

Constraints on the volume of sulfide melt generated through metamorphism of ore deposits

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SUMMARY

Phase relations between mineral assemblages in common sulfide ore deposit types are examined to place constraints on the amount of polymetallic melt produced within the limits of crustal metamorphism. Most sulfide deposit types have the potential to undergo minor melting at moderate temperatures, provided that sulfosalts or tellurides are present. Deposits that contain these phases may start to melt at conditions ranging from lowest greenschist facies to amphibolite facies, whereas those lacking sulfosalts and/or tellurides may only melt once P-T conditions are well into the granulite facies, particularly if galena is also absent. Epithermal gold deposits would melt at the lowest temperatures of all sulfide deposit types. Massive Pb-Zn(-Cu) deposits may start to melt in the lower to middle amphibolite facies, but only if pyrite and arsenopyrite coexist at these conditions. Whereas massive Ni-Cu-PGE deposits can typically generate only trace quantities of melt. Similarly, disseminated Cu deposits, excepting sulfosalt-bearing occurrences, are incapable of generating melt until the granulite facies when melting commences in bornite-rich regions, in the system Cu-Fe-S. Even at granulite facies conditions, the amount of polymetallic melt that can be generated in most deposit types is <1 vol.%. The exception is massive Pb-Zn(-Cu) deposits, where melt volumes significantly exceeding 1 vol.% may be segregated into sulfide magma dykes, allowing mobilization over large distances.

Key words: Ore deposits, metamorphism, mobilisation, sulfide melting.

INTRODUCTION

At an increasing number of metamorphosed sulfide ore deposits around the world, it is being recognized that part of the ore mineral assemblage underwent partial melting during metamorphism (Hofmann, 1994; Mavrogenes *et al.* 2001; Stalder and Rozendaal, 2001; Tomkins and Mavrogenes, 2002; Tomkins *et al.* 2004; Sparks *et al.* 2005). The ore minerals in these deposits that melted include sulfides, sulfosalts (containing As or Sb and S), tellurides and native minerals (metallic minerals lacking S and Te). The metamorphic temperatures at these previously described deposits varied from ~520°C, at the Legenbach deposit in Switzerland (Hofmann, 1994), to as high as 850°C at the

Challenger deposit in Australia (Tomkins & Mavrogenes, 2002).

During high temperature metamorphism and deformation of sulfide ore bodies, sulfide melts are easily mobilized, leading to development of complex mineralogical-structural associations that are impossible to interpret correctly if sulfide melting is not recognized (Tomkins *et al.* 2004). Therefore, awareness that sulfide melting can occur facilitates a more accurate interpretation of ore body evolution, leading to better genetic models to be used in finding similar deposits. Understanding sulfide melt mobilization is also important to mine planning and near-mine exploration because segregation of even small amounts of sulfide magma can significantly redistribute ore metals, especially precious metals, in and around pre-existing mineral deposits (Tomkins & Mavrogenes, 2002; Tomkins *et al.* 2004).

A number of important questions remain regarding sulfide melting that are yet to be comprehensively answered. (1) Under what metamorphic conditions do typical sulfide ore deposits start to melt? (2) Is this process capable of generating enough melt to be mobilized through sulfide magma dykes over hundreds of meters or kilometers? (3) Can this process form new ore deposits? (4) Can our understanding of this process help us find undiscovered ore deposits?

The aim of this study is to explore the initial melting relationships in the common types of sulfide mineral deposits, and in the process, gain an understanding of how much melt can be generated in various deposit types. This will allow us to address the bigger questions related to metamorphism and melting of sulfide ore deposits.

MASSIVE Pb-Zn(-Cu) DEPOSITS

The ore mineral assemblage in volcanogenic massive sulfide (VMS), Mississippi valley type (MVT), and sedimentary exhalative (SEDEX) type deposits is typically dominated by some combination of pyrite, pyrrhotite, sphalerite, galena and chalcopyrite. Stevens *et al.* (2005) estimated that the eutectic amongst these phases occurs between 700 to 730°C at 2 kbar. In the absence of chalcopyrite and pyrite, and any minor phases, melting may not occur until temperatures of ~800°C (at 5 kbar) are reached when galena + sphalerite + pyrrhotite start to melt (Mavrogenes *et al.*, 2001). However, some minor sulfide and sulfosalt phases do occur in many of these deposits and they may be critical to the low temperature generation of small quantities of melt.

In many occurrences, minor Sb-, Bi- and Ag-bearing sulfosalts appear to have formed during cooling, as these

elements exsolved from galena, with which they are commonly associated. There is a large extent of coupled substitution of $\text{Ag}^+\text{Sb}^{3+}$ and $\text{Ag}^+\text{Bi}^{3+}$ for 2Pb^{2+} in galena (e.g., Blackburn & Schwendeman, 1977). Given the typically low concentrations of Sb, Bi and Ag in these deposits relative to Pb (ores typically contain < 0.1 % Ag, Sb and Bi, but percent levels of Pb as galena), it is likely that all would substitute into galena during metamorphism without causing any melting. However, Sb-, Bi- and Ag-doped galena melts at lower temperatures than pure PbS, when combined with other sulfides (e.g., Mavrogenes *et al.* 2001).

One mineral that typically occurs in minor amounts in these deposits and that cannot form as an exsolved mineral during cooling is arsenopyrite. In contrast with Sb, Bi and Ag, there is no known solid solution of As in galena. Arsenopyrite can melt during metamorphism when high $f\text{S}_2$ conditions are generated through decomposition of pyrite. This melting reaction occurs at 491°C at 1 bar and ~560°C at 5 kbar (Clark, 1960; Sharp *et al.*, 1985). This and several other metamorphic processes that can also lead to formation of As-S melt from arsenopyrite breakdown have been discussed extensively by Tomkins *et al.* (in press). Because there is no substitution of As in galena, the presence of even small quantities of As-S melt causes partial melting of galena at temperatures as low as ~549°C (at 1 bar), the melting point of jordanite ($\text{Pb}_9\text{As}_4\text{S}_{15}$). Experiments by Roland (1968), conducted at pressures of up to 2 kbar, found that the melting point of jordanite was not affected by pressure within the uncertainty of measurement.

Other minor As-bearing sulfosalts may also melt at mid-amphibolite facies temperatures (e.g., Maske & Skinner, 1971), but in these deposits they are typically rare compared to arsenopyrite. At middle amphibolite facies conditions (ca. 550-700°C), interaction between As- S_{melt} and galena results in extensive melting of galena. For example, at 650°C (and 1 bar) 1 mole of As- S_{melt} will cause at least 3.8 moles of galena to melt. However, typical bulk compositions in massive sulfide deposits have a very large ratio of galena to arsenopyrite, so the As- S_{melt} would become saturated in PbS and most of the galena would not melt. Nevertheless, it is of interest to know how much melt might be generated in these deposits due to the presence of minor arsenopyrite.

I have derived an equation to calculate the amount of As-Pb-S melt present in any given galena-bearing rock (above 549°C) using the arsenic content from bulk rock analysis (based on the As_2S_3 -PbS phase diagram of Kutolglu, 1969):

$$\text{Vol.\%melt} = \left(\frac{D(\text{As} - 0.016 \text{As}(0.01T - 19))}{3.56 \times 10^7} \right) + \left(\frac{D \cdot X_{\text{As}} (3.92 \times 10^{-22} \cdot T^{7.5} + 3.72)}{316295} \right)$$

Where: As = As content of the rock (in ppm).
T = Temperature in °C.
D = Whole rock density (in kgm^{-3}).
 $X_{\text{As}} = \text{As} / 74.922$.

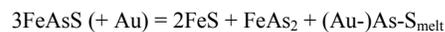
Based on the experiments of Roland (1968), the effect of pressure is likely to be minor. The density of the As-S component of the melt is assumed to be ~3560 kg m^{-3} and that of the Pb-S component to be ~7580 kg m^{-3} , based on the

density of realgar and galena respectively. The result represents the maximum amount of As-Pb- S_{melt} that can exist, assuming that all of the As in the rock forms a melt and that all As- S_{melt} can communicate chemically with excess galena. In many rocks these are not valid assumptions (see Tomkins *et al.* in press) and care should be taken when applying this formula. Although the As content of massive Pb-Zn deposits is typically low, a significant melt fraction can be generated if all of the As in the rock forms a melt. It should be noted that this calculation only considers As-bearing sulfosalts and galena, and the elements As, Pb and S. If other sulfides and sulfosalts contribute to the melt, or other elements partition into the melt from unmelted sulfides, then the melt ratio may actually be higher. Once melting between the major sulfides commences, the volume of melt in massive Pb-Zn sulfide deposits can substantially exceed 1 vol.%.

GOLD DEPOSITS: THE INFLUENCE OF SULFOSALTS

Gold deposits are extremely diverse in their ore mineral associations, which vary largely as a function of formation mechanism. Only alluvial gold deposits are unlikely to melt during metamorphism, simply because in these deposits, gold is not associated with any sulfides or sulfosalts that might depress the melting point of gold to lower temperatures. The gold deposit types that may start to melt during very low temperature metamorphism are those that contain sulfosalts, tellurides, and/or native bismuth or mercury (typically epithermal deposits as well as some orogenic gold deposits). There is a diverse range of minerals and mineral systems in these deposits that can melt at low temperatures, and many, if not all, of the resulting melts have the capacity to incorporate and mobilize gold and silver.

Gold deposits that contain only pyrite, pyrrhotite and arsenopyrite (i.e., many orogenic Au deposits) may not melt during metamorphism until higher temperatures are reached. Arsenopyrite + pyrite may melt at lower amphibolite facies conditions to form As-S melt + pyrrhotite, but only under favourable high $f\text{S}_2$ conditions (see Tomkins *et al.* in press). Generation of high $f\text{S}_2$ conditions during metamorphism is favoured by pyrite-rich rocks that maintain high $f\text{O}_2$ (rocks that are graphite-free, for example) and are low in Fe silicates and oxides. So, if the gold deposit is hosted in graphitic schist, or Fe-rich rocks such as BIF or dolerite, arsenopyrite may not melt until the following reaction occurs:



This reaction proceeds at ~770°C (at 5 kbar, $dP/dT = \sim 14^\circ\text{C/kbar}$; cf. Clark, 1960, Sharp *et al.*, 1985). It is possible that this reaction may also not proceed at low $f\text{S}_2$ conditions, as it is a desulfidation reaction and thus also governed by $f\text{S}_2$. So, it may be that many orogenic gold deposits would not melt even at granulite facies conditions.

As in all deposits, the extent of chemical communication between ore minerals in gold deposits affects melting. The disseminated nature of gold deposits means that there is little physical contact between ore minerals. However, many of the sulfosalts found in some gold deposits melt in isolation at low temperatures. Furthermore, arsenopyrite melting probably does not require physical contact between arsenopyrite and pyrite. The only purpose of pyrite in Reaction 1 is to liberate

sulfur, thereby causing an increase in sulfur fugacity (Tomkins *et al.* in press). Therefore, if enough pyrite is breaking down in the vicinity of an arsenopyrite crystal, a pervasive state of elevated f_{S_2} may be achieved, causing the arsenopyrite to melt. Once some sulfosalts have melted, mobilization caused by deformation will lead to physical interaction between melt and unmelted minerals, including gold, which can then be incorporated into the melt. This mobilization-assisted melting thus leads to generation of a larger proportion of melt than would otherwise be possible.

Because the ore minerals in gold deposits are disseminated, the total bulk sulfide content is low, so any melt that is generated during metamorphism is likely to comprise only a small proportion of the rock volume. In this case the proportional volume of melt can be appraised by conducting bulk rock analysis for As, Sb, Te, Tl, Pb, Cu, Hg, Bi, Au and Ag, the likely melt components, and estimating how much each element contributed to the melt (sulfur must also be taken into account). Clearly, these chemical systems can be very complex, so the result can only give a maximum (by assuming that 100% of each element contributed to the melt) and a best estimate. Bulk rock analysis of gold-rich samples from the Hemlo gold deposit in Canada (a particularly sulfosalt-rich deposit) indicates that the maximum amount of melt averaged over large volumes is <0.5%. However, observations of sulfosalt accumulations that formed during mobilisation of a sulfosalt melt (Tomkins *et al.* 2004) indicate that locally, the melt volume may exceed 25%.

MAGMATIC NI-CU-PGE DEPOSITS

Magmatic Ni-Cu-PGE deposits form by fractionation of sulfide melt from mafic to ultramafic silicate magma (e.g., Naldrett, 1989). When this fractionation process is protracted during cooling, the sulfide melt itself evolves small amounts of lower temperature melts, which are variably enriched in As, Cu, Sb, Pb, Sn, Bi, Te, Pd, Pt, Ag and Au (Li *et al.*, 1992; Gervilla *et al.*, 1997; Pritchard *et al.*, 2004). Although the bulk of the sulfide material in these deposits typically crystallizes at temperatures beyond the realm of normal crustal metamorphism, the highly fractionated low temperature melts may be subject to remelting, and thus mobilization, during amphibolite to granulite facies metamorphism.

The late fractionated component of the sulfide magma in these deposits dominantly crystallizes minerals such as arsenopyrite (FeAsS), gersdorffite (NiAsS), cobaltite (CoAsS), nickeline (NiAs), maucherite (Ni₁₁As₈), löllingite (FeAs₂), westerveldite ((Fe,Ni)As), safflorite ((Co,Fe)As₂), chalcopyrite (CuFeS₂) and cubanite (CuFe₂S₃) (Li *et al.*, 1992; Gervilla *et al.*, 1997; Pritchard *et al.*, 2004). At individual localities, this melt can be Cu-rich (e.g., Sudbury, Canada, Li *et al.*, 1992; Noril'sk, Russia, Naldrett *et al.*, 1997; Uruguayan dyke swarm, Pritchard *et al.*, 2004) or As-rich (e.g., Thompson, Canada, cf. Chen *et al.*, 1993; Carratraca (Spain) and Beni Bousera (Morocco) massifs, Gervilla *et al.*, 1996; Las Aguilas, Argentina, Gervilla *et al.*, 1997). A range of rare tellurides and sulfosalts occur as minor components that crystallize from these late stage fractionates. Of the main minerals in As-rich and Cu-rich fractionates, only the sulfarsenides (minerals with As and S) are likely to melt under normal metamorphic conditions (Clark, 1960; Craig & Kullerud, 1967; Sharp *et al.*, 1985; Singleton & Nash, 1986; Mackovicky *et al.*, 1992), although the others may melt during UHT metamorphism as

temperatures exceed ~850°C (Craig and Kullerud, 1967). Under these conditions a more volumetrically significant melt may develop.

Because many of these deposits contain little or no pyrite and are dominated by pyrrhotite, pyrite decomposition during metamorphism may not buffer f_{S_2} to the levels required to destabilize the sulfarsenides. With a very high ratio of pyrrhotite to pyrite, all of the pyrite will be consumed in maintaining pyrrhotite stoichiometry, which becomes more S-rich with increasing temperature along the Py-Po buffer, as well as in maintaining fluid equilibrium. In such a situation, the sulfarsenides are unlikely to melt during metamorphism through reactions that occur on the Py-Po buffer. However, mass balance calculations suggest that as long as Po:Py < 99:1 and the system is closed to external fluids, pyrite will be preserved to high enough temperatures for Reaction 1 to take place.

It is also possible that sulfarsenides in a pyrrhotite-rich massive Ni-Cu deposit could melt with increasing temperature and f_{S_2} through reactions within the pyrrhotite stability field. During metamorphism sulfur is liberated from pyrrhotite as temperature rises, leading to an increase in sulfur fugacity. This desulfidation produces a steep $\log f_{S_2}$ -T trajectory; a trajectory that in some cases may lead to arsenopyrite melting (see Tomkins *et al.* in press). Cobaltite and gersdorffite may also melt by the same process, although the f_{S_2} -T stability limits of these minerals is not known. This mechanism of sulfarsenide melting may not be applicable to other deposit types containing disseminated rather than massive pyrrhotite, due to processes that inhibit rising sulfur fugacity (Tomkins *et al.*, in press).

The amount of melt that can be derived from melting of the sulfarsenides is more difficult to ascertain through a simple bulk rock analysis, because much of the As also occurs in the arsenides (lacking S), which do not melt. However, most Ni-Cu deposits contain only very small amounts of sulfarsenide minerals (e.g., Chen *et al.*, 1993), so the maximum amount of melt that can be generated from this source is probably typically < 0.05%.

Several rare PGE-bearing sulfosalts and bismuthotellurides that crystallize from late fractionated segregations may start to melt at around the greenschist-amphibolite facies transition and others melt at granulite facies conditions or lower. Due to the rarity of these minerals, the proportion of melt produced by their decomposition is minuscule (probably typically < 0.01 % melt). The extent to which a trace melt, incorporating Pt, Pd, Au, Ag, Bi, Te, Sb, Pb and Sn, could flux the melting of the volumetrically dominant sulfides, sulfarsenides and arsenides is not well understood. Makovicky *et al.* (1992) showed that at 850°C in the system Pt-Fe-As-S, two relevant Pt-bearing melts exist; a Pt-As melt with 28-39 at% As and a Fe-As-Pt melt with up to 23.7 at% Pt, 31.7 at% As and 2.6 at% S. Thus, a complex PGE-bearing bismuthotelluride-sulfosalt melt could cause some (very minor) melting of the main phases at metamorphic conditions experienced in the crust.

DISSEMINATED Cu DEPOSITS

In general, when disseminated copper deposits (such as porphyry, IOCG, redbed and skarn deposits) first form, the ore

mineral assemblage is typically dominated by pyrite and chalcopyrite, and may contain appreciable amounts of bornite and molybdenite. Many minor sulfide minerals may also occur, particularly in some skarn deposits, including a range of Cu-sulfides, galena, sphalerite, gold, bismuth minerals (including native bismuth), tellurides and sulfosalts (particularly tetrahedrite-tennantite). Except in some skarn deposits, the sulfides are invariably disseminated and in most deposits, comprise only a small proportion of the bulk rock.

Ore minerals that melt at low temperatures (sulfosalts, tellurides and native Bi) tend to occur in Au-rich, Cu-poor varieties of these deposits. Examples include the Lucky Draw deposit in Australia (a Au-Bi-Te skarn; Sheppard *et al.*, 1995) and the Nico prospect in northern Canada (a Co-Au-Bi Fe-oxide deposit; Goad *et al.*, 2000). The melting relationships in the sulfosalt- and telluride-rich deposits have been described in the section on gold deposits. In deposits that lack sulfosalts, tellurides and/or native Bi melting may not commence until much higher temperatures are reached. During metamorphism copper sulfides, such as chalcocite (Cu₂S) and covellite (CuS), react with the typically coexisting pyrite to produce Cu-Fe-sulfide solid solutions (bornite_{ss} and ISS) and pyrrothite (Yund & Kullerud, 1966). In the absence of other phases, these assemblages will not melt until the granulite facies is reached. At temperatures near 800°C bornite + ISS or bornite + MSS start to melt (REF), and as such a significant fraction of the sulfide assemblage may melt. Again the volume of melt is dependant on the degree of chemical communication between the relevant phases. Given the relatively restricted amount of bornite in most deposits and the disseminated nature of most ores, the volume of melt that may be generated at granulite facies conditions is probably typically <1%.

Molybdenite, if present, may react with coexisting pyrite to produce a Mo-Fe-S melt at ~735°C (at 1 bar; Grover *et al.*, 1975), but this reaction can only proceed under the very high *f*S₂ conditions required for pyrite to exist at this temperature. Preservation of pyrite to such a high temperature requires oxidized host rocks that are low in Fe silicates and oxides, and that little hydrothermal fluid is generated by breakdown of phyllosilicates. Thus, melting of molybdenite + pyrite may be possible in parts of porphyry Cu deposits that are low in phyllosilicates, and some skarn deposits (molybdenite does not typically occur in IOCG or sediment-hosted stratiform Cu deposits). In this reaction pyrite and molybdenite need to be in physical contact for melting to proceed, a requirement that is not ubiquitously met due to the disseminated nature of these minerals in porphyry deposits and also skarn deposits. Even if pyrite did survive to high temperatures, the low proportion of molybdenite in these deposits (average grades as high as 0.45% MoS₂ is recorded in some deposits, but 0.1 % is more typical; Carten *et al.* 1993) and the poor chemical communication between the minerals implies that the volume of melt that can be generated through this reaction is likely to be low (< 0.1 %). An interesting complication in a metamorphosed porphyry system is that the hydrothermally altered silicate host rock would start to melt before the sulfides, so mobilization of silicate melt could promote mobilization of even trace quantities of sulfide melt (Tomkins and Mavrogenes, 2002; 2003)

CONCLUSIONS

The most important factor governing initiation of melting in a metamorphosed ore deposit is the composition of the original mineral assemblage. A deposit of any type with a significant proportion of sulfosalts is capable of generating a melt within the greenschist or amphibolite facies. In contrast, sulfide deposits lacking sulfosalts or tellurides, may not melt until the upper amphibolite or granulite facies, if at all. Deposits that most typically have appropriate mineral assemblages for low temperature melting are sulfosalt-rich epithermal gold deposits. Massive Pb-Zn deposits, typically being galena-rich and containing minor arsenopyrite, may start to melt in the middle to upper amphibolite facies. Magmatic Ni-Cu-PGE deposits generate only trace quantities during typical crustal metamorphism (UHT metamorphism is regarded as atypical). Disseminated Cu deposits with a significant proportion of bornite may generate melt quantities approaching 1 vol.% under granulite facies conditions. However, these deposits typically form in mountainous upper crustal positions and are thus rarely metamorphosed.

Although many types of sulfosalt-bearing gold deposits are capable of melting at greenschist or amphibolite facies conditions, large volumes of melt are unlikely to accumulate through mobilization. This is because the minerals in gold deposits that melt are invariably sparsely disseminated and the total volume of melt, when the deposit is considered as a whole, is relatively small. In addition, in deformed deposits the silicate host rock typically develops a dispersed array of micro- to meso-scale dilational structures during deformation, into which local aliquots of melt would migrate and then remain, thus precluding accumulation into through-going veins and dykes that drain the rock. Volumes of sulfide melt significant enough to coalesce into larger bodies of magma capable of being mobilized distances of hundreds of meters to kilometers may only be possible in highly metamorphosed massive Pb-Zn±Cu deposits. This conclusion is reached for two reasons. Firstly, only massive sulfide deposits are likely to contain enough sulfide material to form significantly more than 1 vol.% melt. Secondly, the eutectic amongst common sulfides is lowest in systems that contain galena, and too high in systems that do not. Where galena forms a significant proportion of the sulfide assemblage, larger bodies of sulfide magma are likely to form from massive Pb-Zn±Cu deposits where the metamorphic temperature exceeds about 800°C (at 5 kbar), although this critical temperature may be lower in relatively chalcopyrite-rich deposits.

Once these larger volumes of sulfide magma start to form into dykes, outliers of mineralization could start to form at significant distances from the source orebody. Theoretically, in granulite terrains this process could lead to formation of small new satellite orebodies around a source deposit. Once these outliers are identified as sulfide magma dykes sourced from elsewhere, they can potentially be traced by explorationists to the source if it remains unidentified. However, excepting the mechanism suggested by Tomkins & Mavrogenes (2003), where sparse amounts of sulfide melt segregate in concert with migmatitic silicate melt to form metal-rich magma, completely new orebodies are unlikely to form through melting of dispersed sulfide material that was not previously economic.

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