HYDROTHERMAL ALTERATION AND PARAGENETIC SEQUENCE IN THE GOLD OCCURRENCES OF THE AROUCA REGION (NW OF PORTUGAL)

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Abstract

Gold mineralizations in the Arouca region are hosted within silicification bands which correspond to minor shear zones subparallel to the Carboniferous Trough Shear Zone. The development of some of these shear zones is related to the D_3 deformation phase of the Central Iberian autochthon. However, gold mineralization is a late process. The main hydrothermal event is characterised by the development of several quartz+sericite veins together with sericitization of the host rocks. The sulphides (±Au) occur later, especially within fractures, sometimes with later quartz (± chlorite). These sulphides are presently extensively oxidized. Vila Cova is rather a Sb occurrence with minor Au content. These observations suggest, together with field data, that mineralization events are mainly related to the late-D_3 deformation phase. Geochemical analyses also show that the highest gold values are found within late reactivated structures (especially faults), but the hypothesis of a reconcentration process is improbable. Future work will attempt to characterise some of the physico-chemical conditions of gold deposition.

The gold occurrences of Arouca are situated in the vicinity of the Carboniferous Trough Shear Zone (**CTSZ**) which borders the SW limb of the Valongo anticline, both major D_1 variscan structures of the Central Iberian Zone autochthon. These occurrences are part of a major metallogenetic province extending from Póvoa de Varzim (N of Porto) to the Castro Daire granite to the SE, along a NNW-SSE to NW-SE direction (figure 1). Especially in the area between Valongo and the right margin of the Douro river, several Au and Sb-Au mineralizations are known which can be placed in two distinct groups: (a) Au mineralizations hosted by the Arenigian quartzites and developed essentially in the NE limb of the Valongo anticline; (b) Sb-Au mineralizations hosted by the Schist and Graywacke Complex (**SGC**) and Carboniferous lithologies developed mainly to the SW of the **CTSZ**.

In the Arouca area, four gold occurrences were identified which were object of geological exploration campaigns for Sb and Au - Vila Cova, Toural E, Toural W and Chiqueiro (figure 1). These occurrences are domains of silicification of the host rocks where some mineralized quartz veins are found. At Vila Cova and Toural E the occurrences are located within the SW limb of the Valongo anticline hosted by the **SGC** lithologies and Arenigian quartzites respectively. Chiqueiro and Toural W are located to the SW of the **CTSZ** hosted by **SGC** lithologies. In Vila Cova, Toural W and Chiqueiro, the silicification of the host rocks was due to the localized development of minor shear zones subparallel to the **CTSZ**, within which the mineralized quartz veins occur (Gonçalves et al., 1995). The silicification of the host rocks can be essentially pervasive (Vila Cova; Toural E), or result from many minor quartz veins (Toural W; Chiqueiro). The development of such shear zones, as well as part of the earlier quartz veins, can be ascribed to the D_3 deformation event identified in the Central Iberian Zone (Vila Cova still remains unsolved for this particular problem). Nevertheless, the main mineralizing event is essentially related to the late-D_3 deformation phase which caused the late reactivation of the **CTSZ** as a reverse left-lateral shear zone, and consequently some of the earlier structures.

The weathering of the mineralized structures constitutes a striking feature in the area. As a mater of fact, with the exception of Chiqueiro, in all the other 3 areas there is

extensive oxidation of the former ore minerals, mainly with the formation of iron ox-ides/hydroxides. In Chiqueiro, part of the primary ores (especially arsenopyrite) are pre-served in spite of the relative abundance of scorodite and other unidentified arsenates. Qualitative microprobe analysis also showed that within the main mineralized quartz structures of Vila Cova, Sb, Fe, and As occur in the oxidized material.
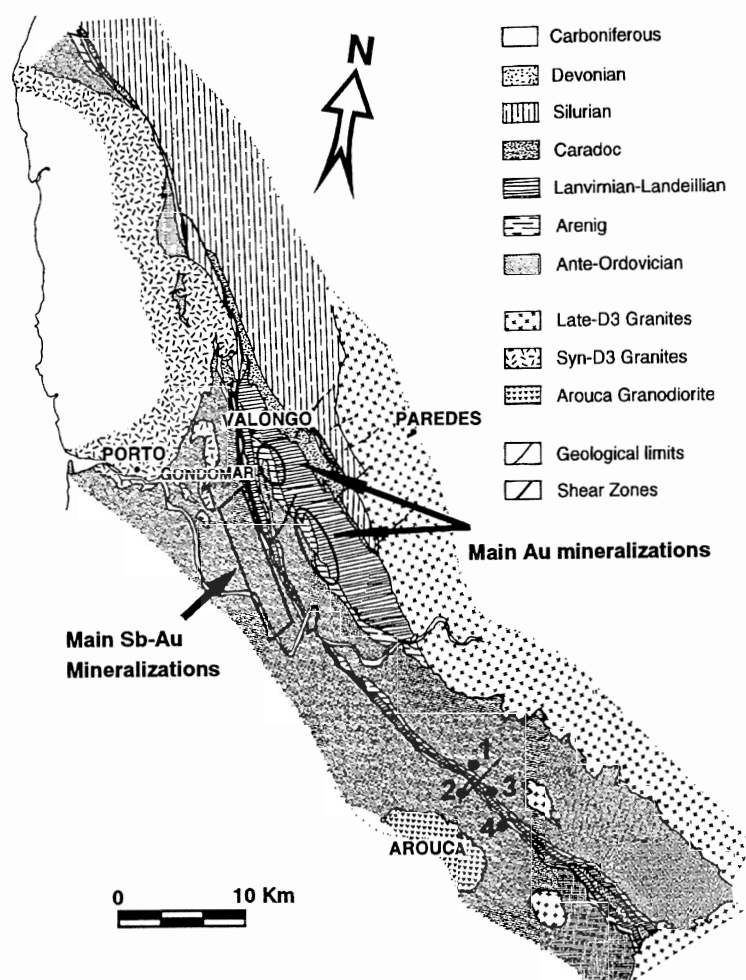

Figure 1. Simplified geological map of the Valongo anticline area (Central Iberian autochthon) with the location of the main Au and Sb-Au mineralizations as well as of the studied gold occurrences: **1:** Vila Cova; **2:** Toural W; **3:** Toural E; **4:** Chiqueiro.

The petrographic characterisation of several polished thin sections allowed the definition of a general synthetic paragenetic sequence for all areas (figure 2). The most striking character of the mineralized zones is the extensive deposition of quartz, giving rise to the silicification bands. This quartz generally represents the first episode of mineral deposition, and is globally termed *quartz I*. In Vila Cova, and to a lesser extent in Toural E, the quartz is microcrystalline obliterating former veins, frequently pre-tectonic ones. The deformation is characterised by the development of several ductile microstructures (subgrains, deformation bands and lamellae, and recristalization). In Toural W and Chiqueiro the silicification bands result both in the formation of the main mineralized veins and related minor quartz veins in the host rocks. The microstructures exhibited by *quartz I*

in these areas is different from the former ones. Generally, the quartz is less deformed and shows rare recristalization. During the last impulses of this stage in Chiqueiro, the precipitation of *arsenopyrite I* occur within the major veins.

The main hydrothermal event is later and characterised by the deposition of a moderately deformed *quartz II* + *sericite*, sometimes accompanied with *arsenopyrite II* (only in Chiqueiro) and minor *pyrite*, *chalcopyrite* (± *arsenopyrite*) observed as relicts within *goethite* and *hematite*. Deformation proceeds with brecciation and fracturing of the quartz veins and host rocks, accompanied by the deposition of *pyrite* and *chalcopyrite* (also relicts within *goethite* and *hematite*), *quartz III* (generally undeformed), and *chlorite*. The formation of *chlorite* is clearly a late process associated with the fracturing of the rocks. This is particularly evident in Toural E where a complete chloritization of the rock matrix within the late fault zones is observed. In this particular case, several chlorite generations are developed: (a) *chl I* replacing former *sericites* (of metamorphic and hydrothermal nature); (b) veins of *chl II* in later fractures; (c) veins of *chl III* + *qz* + *anatase*; the deposition of *wavellite* marks the final stage of the hydrothermal system.
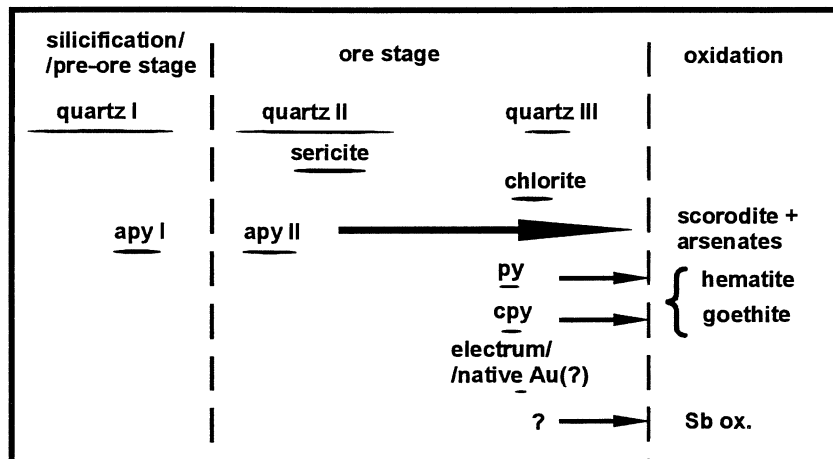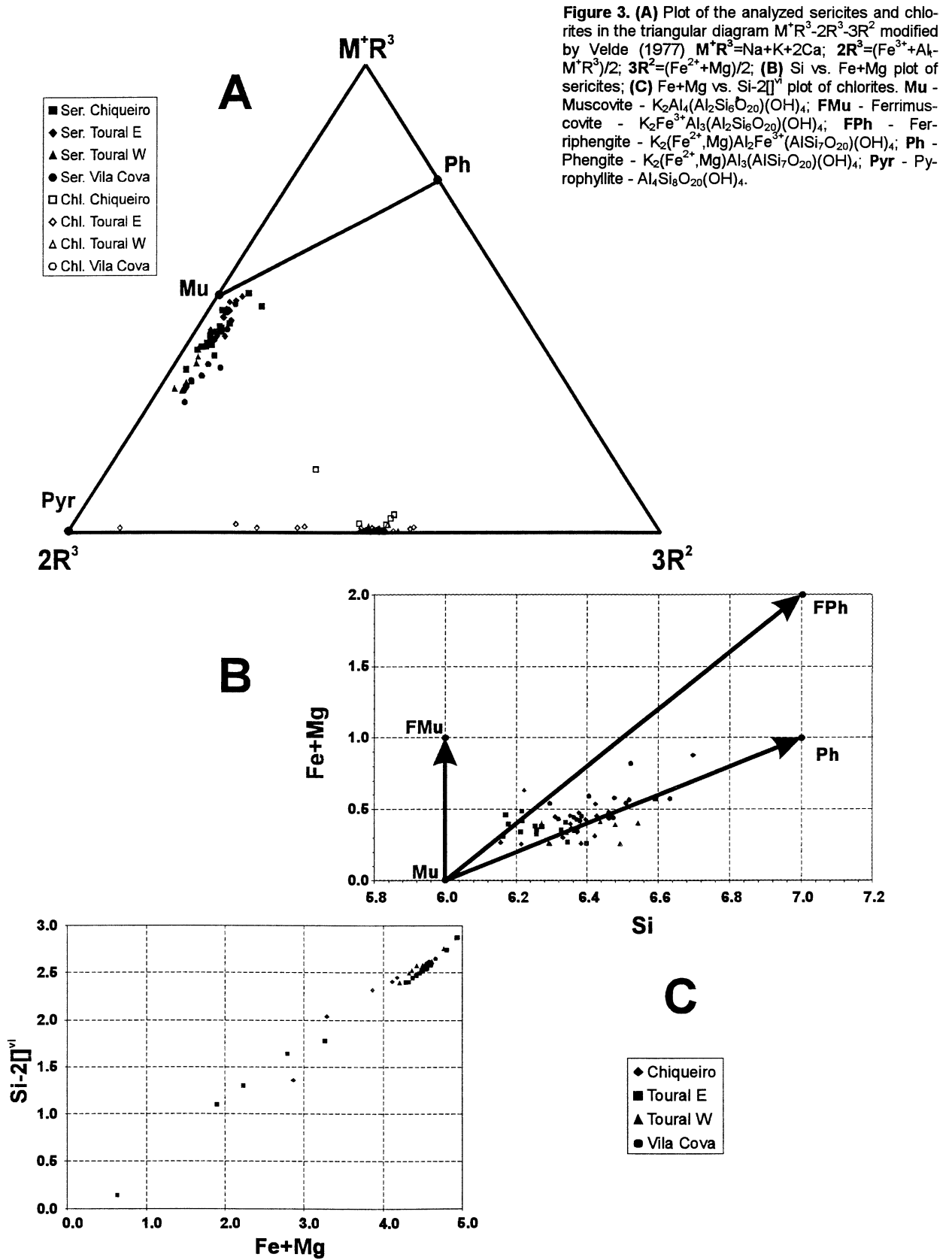
silicification/ /pre-ore stage		ore stage		oxidation
quartz I		quartz II	quartz III	
		sericite		
			chlorite	
apy I		apy II ━━━━━━━➤		scorodite + arsenates
			py ━━➤	⎰ hematite
			cpy ━━➤	⎱ goethite
			electrum/ /native Au(?)	
			? ━━➤	Sb ox.

Figure 2. Synthetic paragenetic sequence for the occurrences of Arouca region. The **apy I**, **apy II**, and **electrum** are found exclusively in Chiqueiro (minor **apy** Is also found in Toural E). The Sb oxides were identified only in Vila Cova.

The characterisation of hydrothermal minerals (*sericites* and *chlorites*) was achieved with the definition of the main substitution vectors. *Sericites* show a systematic deficiency in K (fig. 3a) without any significant increase in Si in the tetrahedral position (therefore excluding illites), and a deviation from the ideal dioctahedral structure towards a trioctahedral one. The inverse correlation between the total occupancy in the octahedral and dodecahedral sites may put in evidence the mechanism of substitution through the vector $R^{2+(vi)}\square^{xii}_2 R^{+(xii)}_{-2}\square^{vi}_{-1}$, which may explain the K deficiency and part of the dioctahedral deviation. Nevertheless, for this point, the analyzed *sericites* stay systematically near the ideal substitution line $R^{2+(vi)}_3 Al^{vi}_{-2}\square^{vi}_{-1}$, which probably suggests the intervention of both vectors. The incorporation of most Fe and Mg in the *sericite* structure is however best explained by the tschermak substitution $(Si^{iv}R^{2+(vi)}Al^{vi}_{-1}Al^{iv}_{-1}$ - fig. 3b). Some analyses showed significant iron content, but the available data is insufficient to fully characterise them. Nevertheless, the coexistence of an oxide paragenesis may also favour that the oxidized *sericites* (typically with 2-4 wt% of FeO) may have Fe^{3+}, as suggested by Guidotti (1984), since it also could explain the *sericite* plots above the muscovite-phengite line in fig. 3b.

Figure 3. (A) Plot of the analyzed sericites and chlorites in the triangular diagram M^+R^3-$2R^3$-$3R^2$ modified by Velde (1977) M^+R^3=Na+K+2Ca; $2R^3$=(Fe^{3+}+Al-M^+R^3)/2; $3R^2$=(Fe^{2+}+Mg)/2; (B) Si vs. Fe+Mg plot of sericites; (C) Fe+Mg vs. Si-2[]^{VI} plot of chlorites. Mu - Muscovite - $K_2Al_4(Al_2Si_6O_{20})(OH)_4$; FMu - Ferrimuscovite - $K_2Fe^{3+}Al_3(Al_2Si_6O_{20})(OH)_4$; FPh - Ferriphengite - $K_2(Fe^{2+},Mg)Al_2Fe^{3+}(AlSi_7O_{20})(OH)_4$; Ph - Phengite - $K_2(Fe^{2+},Mg)Al_3(AlSi_7O_{20})(OH)_4$; Pyr - Pyrophyllite - $Al_4Si_8O_{20}(OH)_4$.

The analyzed *chlorites* of Toural E show different Fe/(Fe+Mg) ratios in relation to the other areas. In these areas, the ratio is close to 0.5 in contrast to Toural E, where *chlorites* are systematically Fe-rich (with the exception of *chl III*). Also the specimens of *chl II* of Toural E are more Si and Al-rich (fig. 3a), which is interpreted as a result of partial argilization. The sum of the octahedral sites is in most cases close to the ideal 6 per formula (basis of 14 O_2), and the plot of these analyses is also close to the ideal tschermak substitution line, represented by the vector $R^{3+(vi)}Al^{iv}R^{2+(vi)}_{-1}Si^{iv}_{-1}$. However, the $Al^{vi}+2Ti+Cr$ content is systematically greater than Al^{iv} in some of the specimens, with the consequent deficiency in the octahedral occupancy. The observation of fig. 3c indicates that the substitution vector $Si^{iv}_2\square^{vi}_1R^{2+(vi)}_{-1}Al^{iv}_{-2}$ is most probably the main mechanism for these specimens. It is also important to note that the lack of data relating the Fe^{3+} content in *chlorites* constitutes one major limitation to this analysis.

The main mineralized structures of Arouca were essentially developed during the late-D_3 deformation phase. The paragenetic sequence and geochemical analyses of several rock samples corroborate this conclusion. As a mater of fact, the analyzed arsenopyrite rich veins of Chiqueiro show that most of them correspond to a low grade ore (less than 0.5 ppm Au). On the other hand, when these same veins are late becciated, they show 2-7 ppm Au. In Toural E, in spite of the low values (only up to 0.25 ppm Au), it is also within the late faults that the highest values are found. Vila Cova (1 sample analyzed) is essentially an Sb occurrence with only minor Au. This is in accordance with the petrographic data which shows that most of the oxidized material (essentially former sulphides) is within late fractures in early quartz, or together with the later quartz (even in Vila Cova). It is worth noting the absence of Ag in all samples, suggesting that probably most of the Au is in the native form. It was not possible to determine the Sb supporting phase. Electrum was found within late fractures in a quartz vein and identified by qualitative microprobe analysis in only one sample of Chiqueiro. The data from Chiqueiro might suggest that gold mineralization could be a result of later reconcentration in secondary structures from a former low grade ore (particularly arsenopyrite-rich), in a similar way to the shear zone model proposed by Bonnemaison & Marcoux (1990). However, there are no evidences that support this hypothesis. The limited extent of the veins, together with the lack of correlation between As and Au, led us to consider that most probably the main Au mineralization is an independent process; this is actually a common feature of most Au-mineralizations in Central Iberian Zone. Work in progress should enable the characterisation of the mineralizing fluids and, therefore of the main physico-chemical conditions of gold deposition.

References

Bonnemaison, M. & Marcoux, E., 1990. Auriferous mineralization in some shear-zones: A three stage model of metallogenesis. Mineral. Deposita, 25: 96-104.

Gonçalves, M., Mateus, A. & Barriga, F. J. A. S., 1995. Structural control of gold occurrences in the Arouca region and their relationships to the Carboniferous Trough Shear Zone (NW Portugal). In: M. D. Rodriguez Alonso & J. C. Gonzalo Corral (Eds), XIII RGOP/PICG - 319-320, p. 180-184. Salamanca.

Guidotti, C. V., 1984. Micas in metamorphic rocks. In: S. W. Bailey (Ed), Micas. Reviews in Mineralogy, vol. 13, p. 357-467. Mineralogical Society of America.

Velde, B., 1977. A proposed phase diagram for illite, expanding chlorite, corrensite and illite-montmorillonite mixed layered minerals. Clays and Clay Minerals, 25: 264-270.