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ISOTOPIC CONSTRAINTS ON THE CLASSIFICATION OF THE JALES AU DISTRICT (NORTHERN PORTUGAL)

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KEY WORDS: gold, Variscan, intrusion-related, Portugal.

PALAVRAS-CHAVE: ouro, Varisca, relacionado com intrusões, Portugal.

ABSTRACT

The Jales Au district encompasses three areas with epigenetic mineralization of Variscan age. Mineralization is hosted in syn- to late-tectonic Variscan granites and in greenschist facies metasedimentary rocks, ranging in age from Cambrian to Lower Devonian. Sulfide paragenesis proceeds from early Fe-As sulfides to late base-metal sulfides. The deposits proximal to the intrusion (Campo and Gralheira) are relatively sulfide-rich and contain paragenetically late electrum associated with significant amounts of base-metal sulfides, whereas the deposit distal to the intrusion (Três Minas) is sulfide-poor and contains high-fineness Au that is paragenetically early. In spite of displaying characteristics related to orogenic Au deposits, the regional association with W-Sn deposits, close temporal relationship of mineralization with granitic intrusions, metal associations characterized by significant Bi contents or Bi-Au correlation, a paragenesis defining a low fO_2 -low fS_2 environment and S (and Pb) isotope studies suggest that the mineralization is related to syn- to late-tectonic reduced granites and could also be classified as intrusion-related.

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RESUMO: A classificação do distrito aurífero de Jales no quadro da geoquímica isotópica

O distrito aurífero de Jales inclui três áreas com mineralização epigenética de idade varisca. A mineralização está encaixada em granitos variscos sin- a tardi-tectônicos e rochas metasedimentares da fácies dos xistos verdes, com idades que vão do Câmbrio ao Devónico Inferior. A paragênese sulfuretada é constituída por sulfuretos de Fe-As precoces e sulfuretos de metais base tardios. Os depósitos proximais à intrusão granítica (Campo e Gralheira) são relativamente ricos em sulfuretos e contêm electrum tardio associado a quantidades significativas de sulfuretos de metais de base, enquanto o depósito mais distal à intrusão (Três Minas) é pobre em sulfuretos e contém Au de alta fineza que é precoce. Apesar de apresentar características relacionadas com depósitos de Au orogénicos, a relação regional com depósitos de W-Sn, a proximidade temporal da mineralização com intrusões graníticas, as associações elementares caracterizadas por conteúdos significativos de Bi ou boa correlação Bi-Au, uma paragénese definindo um ambiente com baixa fO_2 -baixa fS_2 e estudos de isótopos de S (e Pb) sugerem que a mineralização está relacionada com granitos reduzidos sin- a tardi-tectônicos e poderá também ser classificada como "intrusion-related".

INTRODUCTION

The genetic model for the mineralization at the Jales district has been controversial. This may be partially due to the fact that different authors studied different parts of the district. NORONHA *et al.* (1993) and NORONHA *et al.* (1995) related the mineralization to metamorphic and meteoric fluids whereas Neiva (1992) invoked fractionated magmatic fluids. In addition to these epigenetic models, syngenetic paleoplacer and exhalative models have been proposed for the Três Minas area by RIBEIRO (1984) and SHEPHERD & OLIVEIRA (1990), respectively.

Some of the characteristics of the Jales deposits, including tectonic setting, structural controls, fluid composition, metal association and mineral assemblages, are indeed compatible with the so called mesothermal or orogenic lode Au deposits, related to metamorphic or meteoric fluids (NESBITT *et al.* 1986; GROVES *et al.* 1998). However, the characteristics of this deposit type often overlap those of the more recently described intrusion-related deposits (THOMPSON *et al.* 1999). This overlap frequently contributes to an equivocal classification of Au deposits, especially in the cases where magmatism and metamorphism took place within a short interval that cannot be resolved by radiometric techniques. The difficulty in distinguishing between orogenic and intrusion-related deposits throughout the world is extended to the studied district, and largely results from the fact that exact criteria to define and distinguish these deposit types remain to be fully determined.

In this paper, we report on the geological setting and geochemical features of mineralization at the three areas, including new Ar/Ar dates and Pb and S isotopic data. The results suggest that mineralization may be of the intrusion-related type, as defined by NEWBERRY *et al.* (1995), MCCOY *et al.* (1997), THOMPSON *et al.* (1999) and THOMPSON & NEWBERRY (2000).

GEOLOGICAL SETTING

The Jales district includes three mineralized areas, from south to north: Campo, Gralheira and Três Minas. These three areas have been subjected to mining during Roman times. In modern times only the Campo deposit was mined, producing approximately 25 tons of Au and 100 tons of Ag (1933-1992).

The Jales district is part of the Iberian Massif, which constitutes the western-most exposure of the European Variscan orogen, resulting from the collision between Laurasia and Gondwana. The Jales district is located within the Axial Domain of the Iberian Massif, the Central Iberian Zone, characterized by subvertical structures and intrusion of large granite batholiths (RIBEIRO, 1990) and best-known for its W-Sn endowment.

In the studied district, both autochthonous and parautochthonous metasedimentary rocks are present. Both have been metamorphosed to greenschist facies, with isograds parallel to the granite intrusion contacts. The autochthonous metasedimentary rocks include the pre-Ordovician Douro Group, composed of flysch-type sedimentary rocks, unconformably overlain by Ordovician quartzites and siliceous mica schists. The parautochthonous Silurian and Devonian sequences that overlay the Autochthonous Domain, above the basal thrust, include the lower Fragas Negras unit and the upper Curros unit, with a gradational contact in between (RIBEIRO, 1998). The Fragas Negras unit is mainly composed of black phyllites, quartzophyllites and quartzites, with minor marbles and calc-silicate rocks. The Curros unit consists of phyllites, schists and some quartzophyllites and quartzites.

The Jales district is bound on the south by syntectonic two mica granites (the Jales massif) and on the west by a post-tectonic biotite granite (the Vila Pouca de Aguiar massif). The Jales massif comprises remnants of an early- to syn-tectonic fine-grained granite, occurring as irregular roof pendants within the larger mass of syn- to late-tectonic granites (NEIVA *et al.* 1995; DIAS *et al.* 1998). The syn- to late-tectonic granites are represented by a medium- to coarse-grained two-mica granite and a volumetrically minor fine-grained tourmaline-muscovite granite. The early- to syn-tectonic fine-grained granite has been dated at 320 ± 6 .

Ma (Rb/Sr whole-rock) and the syn- to late-tectonic medium- to coarse grained two-mica granite has been dated at 308.5 ± 2.4 Ma (Rb/Sr muscovite-rock pairs), by NEIVA *et al.* (1995). The post-tectonic biotite granite was dated at 285 ± 6 Ma (Rb/Sr whole-rock by SHEPERD & OLIVEIRA, 1990).

Fig. 1 portrays the ages of the mineralization and how they relate to the cooling history of the syn- to late-tectonic two-mica granites, indicated by the crossing of successive lower closure temperatures. Three hydrothermal muscovite samples from Três Minas mineralization yielded an average Ar/Ar age of 302.1 ± 0.4 Ma (ROSA, 2001), indistinguishable from Ar/Ar ages obtained by NEIVA *et al.* (1995) for the alteration muscovite associated with mineralization at Campo; 300.7 ± 2.8 Ma in altered granite, and 303.0 ± 2.8 Ma for altered mica-schist. Hence, all the mineralization in the district is of Variscan age. This age data is inconsistent with the syngenetic models for Três Minas. In addition, for both isotopic systems (Rb/Sr and Ar/Ar) the age at which the muscovite cooled below the respective closure temperature is identical for igneous and alteration muscovites (fig.1), indicating that mineralization is synchronous with late cooling of the granite. Since textural evidence revealed that the shearing associated with mineralization overprinted and destroyed the metamorphic fabric characterized by the development of polygonal textures (ROSA, 2001), it can also be concluded that mineralization occurred some time after peak metamorphism.

The Campo mineralized area is related to late-tectonic tension fractures in the Jales massif. The Gralheira mineralization is hosted by a late-tectonic shear zone mostly within Ordovician siliceous mica schists but partially within the Jales massif. Finally, in the Três Minas area, mineralization is also related to a late-tectonic shear zone, exclusively within the Curros Unit of the parautochthonous sequences. The three areas are located at increasing distances from the Jales massif and are therefore amenable to the definition of zonation patterns.

In addition to mineralized veins, barren crustiform quartz veins with pyrite and calcite occur throughout the district. These veins crosscut mineralized structures and are therefore considered post-tectonic.

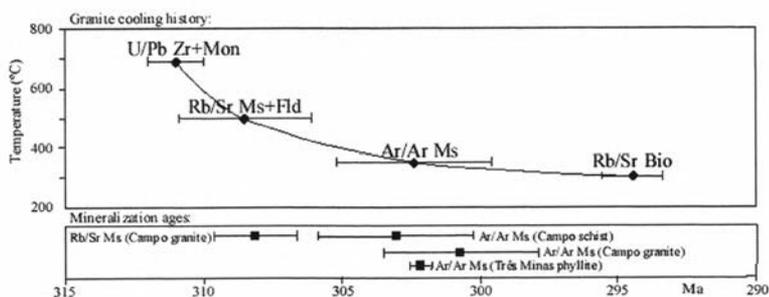


Fig. 1 – Thermal evolution of the Jales syn- to late-tectonic granite (diamonds) and hydrothermal alteration dating (boxes). Is uncertainties. Data from NEIVA *et al.*(1995), ALMEIDA *et al.*(1998) and ROSA (2001).

MINERALIZATION AND ALTERATION

PARAGENESIS

Five stages of mineralization and/or alteration have been recognized in the district; only at Campo are all the stages represented, however (ROSA, 2001). Two paragenetic sequences are presented (figs. 2 and 3), representing the mineralization and alteration assemblages in the deposits proximal and intermediate to the granite intrusion (Campo+Gralheira) and in the more distal deposit (Três Minas), respectively. The different parageneses define a zonation relative to the Jales granite massif, with decrease of the sulfide abundance in general, and of base-metal sulfides in particular, towards distal areas.

Stage I is represented by barren black quartz, which has a ribbon texture due to intense ductile deformation. This stage was accompanied by wall-rock alteration of biotite and chlorite to muscovite, and alteration of igneous plagioclase to muscovite and microcline. This stage was not identified at Gralheira and Três Minas.

Stage II is the Fe-As sulfide stage, characterized by early pyrrhotite and löllingite and later arsenopyrite and pyrite. The löllingite is rare as it has been replaced by arsenopyrite, with löllingite occurring as inclusions within arsenopyrite.

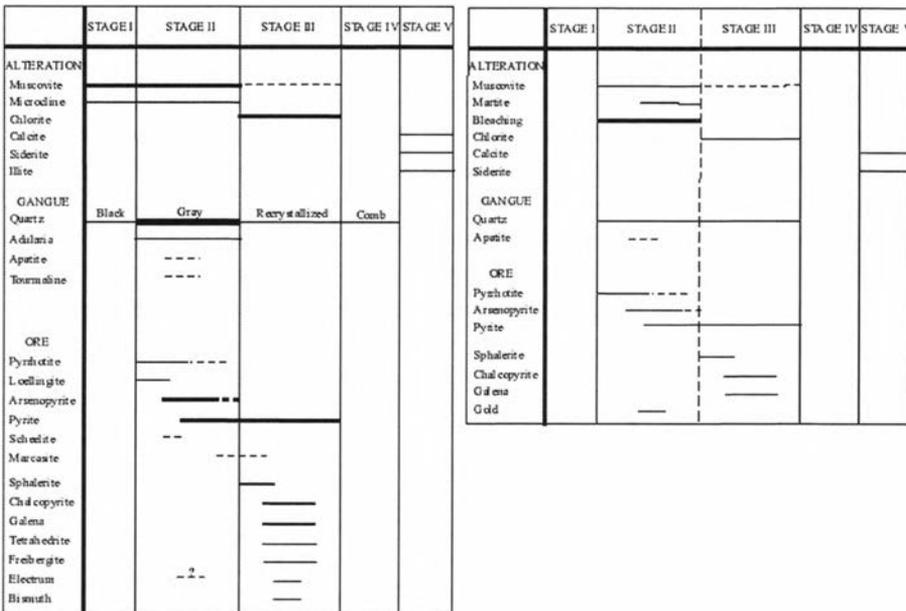


Fig. 2 (left) Generalized paragenetic sequence for the Campo and Gralheira areas (proximal to intermediate to intrusion). The bar width is proportional to mineral abundance.

Fig. 3 (right) Generalized paragenetic sequence for the Três Minas area (distal to intrusion). The bar width is proportional to mineral abundance.

These sulfides occur in gray quartz and locally with adularia. Minor scheelite and marcasite may be related to this stage. At Campo and Gralheira, during this stage more muscovite and microcline were formed similar to Stage I. At Três Minas this stage was accompanied by bleaching of the carbonaceous quartzites hosting the mineralization and martitization of metamorphic magnetite.

Stage III is the base-metal stage, characterized by brecciation of the earlier Fe-As sulfides and the recrystallization of quartz along fractures. Base-metal sulfides and chlorite were deposited along these fractures and as matrix to breccias. Chlorite also replaced muscovite. This stage is very rare at Três Minas, where only trace amounts of base-metals sulfides and chlorite were identified.

Stage IV is defined by the deposition of barren comb quartz along brittle fractures. This stage was not identified at Três Minas. Stage V is characterized by the crystallization of minor amounts of calcite, siderite and illite.

Au occurs as coarse-grained electrum at Campo and Gralheira and as fine-grained high-fineness Au at Três Minas. It is paragenetically earlier at Três Minas, where it belongs to Stage II, in contrast with the other two areas, where it is associated with base-metal sulfides and chlorite of Stage III.

METAL ASSOCIATION

Reflecting the different mineral assemblages, geochemical data presented by ROSA (2001) indicates that the Três Minas area has a distinct metal association signature, in contrast to the other two areas. Whereas the Au enrichment factor is similar at the three areas, base metals and Ag are more than one order of magnitude less enriched or are even depleted at Três Minas (fig. 4).

Of importance regarding the classification of the mineralization at the Jales district, as will be discussed below, are the high Bi contents in areas proximal to the Jales massif and the high Bi-Au correlation at the distal area, represented by Três Minas (fig. 5).

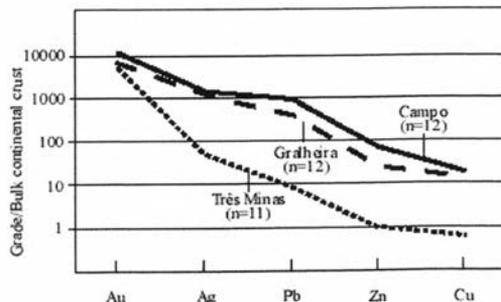


Fig. 4 – Average metal grades normalized to those of bulk continental crust. Bulk continental crust values from the Geochemical Earth Reference Model database (EarthRef.org).

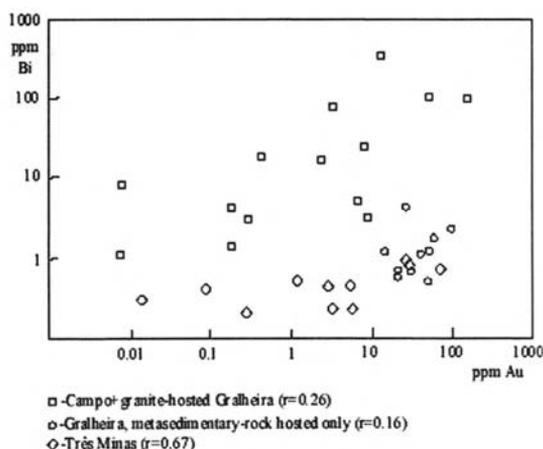


Fig. 5 – Au-Bi correlation and abundances. Correlation factors (r) for each area indicated within parentheses. Note high Bi contents at Campo and positive correlation of Au and Bi at Três Minas.

STABLE ISOTOPES

SULFUR

In order to constrain the source of hydrothermal fluids, S isotope analyses were conducted on sulfide minerals (fig. 6). Within each of the groups considered, metamorphic pyrrhotite, sulfides related to Au mineralization, and pyrite from post-tectonic veins were analysed. The results show a trend of increasingly heavier S from the older to the younger sulfide minerals.

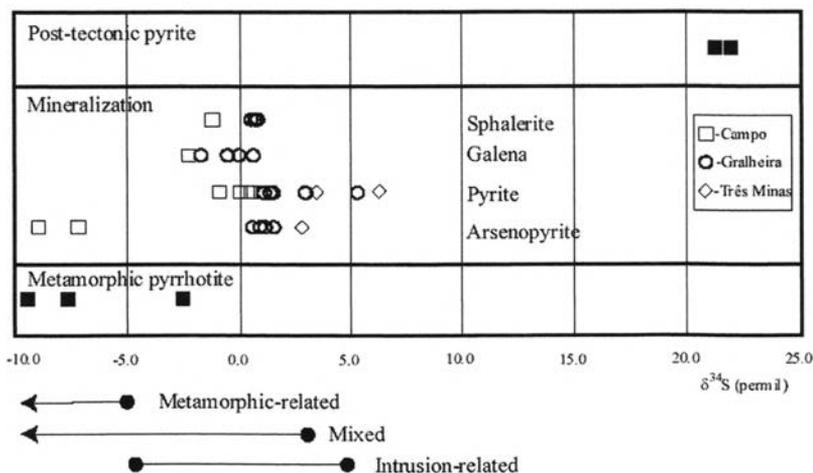


Fig. 6 – S isotope data of sulfides from the Jales district, relative to CDT notation. Ranges of isotopic compositions for sulfides of Alaskan Au deposits compiled by NEWBERRY *et al.* (1995).

The metamorphic pyrrhotite from schistosity-concordant metamorphic quartz lenses and highly deformed thin pyrrhotite ribbons in phyllites has the lightest S, averaging approximately -6.5‰. This pyrrhotite is interpreted to result from the metamorphism of diagenetic pyrite. As indicated by the experimental studies of RIPLEY & SNYDER (2000), there is no significant S isotope fractionation during conversion of pyrite to pyrrhotite during metamorphism, and therefore the isotopic composition of pyrrhotite is a reliable indicator of the isotopic composition of H₂S released during metamorphism and of the original diagenetic pyrite. It is therefore concluded that diagenetic pyrite and fluids released during metamorphic devolatilization also had a sulfur isotope composition close to -6.5 ‰. For the diagenetic pyrite, such a composition is compatible with bacterial reduction of sulfur.

Sulfides belonging to mineralized veins and structures belong to hydrothermal Stage II (arsenopyrite and pyrite) and Stage III (galena and sphalerite). These sulfides plot mostly between -2 and +2‰, with a few arsenopyrite and pyrite outliers. These sulfides show the typical equilibrium fractionation sequence (OHMOTO & RYE, 1979), with $\delta^{34}\text{S}_{\text{gn}} < \delta^{34}\text{S}_{\text{sph}} < \delta^{34}\text{S}_{\text{py}}$. Additionally, for the same sulfide mineral, the S isotopic compositions are heavier at Três Minas than at Campo, with Gralheira displaying intermediate values. Therefore there is an apparent zoning away from the granite intrusion, with S becoming progressively heavier as the distance to the granite increases.

The mineral paragenesis of the Au-mineralized veins contains arsenopyrite, löllingite and iron-rich sphalerite that indicate that the hydrothermal fluid was relatively reduced. For a fluid temperature of approximately 300°C, consistent with previous chlorite solid solution modeling (ROSA, 2001) and fluid inclusion data (SHEPHERD & OLIVEIRA, 1990; NORONHA *et al.*, 1993), and a pH around 6, compatible with buffering by the K-feldspar and muscovite pair, H₂S is the predominant S species in the fluid, hence $\delta^{34}\text{S}_{\text{H}_2\text{S}} = \delta^{34}\text{S}_{\text{fluid}}$ (OHMOTO & RYE, 1979). At 300°C, fractionation factors between sulfides and H₂S in the hydrothermal fluid are not large, with galena being 1.9‰ lighter and pyrite being 1.2‰ heavier than the H₂S in the fluid (OHMOTO & RYE, 1979). From the sulfide isotopic analyses available it is concluded that the hydrothermal fluid would have had a bulk sulfur isotope composition of approximately 0‰, with somewhat lighter values in Campo and somewhat heavier values at Três Minas.

Finally, the pyrite collected from post-tectonic quartz±calcite veins has the heaviest S, approaching +22‰. The origin of this heavy S is not clear but was also reported in post-Variscan veins in the nearby Panasqueira W-Sn deposit (KELLY & RYE, 1989). These authors considered that the heavy S could be derived from heavy sulfate in early Paleozoic water that formed the Douro Group. Since Panasqueira has a similar geologic setting to the Jales district it is concluded that the source for the post-tectonic heavy S could be the same.

LEAD

The Pb isotopic composition of minerals with low U/Pb ratios, such as feldspars and sulfides, helps to discriminate between potential sources for Pb, and presumably other metals, in mineral deposits. K-feldspars from the syn- to late-tectonic granites and adularia and sulfides from the mineralized veins were analyzed. The results were plotted in the uraniumogenic Pb isotope diagram of fig. 7, in which the growth-curve of STACEY & KRAMERS (1975) is drawn for reference. The Pb isotope composition of K-feldspars from the syn- to late-tectonic granites lie very close to Stacey-Kramers growth curve, indicating model ages consistent with the geochronological ages obtained for these rocks (≈ 300 Ma). Thus, the crustal source for the syn- to late-tectonic granites was not significantly enriched in U and Th relative to the STACEY & KRAMERS (1975) model.

In contrast with the plutonic K-feldspars, the vein minerals, both sulfides and adularia, have more radiogenic Pb compositions. These minerals display model ages that are incompatible with the Ar/Ar data, even showing future ages. The more radiogenic Pb of the mineralized veins can be best explained by considering one or a combination of the two following processes: 1) contamination of Pb-poor vein

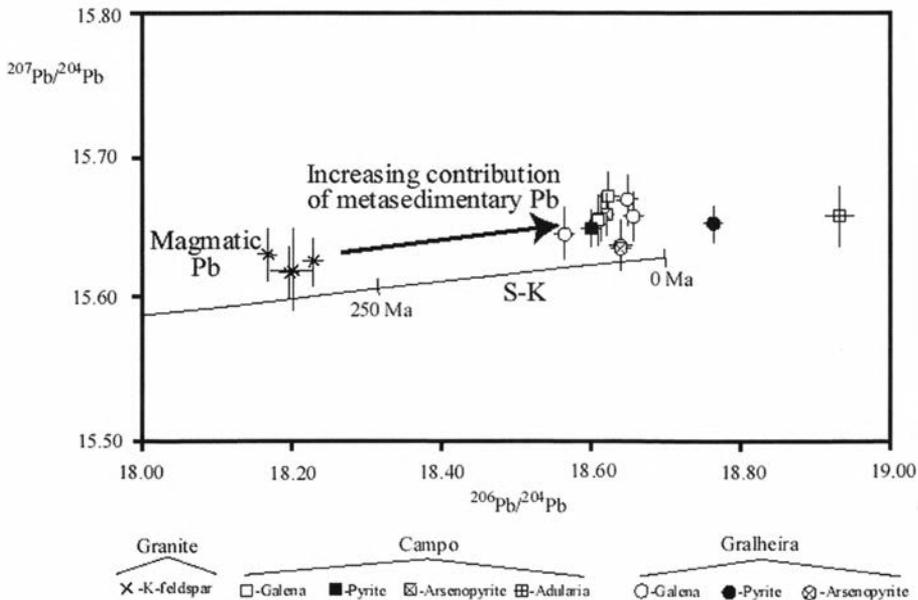


Fig. 7 — Uranogenic Pb isotope diagram, with STACEY & KRAMERS (1975) growth-curve. Some of the more radiogenic Pb in the vein minerals may have resulted from contamination of Pb-poor vein minerals with U, which decayed to give more radiogenic present-day Pb isotopic values. Arrow indicates increasing contribution of Pb leached from the metasedimentary rocks to the Pb in mineralized veins. Uncertainties are at 95% confidence level.

minerals with U, which decayed to give more radiogenic present-day Pb isotopic values; and/or 2) an input of radiogenic Pb from old and U and Th-rich metasedimentary rocks at the time of vein mineral deposition. Metasedimentary rocks, especially black phyllites, commonly have significant amounts of U and Th and therefore contribute Pb with high isotopic ratios. Considering that the degree of contamination with U should be the same for each mineral species, the fact that, for the same mineral, the Pb isotopic ratios generally increase outwards from the magmatic system, from veins hosted in granite (Campo) to veins hosted in metasedimentary rocks (Gralheira), suggests that the trend displayed in fig.7 resulted to a certain degree from the mixing of Pb from two different homogeneous sources, a contribution of magmatic Pb derived from the granites, and more radiogenic Pb leached from metasedimentary rocks. The relative contribution from each of the two proposed sources is dependent on the Pb content and isotopic composition of the metasedimentary rocks and cannot be evaluated with the present data.

DEPOSIT CLASSIFICATION AND FLUID SOURCE

A magmatic source for the fluids related to the Jales mineralization is supported by some features that are typical of intrusion-related systems as defined by NEWBERRY *et al.* (1995), MCCOY *et al.* (1997), THOMPSON *et al.* (1999) and THOMPSON & NEWBERRY (2000). These include the regional association with W-Sn deposits, high Bi contents or Au-Bi correlation (fig. 5) and the low fO_2 -low fS_2 environment for mineralization as documented by the presence of mineral phases such as löllingite, pyrrhotite and native Bi.

Also, as discussed in the section on the geological setting, radiometric data indicate that mineralization appears to be related to protracted cooling of the syn- to late-tectonic granites, and textural evidence indicates that mineralization happened after peak metamorphism. This means that fluids released during prograde metamorphism would no longer have been available by the time the granite massifs were emplaced and mineralization took place, and therefore the mineralization is more likely related to magmatic fluids.

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Additionally, despite the proposed contribution of a certain amount of metasedimentary Pb, Pb isotope data is not incompatible with an intrusion-related model. Mixing lines, similar to the one in fig. 7, with Pb contributed from magmatic and sedimentary sources, have been proposed for the Telfer deposit in Australia by GOELLNIGHT *et al.* (1989) and for the Pogo deposit in Alaska by SMITH *et al.* (1999).

Finally, S isotope data are most reasonably explained by a predominantly magmatic fluid, with only minor and localized contribution from nonmagmatic fluids. The $\delta^{34}S$ values of the sulfides in the mineralized veins resulted from a

fluid with $\delta^{34}\text{S}\approx 0\%$, a value generally interpreted to be indicative of a magmatic source. The fact that Variscan granitic rocks of western Spain have been confirmed to have $\delta^{34}\text{S}$ values between -3.9 and 4.8% (RECIO *et al.*, 1991), further supports that the S probably had a magmatic source. A significant metamorphic source for the S can be excluded for the following reasons. The desulfidation reaction converting diagenetic pyrite into pyrrhotite with release of S causes no significant isotopic fractionation (RIPLEY & SNYDER, 2000). Because the analyzed metamorphic pyrrhotite is isotopically light ($\delta^{34}\text{S}\approx -6.5\%$) it is doubtful that heavier fluids ($\delta^{34}\text{S}\approx 0\%$) could be generated by desulfidation. However, a small metamorphic contribution cannot be excluded for some of the S in arsenopyrite in the Campo area, which yielded light S compositions. Even lighter S isotope results were described at the intrusion-related Au deposit of Donlin Creek, Alaska, by SZUMIGALA *et al.* (2000).

The slightly heavier sulfides from Gralheira and especially Três Minas are possibly the result of a small input of heavier S from a nonmagmatic source, probably the same source as the S in the post-tectonic pyrite.

Fig. 6 includes the ranges of S isotopic compositions for sulfides of different Alaskan Au deposits, as compiled by NEWBERRY *et al.* (1995). This shows a good overlap between the data collected during this study and the intrinsic Au deposits, for which a magmatic origin has been established by NEWBERRY *et al.* (1995). In contrast, in extrinsic and mixed deposits, for which a metamorphic origin has been established, at least in part, the sulfides have much lighter S isotopic compositions ($\delta^{34}\text{S} < -10.0\%$). Since such isotopically light sulfides have not been recorded, this further strengthens the argument for a magmatic source for the S at the Jales district.

SUMMARY AND CONCLUSIONS

The Jales district mineralization seems to belong to an ever growing class of deposits of enigmatic nature and ambiguous interpretation. In spite of this, and although no exact criteria to distinguish orogenic from intrusion-related Au deposits have been established to date, it is suggested that the Jales district is most likely an example of intrusion-related Au deposit formed by the accumulation of Au in late magmatic fluids, due to its incompatible behavior during evolution of reduced magmas (MCCOY *et al.*, 1997).

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