogy.

3.1. Mineralogy

Low sulfide advanced argillic alteration is characterized by Al-Si phases including kaolinite, pyrophyllite, andalusite, diasporite, dumortierite and corundum (Figs. 1 and 2; Table 1). Specular hematite (\pm magnetite) are present in these low-sulfide examples,

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