Geochronology of the very low-grade metamorphism in metabasites: an approach from isotopic geochemistry

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Introduction

Recent advances in very low-grade metamorphism of metabasites have been centered on the quantification of the intensive P-T-X conditions using internally consistent thermodynamic database (e.g. Powell et al., 1993; Robinson et al., 2004, 2005; Day and Springer, 2005). However, in contrast with research in low, medium or high-grade metabasites, precise determination of the timing on which very low-grade processes occurred has been a very scarcely exploited topic, mostly as a consequence of the reduced number of neophormed minerals (mostly dominated by hydrated Ca-Al silicates and mafic phyllosilicates, e.g. Schiffman and Day, 1999, Bevins 2007, this volume) during these low P-T processes. This limitation can be resolved when metabasites are intercalated with metapelites, for which K-Ar dating of clay minerals is a reliable methodology to determine timing in diagenetic to slightly-metamorphosed sediments (Clauer 2007, this volume). Nevertheless, in active continental margins and island-arc domains, lithologies are mostly dominated by volcanic and volcaniclastic rocks that, as in the case of the Andean Cordillera of central Chile, are dominated by basic to intermediate rocks. Moreover, the evolution of the Andean Cordillera is the consequence of discrete basins formation, development of magmatism (volcanic arcs coeval with a plutonic root), subsidence- and consequent very low-grade metamorphism- and final exhumation to achieve it present relief. On this context, the precise timing of metamorphism and, moreover, the quantification of the time-interval between deposition of volcanic materials in a regional subsiding basin and the generation of low-grade metamorphic assemblages in those same materials due to burial is critical to the global understanding of the building of this segment of the Cordillera. Nevertheless, it is to remark the scarce number of papers developed concerning this topic previous to the 90's (e.g. Åberg et al., 1984), mostly because of the difficulty to obtain reliable ages for these two events. This difficulty is mainly due to the reduced number of mineral phases compositionally suitable to apply methods such as the K-Ar or $^{40}Ar/^{39}Ar$ on the one hand and, on the other, because of the small amount in which these phases are commonly found. Among them, the K-feldspar adularia, the sericitic micas and the celadonites are the best known examples of K-bearing secondary minerals present in metabasites in sub-greenschist facies. Moreover, when secondary mineral phases are well developed, primary minerals are generally too altered to allow correct dating of the volcanism.

The present work is a review of the progress achieved in the understanding of timing of the very low-grade metamorphic events in the Mesozoic arc/back-arc sequences from the Coastal and Andean cordilleras in central Chile. Also, our progress in inserting metamorphism in the geodynamic evolution in time of ensialic marginal basins generated under spreading-subsidence conditions along active continental margins is reviewed. A revision of the different geochronological methods applied to the determination of the timing of metamorphism would be also developed.

Rb-Sr geochronology

Investigations concerning low-grade alteration effects in volcanic rocks show that the alteration might be accompanied by opening and significant disturbances of the Rb-Sr system (e.g. Åberg, 1985). The general expression governing the Rb-Sr geochronology is given by the equation

$$\left(\frac{^{87}Sr}{^{86}Sr}\right)_o = \left(\frac{^{87}Sr}{^{86}Sr}\right)_m + \left(\frac{^{87}Rb}{^{86}Sr}\right)_m \left(e^{\lambda t} - 1\right) \quad \text{(eq.1)}$$

on which o indicates the initial ratios and m the present-day measured ratio. Different altered rocks with variable Rb/Sr ratios could plot along a pseudo-isochron line giving information about the timing of alteration.

Åberg et al. (1984) applied the Rb-Sr systematics to the lower Cretaceous basaltic rocks from the Coastal Range of central Chile (Figure 1a). Rb-Sr data for six "unaltered" samples of these basalts plot close to a reference line for 117 Ma (assumed age of volcanism based on palaeontological data from interbedded limestones). The intersection with the y-axis (0.7038±0.0004) would represent the initial Sr isotopic signature. When plotting strongly altered basalts, a similar Sr(i) was obtained (0.7040±0.0002). Moreover, Ca-Al silicate present in one amygdule sample, in which Rb-bearing minerals are not present, gave a similar Sr(i) (0.7040±0.0001) and consequently reflects the Sr(i) of its host rocks at the time of metamorphism, i.e. when the amygdule was formed. The Rb-Sr whole rock systems closed at 102±3 Ma (Figure 1b), which would be about 10-20 Ma after the flows were extruded.

Munizaga et al. (1988) carried out whole-rock Sr isotopic analyses on volcanic rocks hydrothermally and pervasively altered belonging to the stratabound copper deposits of El Soldado and Lo Aguirre, stratigraphycally located below the basaltic sequences studied by Åberg et al. (1984). Rb/Sr isochrons of 109±4 Ma and of 113±3 Ma were obtained for these samples respectively. These figures can be interpreted in terms of dating regional very low-grade metamorphism affecting these Mesozoic volcanic sequences.

Nevertheless, in spite of these previous rather favourable results obtained using the Rb/Sr systematics to date very low-grade metabasites, this method could not be considered as a practical or valid geochronometer due the isotopic heterogeneity that could present different basaltic lavas. Moreover, differential mobility of Rb and Sr in low-T fluid/rock processes (controlled by permeability, porosity, texture, etc., of primary rocks) could imply different $(^{87}Rb/^{86}Sr)_m$ and $(^{87}Sr/^{86}Sr)_m$ ratios and errochrons could be more frequent than real isochrons.

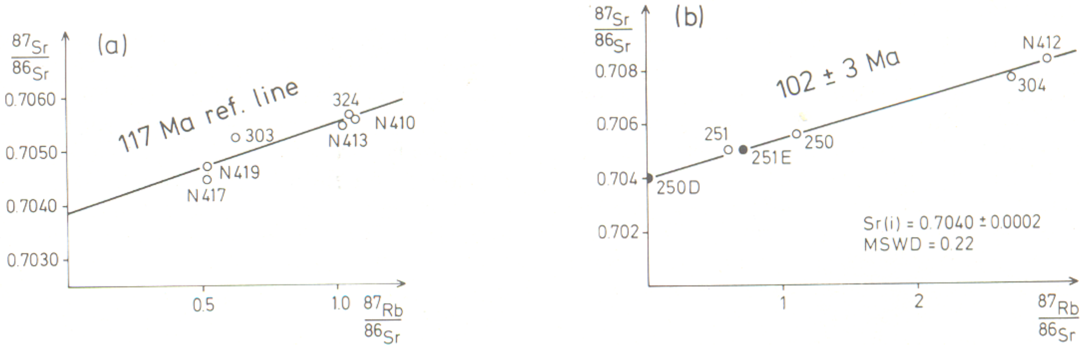

FIGURE 1. *Rb-Sr whole rock diagrams for Lower Cretaceous porphyritic flood basalts, central Chile (modified from Åberg et al., 1984). (a) Samples of "unaltered" basalt from six flows. The scatter is probably due to incipient alteration, present in even the best-preserved samples. The reference line for 117 Ma is based on palaeontological data. (b) Samples of strongly altered basalt (circles) and Ca-Al silicate in amygdales (filled circles) from three of the flows plotted in (a).*

K-Ar geochronology

The K-Ar systematics is a rather simple geochronological method based on the natural $^{40}K \rightarrow ^{40}Ar$ and $^{40}K \rightarrow ^{40}Ca$ isotopic decays. The age expression: is given by the equation

$$t = \frac{1}{\lambda} \ln\left[\frac{^{40}Ar^*}{^{40}K} \left(\frac{\lambda}{\lambda_{Ar}} \right) + 1 \right] \quad \text{(eq. 2)}$$

where $\lambda = \lambda_{Ar} + \lambda_{Ca}$ and $^{40}Ar^*$ refers to ^{40}Ar that is radiogenically produced *in situ.* There are some assumptions that must be met for this expression to yield a reliable crystallization age, the most important being (1) absence of any loss of radiogenic $^{40}Ar^*$ from the material since its formation; (2) absence of loss or gain of ^{40}K other than by radioactive decay (i.e., the material must be chemically closed); and (3) absence of excess argon in the sample.

As in the Rb/Sr systematics, the application of this methodology to metabasites is rather restricted due the relatively low-K content in the typical very low-grade metamorphic minerals present on these systems. Among the K-bearing minerals, sericite, K-feldspar (adularia) and celadonite seem to be the most appropriate to be dated with this methodology. However, the grain size of these minerals commonly makes its separation from the host rock difficult. Moreover, it is rather frequent that sericite and K-feldspar develope on primary Ca-rich plagioclase which, in some cases, preserves relict patches of unaltered igneous plagioclase (e.g. Morata et al., 1997). In these cases, a K-Ar age obtained from altered plagioclase could render a mixed age between magmatism and metamorphism.

Some works have been carried out using the K/Ar systematics on pervasively and highly altered basic whole-rocks with different alteration degree (e.g. Munizaga et al., 1988, Puga et al., 1988). In these cases, the youngest age obtained was considered as the best estimation of the timing of very low-grade metamorphism.

In the central Andes and in the Coastal Cordillera in central Chile, the K-Ar systematics has been successfully applied to celadonites filling amygdules in very low-grade metamorphosed Mesozoic basic lavas (Belmar et al., 2001; Morata et al., 2006). In most cases, celadonite appears as a thin film at the outer rim of amygdules (Figure 2), intimately intergrown with chlorite and mixed-layer chlorite/smectite, in equilibrium with metamorphic minerals of the zeolite and prehnite-pumpellyite facies. Analytical results give ages with geological significance, showing two different metamorphic events developed 15 and 45 Ma latter the volcanism present in these Mesozoic sequences. Nevertheless, the application of the K-Ar systematics to other K-rich fine-grained phyllosillicates (*e.g.* illite in the groundmass of very low-grade metamorphosed volcaniclastic rocks) gives ages without regional significance probably due to (i) the growing mechanisms of illite in mafic systems and (ii) the facility to develop illite in the groundmass of volcaniclastic rocks during local alteration processes (Morata et al., 2006).

One of the major problems of the K/Ar systematics is related to the fact that K and Ar are measured separately on different sample aliquots and under different analytical techniques (atomic absorption *vs* mass spectrometry). Obviously this fact could introduce some degree of heterogeneity (e.g. heterogeneous distribution of daughter $^{40}Ar^*$) to our measurements giving values without any geological meaning.

Ar-Ar incremental heating geochronology

In the $^{40}Ar/^{39}Ar$ methodology, both isotopes can be measured on the same material so there is no need to assume sample homogeneity as in the K-Ar systematics. The $^{40}Ar/^{39}Ar$ incremental heating technique provides very useful information about Ar distribution on the sample. The final

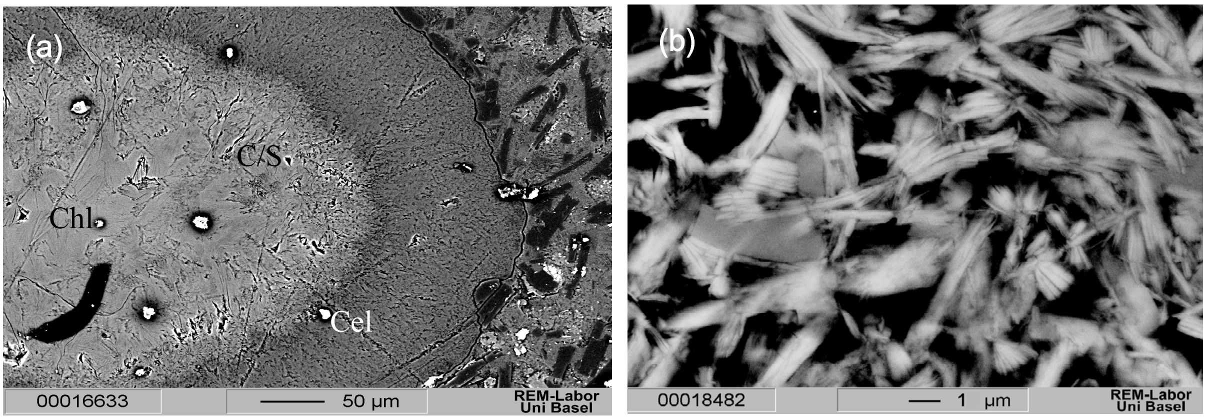

Figure 2. *(a) Back-scattered image of amygdule in andesitic lava showing the intimate relationships between celadonite (Cel), chlorite (Chl) and mixed layer chlorite/smectite (C/S). (b) Detail of celadonite crystals.*

result of the step heating experiments is a plot of apparent age versus the amount of argon released at each temperature step. These plots are called argon release spectra. The expression to solve the age of an unknown sample is giving by the equation

$$t = \frac{1}{\lambda}\ln\left[\frac{^{40}Ar}{^{40}K}J + 1\right] \qquad \text{(eq. 3)}$$

being

$$J = \frac{\lambda}{\lambda_{Ar}}\frac{^{39}K}{^{40}K}\Omega \qquad \text{(eq. 4)}$$

where Ω is an efficient factor defined experimentally in the nuclear reactor.

The ^{40}Ar/^{39}Ar methodology was used for the first time in dating very low-grade metabasites of the Lower Cretaceous of the Coastal Range of central Chile by Aguirre et al. (1999). Later, Wilson et al. (2003) and Fuentes et al. (2005) dated metabasites of these volcanic sequences. Crystals of: (i) transparent plagioclase, (ii) strongly sericitized plagioclase and (iii) adularia were analysed by the ^{40}Ar/^{39}Ar step heating procedure (Figure 3). Plagioclase were separated from basic flows using a magnetic separator and then carefully selected under a binocular microscope in order to analyse a ca. 20-mg fraction or small clusters (30–32) of transparent grains only. The most milky-white plagioclase crystals (strongly sericitized), 500–1000 µm in size, were also selected in order to analyse sericite directly in plagioclase (discrete sericite crystals could not be separated from plagioclase). Finally, crystals of adularia between 800 and 1500 µm were directly hand-picked from amygdules and then carefully selected under the binocular microscope.

The analytical results using this methodology permits dating both the volcanism and the metamorphism (Figure 4), allowing us to determine the time interval between these two events.

Apparently valid ages of both, the emplacement of the lava flows (by dating transparent plagioclase) and their subsequent very low-grade metamorphic event (by dating sericite and/or adularia) were obtained from the same basic lava series, and even on the same rock sample (Figure 4). Sericite, extensively developed in altered basalts, could be dated by analysing single grains of strongly sericitized plagioclase. The validity of this age is supported by concordant ages obtained on adularia (Fuentes et al., 2005).

Morata et al. (2006) also used the ^{40}Ar/^{39}Ar methodology in dating celadonites in amygdules. Nevertheless, the ^{40}Ar/^{39}Ar spectra during the incremental-heating experiment lost ^{39}Ar by recoil without development of a plateau age, giving a disturbed spectrum without geological significance (Figure 5). Moreover, the spectrum obtained is similar to the spectrum for the SED-2

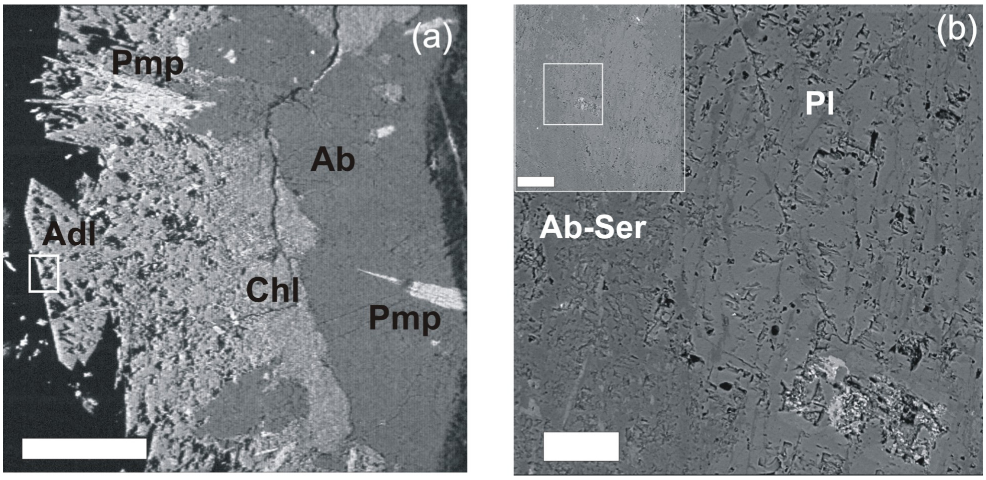

FIGURE 3. *Scanning electron micrograph (secondary electron images) of adularia in amygdules from lavas of the Lower Cretaceous in the Coastal Range of central Chile (modified from Fuentes et al., 2005). (a) Sample with adularia (Adl) in the inner part of an amygdule filled by pumpellyite (Pmp), chlorite (Chl) and an outer rim of albite (Ab). Scale bar=20 µ. (b) Detail of strongly sericitized plagioclase phenocryst (small inset, scale bar=200 µ). Darker gray corresponds to a fine intergrowth of albite-sericite (Ab-Ser) and light gray to plagioclase relict. Scale bar=50 µm.*

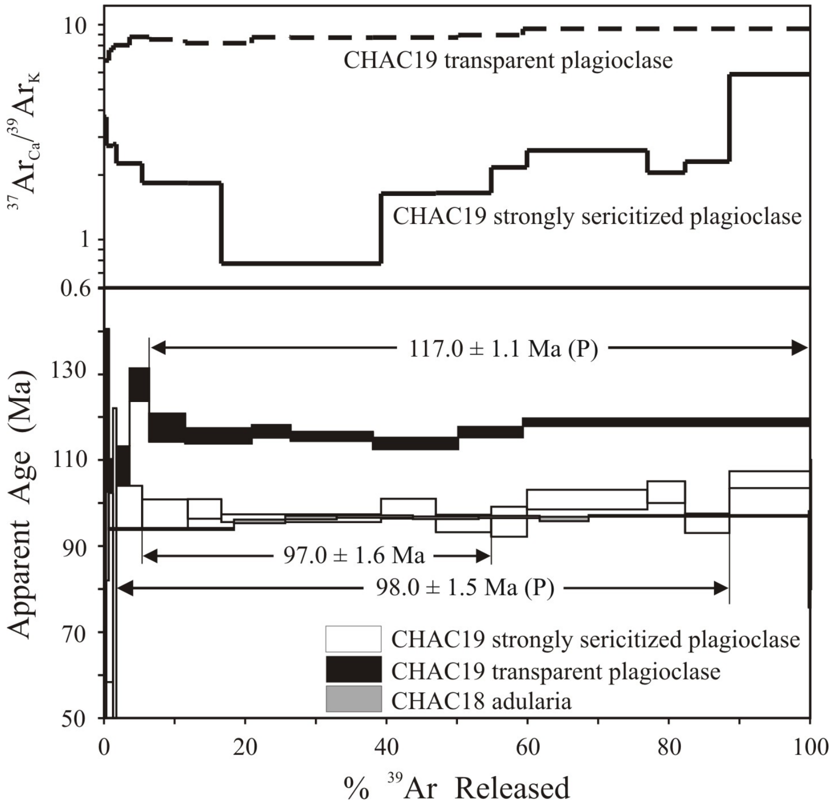

FIGURE 4. *$^{40}Ar/^{39}Ar$ age and $^{37}Ar_{Ca}/^{39}Ar_K$ ratio spectra obtained on a cluster of 32 grains of fresh plagioclase, single grains of strongly sericitized plagioclase and one adularia single grain (adapted from Fuentes et al., 2005).*

sericite of Foland et al. (1992), interpreted by these authors as consequence of ^{40}Ar/^{39}Ar heterogeneities due to ^{39}Ar redistribution (for example from high- to low-K regions or grains) during irradiation, probably related with the fine grain size of dated samples (Figure 3b).

The ^{40}Ar/^{39}Ar methodology has also been applied recently by Oliveros et al. (2007b) to date different low-T alteration events in Jurassic metabasaltic-andesites in the Coastal Range of northern Chile on which Cu-sulphide mineralizations are present. Analyzed alteration minerals such as adularia, sericite, and actinolite apparently give valid ^{40}Ar/^{39}Ar plateau and miniplateau ages. These results allowed Oliveros et al. (2007b) to evidence the occurrence of several alteration events and constraint the timing of Cu-minerallization.

Finally, high spatial resolution studies using a ^{40}Ar/^{39}Ar laser-probe technique permit *in situ* ablation of the neoformed/recrystallized minerals related to low-T processes and thereby constrain the age of alteration. Arancibia (2004) applied UVLAMP ^{40}Ar/^{39}Ar laser probe to date neoformed white-mica during mylonitic deformation under low-grade metamorphic conditions in the Coastal Cordillera of central Chile. This technique would provide an important input in the estimation of the time of neoformation minerals, establishing the relationships between dated mineral and petrographic texture.

U-Pb geochronology

There are two naturally occurring isotopes of uranium that give rise to useful geologic chronometers. ^{238}U decays to ^{206}Pb and ^{235}U decays to ^{207}Pb. Each one is an independent isotope decay scheme and each gives rise to an independent age equation:

$$\frac{^{206}Pb^*}{^{238}U} = e^{\lambda_1 t} - 1 \qquad \text{(eq. 5)}$$

$$\frac{^{206}Pb^*}{^{235}U} = e^{\lambda_2 t} - 1 \qquad \text{(eq. 6)}$$

where λ_1 and λ_2 are the two respective decay constants and Pb* refers to radiogenically produced lead. Results are normally plotted on the so called concordia diagram on which it is possible to graphically determine if the two ages calculated by the two independent isotopic systems are the same, and also provides a means for interpreting the significance of the ages if they are not.

Minerals in metamorphic rocks that are routinely used for U-Pb dating are zircon, allanite, monazite, rutile, titanite and garnet. Among these minerals, only titanite is a common phase in very low-grade metabasites. It is commonly accompanied by other metamorphic minerals suitable for isotopic dating which allows cross-check of the chronological results. Titanite normally appears replacing primary Ti-rich oxides, infilling amygdules or in the groundmass of volcanic rocks (Figure 6). However, grain size of titanite in all these microdomains is normally too small (50-100 μm) for mineral separation. To solve this problem, *in situ* petrographic thin section laser ablation–(± multiple collector)-inductively coupled plasma mass spectrometry (LA–(± MC)–ICP-MS) has made significant advances in generating precise and accurate ages (e.g. Simonetti et al., 2006). Geochronological investigation of titanite using LA–ICP-MS offers several advantages over other dating techniques. These include: (1) simple sample preparation procedures, (2) measurement of isotopic ratios at high spatial resolution (20–100μm), (3) rapid analysis, typically on the order of several minutes, and (4) low cost compared with other U–Pb analytical protocols, such as SHRIMP (sensitive high resolution ion microprobe) or ID- TIMS (isotope dilution-thermal ionization mass spectrometry). This technique also provides the opportunity to directly link age information for a particular sample with deformational fabrics, and pressure–temperature data derived from electron microprobe analysis in the same thin section.

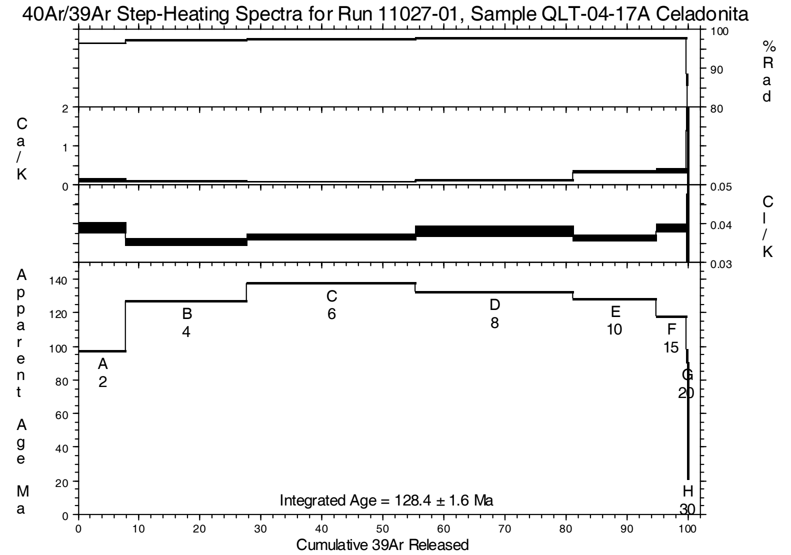

FIGURE 5. *Typical recoil pattern probably by loss of ^{39}Ar due to the very fine-grain of celadonite (<1 μm, Figure 5b) crystals (modified from Morata et al., 2006).*

Titanite typically contains significant quantities of common Pb, therefore the accuracy of U–Pb dates obtained from titanite is critically dependent on the correct assessment of the common Pb component (Simonetti et al., 2006).

Oliveros et al. (2007a) have used LA-MC-ICP-MS to date titanite in an amygdule and in the groundmass (crystals grain size < 200 μm) of Upper Jurassic-Lower Cretaceous basic lava flows metamorphosed in the prehnite-pumpellyite facies from the central Andes. Preliminary results give a mean age of 84.6 ± 2.2 Ma for the titanite in amygdule, with a moderate content of radiogenic Pb in the analyzed minerals indicating that the age obtained is reliable. On the other hand, the small and anhedral titanite crystals in the groundmass for which the content of radiogenic Pb is low, yielded a more imprecise mean age of 108.0 ± 6.4 Ma. However, this last age is in the range of the K-Ar ages of 109 ± 3 and 92 ± 3 obtained for metamorphic celadonite in the same rocks, showing the reliability and power of this technique in the study of very low-grade metamorphism in metabasites.

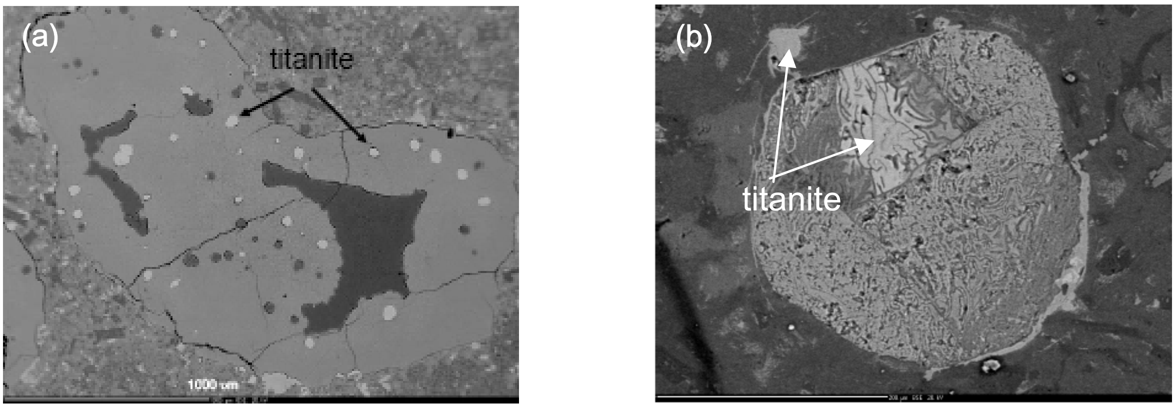

FIGURE 6. Titanite in very low-grade metabasites from the Mesozoic of the Andean Cordillera in central Chile. (a) Titanite grains in a quartz-celadonite amygdule; (b) titanite replacing magmatic Ti-magnetite (from Oliveros et al., unpublished)

From geochronological data to geotectonic significance

The correct interpretation of a radiometric age must be considered based on the closure temperature (*Tc*) of the mineral analysed. Therefore, the measured age of the sample will be the age at which mineral went through its closure temperature. Estimation of *Tc* for each mineral implies the knowledge, among other parameters, of activation energy for chemical diffusion and the size of the diffusion domain in the crystal (see a detailed discussion in McDougall and Harrison, 1989). Unfortunately, for some very low-grade metamorphic minerals used for ~~a~~ geochronological purposes, *Tc* is not well constrained and some assumptions must be necessary.

Once a very low-grade metamorphic region has been dated by one of the methologies previously discussed, some geotectonic implications can be proposed. In this sense, for the case of the Mesozoic volcanic sequences in the Coastal Cordillera of central Chile (Aguirre et al., 1999, Fuentes et al., 2005, Morata et al., 2005), coincidence in the metamorphic and plutonic ages suggests that, additionally to burial, an anomalous thermal gradient reflected by regional magmatism was present during the metamorphic event originating the granitoid plutons. Nevertheless, the epizonal emplacement (*c*. 2.0 kbar) of the plutons characterized by fast cooling and exhumation (Parada et al., 2005) would not be capable to provide the necessary thermal budget to produce by itself the regional metamorphism. On the contrary, the regional magmatic event (and the metamorphism?) would be rather related to asthenospheric upwelling during extension and crustal attenuation taking place during the generation of the Early Cretaceous basins in the region. Moreover, the ^{40}Ar/^{39}Ar methodology allow us to estimate in about 20-22 Ma the time interval between volcanism and metamorphism (Aguirre et al., 1999; Fuentes et al., 2005). In this context, the precise dating of the very low-grade metamorphism permits to constrain the maximum ages of the subsidence and extension (dating the climax of metamorphism) and, consequently, the timing of closure and inversion of basins as a consequence of changes in the regional geodynamic setting from extensional to compressional. Moreover, the long standing process of subsidence and burial metamorphism of volcanic materials in the intra-arc basin of the Coast Range of central Chile might have important metallogenic implications. In fact, dehydration processes due to advanced burial metamorphism might be responsible for the formation of the typical Chilean manto type copper deposits which are particularly concentrated in the Cretaceous rocks from the region studied (see Aguirre et al., 1999).

Conclusions

Precise and reliable isotopic dating in very low-grade metabasites could be obtained by various geochronological methods. The ^{40}Ar-^{39}Ar systematics permits to obtain primary (=magmatic) and metamorphic ages in a same outcrop and even in a same sample. Moreover, the application of the *in situ* LA-MC-ICP-MS in titanite reveals as an extremely useful and powerful tool to constrain the timing of metamorphism.

The combination of precise ages with thermodynamic modelling and calculation of P-T conditions, would lead to a global understanding of the regional very low-grade metamorphic events placing them in a geodynamic context and constraining the evolutionary geological histories in areas affected by such low P-T conditions.

Acknowledgements

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