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K-Ar ages in biotites and K-feldspars from the Catalan Coastal Batholith: Evidence of a post-Hercynian overprinting

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Key words: Granitoids, Hercynian, calc-alkaline, epizonal, geochronology, biotite, chlorite, K-feldspar, resetting, Mesozoic

ABSTRACT

A K-Ar isotopic study was carried out on biotite, K-feldspar and amphibole separates from several of the main acid and intermediate plutonic units of the epizonal late-Hercynian Catalan Coastal Ranges batholith (NE Spain).

According to the study, apparent ages of biotite separates from granites, granodiorites and tonalites, containing more than 7% potassium, ranged from 282 ± 6 to 296 ± 6 Ma with an isochrone age of 284 ± 4 Ma. This value is considered to be the best estimate of a common closure biotite age for all the corresponding intrusions.

In contrast, most of the potassic feldspars showing argon loss to different degrees, gave apparent ages reaching down to 187 ± 4 Ma. This indicates that a slight post-Hercynian disturbing event partially opened the K-Ar feldspar system, but not the biotite system. Nevertheless, K-feldspars from a few localities (even those affected by some deuteric alteration) gave ages close to that of the biotite. Thus, it is suggested that K-feldspar can preserve its radiogenic Ar for a long period of time, as long as it remains unaffected by subsequent processes of recrystallization or overheating.

The disturbing event must have happened after the exhumation of the plutons, since the Lower Triassic rocks lie unconformably over the eroded granitoids (hence they would have been nearly at surface temperature about 250 Ma ago). The age and characteristics of this event are loosely constrained, but the cluster of apparent ages around 200 Ma, together with the fact that K-feldspars younger than 187 Ma have not been found, suggest that it occurred during the Mesozoic.

In addition to argon loss in K-feldspars, a significant increase in apparent ages of chlorite-bearing separates, ranging from 282 ± 6 to 311 ± 6 Ma, was observed in correlation with their chlorite content. We suggest that, in the studied granitoids, chlorite contains extraneous $^{40}Ar^{*}$ whereas pure biotite does not seem to trap significant amounts of radiogenic argon. Consequently, since interlayered chlorite cannot be easily separated from biotite, some K-Ar ages obtained from biotites of regions that have undergone slight thermal or hydrothermal overprints may be considerably increased.

RESUME

Les biotites, les feldspaths et les amphiboles des unités plutoniques acides et intermédiaires hercyniennes tardives formant le Batholite Côtier Catalan (NE de l'Espagne) ont été datés par la méthode K-Ar.

Les âges apparents obtenus sur les biotites contenant plus de 7% de potassium et provenant des granites, des granodiorites et des tonalites sont compris entre 282 ± 6 et 296 ± 6 Ma tandis que l'âge par isochrone est de 284 ± 4 Ma. Cet âge peut être admis comme comme âge de fermeture du système cristallin de la biotite pour toutes les intrusions étudiées.

En revanche, la plupart des feldspaths potassiques présentent différents degrés de perte d'argon radiogénique ayant pour conséquence l'obtention d'âges apparents aussi faibles que 187 ± 4 Ma. On peut en conclure qu'un léger événement thermique ou hydrothermal post-hercynien a partiellement ouvert le système cristallin des feldspaths sans pour autant affecter celui des biotites. Il faut cependant mentionner que certains feldspaths (même ceux présentant les traces d'une altération deutérique) donnent un âge très proche de celui des biotites. Ainsi, les feldspaths sont capables de conserver leur argon radiogénique durant de longues périodes tant qu'ils ne sont pas affectés par des processus de recristallisation ou de réchauffement.

Cet événement perturbant a dû se produire après la remontée en surface des plutons puisque le Trias inférieur repose en discordance sur les granitoïdes érodés. Cela signifie qu'ils étaient froids il y a 250 Ma. L'âge et les conditions P-T de cet événement ne sont pas connus avec précision cependant le regroupement des âges apparents autour de 200 Ma et l'absence de valeurs inférieures à 187 Ma signifie qu'il a dû se produire durant le Mésozoïque.

On observe d'une part la perte de $^{40}Ar^{*}$ par les feldspaths et d'autre part une augmentation significative des âges apparents des concentrés de minéraux contenant de la chlorite qui se situent entre 282 ± 6 et 311 ± 6 Ma. Cette augmentation peut être directement corrélée avec la proportion de chlorite. De fait, les chlorites des granitoïdes doivent contenir de l'argon radiogénique hérité mais ce ne semble pas être le cas pour les biotites. Puisque les chlorites interstratifiées ne peuvent être séparées des biotites, les âges obtenus dans les zones ayant subi une légère influence de caractère thermique ou hydrothermal peuvent être fortement augmentés.

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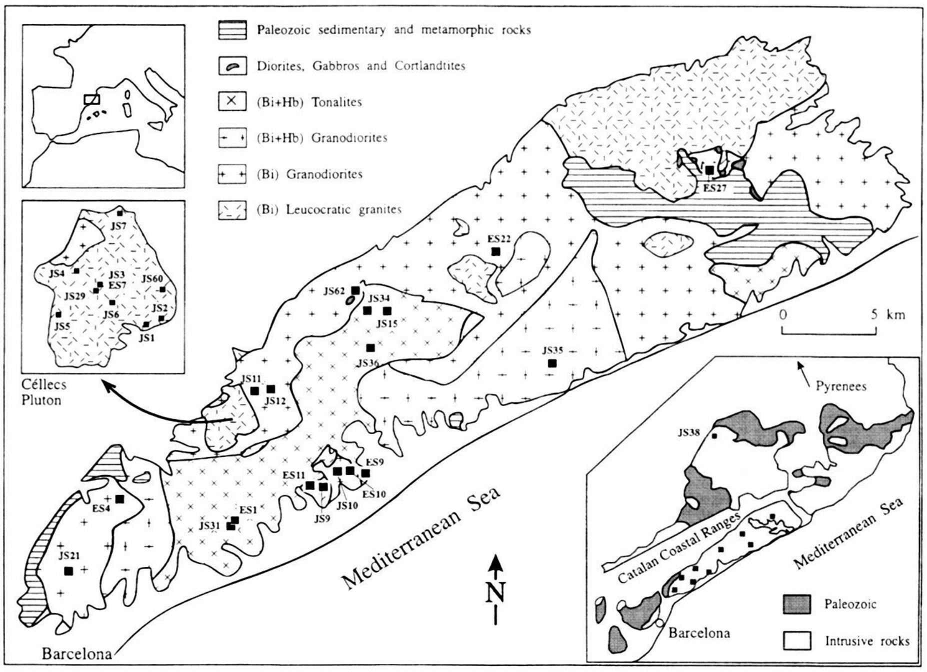

Fig. 1. Geological map of the Montnegre Massif (modified after Enrique, 1990), with the location of the analysed samples.

1. Introduction

The K-Ar isotopic dating method has been widely used for many years in geochronological and cooling-history studies of igneous and metamorphic rocks. Although $^{40}Ar/^{39}Ar$ data generally provide more information than the conventional K-Ar method, the relative simplicity of the latter makes it a useful tool in preliminary studies of geochronologically poorly known areas.

Some of the main problems in the interpretation of data are inherent to the ^{40}K-^{40}Ar system and for this reason they are common to both conventional K-Ar and $^{40}Ar/^{39}Ar$ methods. For example, the relative loss or gain of radiogenic argon in relation to potassium may distort the original igneous or metamorphic cooling ages of the analysed minerals. However, a detailed study of such anomalies may provide very valuable information about the thermal history of those minerals.

Biotite is one of the most frequently used minerals in K-Ar and $^{40}Ar/^{39}Ar$ studies because of its wide distribution, high potassium content and relatively high blocking temperature of about 300°C (e.g. McDougall & Harrison 1988) or 450°C (Villa & Puxeddu 1994). However, this mineral may easily be altered by several natural processes which affect its argon content, and may give rise to misleading interpretations. For example, it has been demonstrated that, during weathering processes Ar and K are progressively lost as biotite is replaced by clay minerals (Mitchell et al. 1988). Likewise, Lo & Onstott (1989, 1995) report that during the chloritization reactions caused by greenschist metamorphism, biotite loses both K and Ar. Moreover, other studies indicate that chlorite can have anomalously high $^{40}Ar/^{39}Ar$ ratios, giving much older ages than the primary biotite (Monié et al. 1994, fig.10).

The study of the original $^{40}Ar^{*}/K$ ratio in partially chloritized biotites using the $^{40}Ar/^{39}Ar$ technique is limited by ^{39}Ar recoil (Lo & Onstott 1989). In this case only integrated ages can be used, making the Ar/Ar method equivalent to the K/Ar method. Consequently, in our opinion, the K-Ar method may be also very useful in the study of the chloritization process of biotite from granitoids.

The main aims of the present study are: 1) to determine biotite, amphibole and K-feldspar K-Ar ages from different intrusive units of the epizonal Hercynian Montnegre Massif (Catalan Coastal Ranges, NE Spain); 2) to determine the effect of chlorite in biotite separates on the apparent age of the latter; and 3) to constrain the age of both chloritization and K-feldspar Ar loss.

2. Geological setting

Two large Hercynian areas are present in the northeastern Iberian Peninsula: the Pyrenean Axial Zone and the northern Catalan Coastal Ranges. They have been isolated from each other and from the main remnants of the Hercynian fold belt (Iberian Massif and French Central Massif) by Alpine tectonics and Tertiary sedimentary basins. For this reason, their real extent and geological links with the neighbouring fragments are, at present, imprecisely known (Julivert & Martínez 1980)

A substantial part of the Hercynian outcrops of the Catalan Coastal Ranges consists of a composite epizonal calc-alkaline batholith emplaced mainly by magmatic stoping. It displays a contact metamorphic aureole which gave rise to different varieties of hornfels in the Cambrian-to-Lower Carboniferous host rocks (Fontboté & Julivert 1954). This thermal metamorphism overprinted a pre-existing low to very low regional metamorphism mainly of greenchist facies. The petrological and geochemical characteristics of the igneous rocks, their large compositional range, and the scarcity of strongly peraluminous types is reminiscent of a continental margin subduction-related plutonism (Enrique 1984, 1990).

The Montnegre Massif (NE of Barcelona) represents about one-third (500 Km^2) of the total exposed surface of the batholith (Fig. 1). It consists of many intrusive plutonic units, normally of very uniform composition, and with brittle-type (sharp and rectilinear) contacts between them. The absence of chilled margins at the contacts suggests that the contrast in temperature with previous adjacent intrusions was small, hence a narrow time span between them can be inferred.

A succession of basic-to-acid intrusions is observed from the relationship of the contacts between different intrusions. These intrusions include quartzdiorites, tonalites, granodiorites and leucocratic monzogranites.

All plutonic units are cross-cut by a dense porphyritic dyke swarm, generally trending ENE to NE. The composition of the dykes is primarily granitic and granodioritic and, to a lesser degree, dioritic.

The batholith and the associated hypabyssal rocks postdate all major Hercynian structures, and are covered by Triassic sediments in some localities. Thus, the intrusion must have occurred at the end of the Carboniferous or during the Permian.

From a morpho-structural point of view, the Montnegre Massif belongs to a series of Neogene horsts and grabens that run parallel to the Mediterranean coast. The grabens are filled with Miocene or younger sediments, whereas the horsts are formed by Paleozoic rocks, covered in some places by Mesozoic or Paleogene sediments (Llopis 1947).

The folding and metamorphism of Paleozoic material is solely due to the Hercynian orogeny (Fontboté & Julivert 1954). Alpine structures are restricted to some Eocene-Oligocene compressional faults on the northern limit of the graben and to normal faults trending NE and NW that define the present day morphology of the Catalan Coastal Ranges. (Anadón et al. 1979; Guimerà & Àlvaro 1994).

The Montnegre Massif forms the North-western margin of the Barcelona Graben. It is characterized by a SE-dipping normal fault system with kilometric slip, attaining 6 Km of normal slip at the Barcelona fault (Roca & Guimerà 1992). The same fault system was active during the Mesozoic extensional tectonics which gave origin to the Iberian Basin (Iberian Range, Ebro Basin, and Catalan Range). The main rift stages (with significant crustal thinning) took place mainly during two periods: Late Triassic- early Lias and late Jurassic-early Cretaceous (Salas & Casas 1993). Between these periods, the fault tectonic activity did not disappear but was very attenuated (Roca et al. 1992).

Tab. 1. Modal analyses (in percent) of Montnegre rocks (after Enrique, 1990). In parenthesis the standard deviation. n=number of samples.

	n	Q	Pl	KF	Bi	Hb	Px	Acc	Chl+Ep
Granites	6	35.3 *(5.2)*	29.3 *(3.6)*	31.8 *(8.5)*	3.2 *(1.3)*	-	-	0.1 *(0.1)*	0.3 *(0.3)*
Granodiorites	14	33.6 *(4.1)*	40.9 *(3.7)*	13.4 *(4.2)*	11.3 *(3.0)*	-	-	0.3 *(0.1)*	0.5 *(0.3)*
Tonalites	15	27.8 *(3.8)*	49.6 *(4.8)*	5.5 *(1.6)*	13.9 *(2.6)*	1.6 *(1.5)*	-	0.4 *(0.2)*	0.8 *(0.7)*
Quartz-diorite	1	9.7	62.1	2.0	13	3.8	7.2	2.2	-
Gabbro	1	0.5	51.1	0.3	-	47.7	-	0.4	-

3. Mineral composition

The rock-types selected for the K-Ar analyses were leucogranites, granites, granodiorites, tonalites and hornblende-gabbros. The primary mineralogy is well preserved in all of the studied rocks and only very minor subsolidus alteration is observed (see modal composition in Table 1).

Biotite is the most characteristic and widespread mafic mineral in Montnegre plutonic rocks (only absent from a few hornblende-gabbros), with a size ranging from 0.1 mm to 10 mm long. Apatite, zircon and ore minerals are frequently found as inclusions. Biotites are of intermediate composition between annite and eastonite. Their Fe/(Fe+Mg) ratio ranges from 0.8 in leucogranites to 0.5 in quartz-gabbros (Solé 1993). Biotite is usually replaced by chlorite in small amounts. The proportions of chlorite in biotite separates were estimated by X-ray diffraction and potassium analysis.

Amphiboles are present in all the more basic rocks and appear in some granodiorites. Their composition is quite homogeneous, consisting mainly of magnesio-hornblendes. Their Mg/(Mg+Fe2+) ratio is between 0.6 and 0.7, being higher only in hornblende-gabbros, where it can reach 0.9 (Solé, 1993).

Potassic feldspars are present in all rocks, from leucogranites to diorites. Their size ranges from 2 to 10 mm. Inclusions are scarce, usually plagioclase or biotite, and occasionally quartz. Both orthoclase and microcline are present, with a composition of Or80–96. Potassic feldspars are generally microperthitic, with albite lamellae (Ab97) of about 10–30 µm wide (Solé 1993).

Plagioclase is always zoned, with more calcic nuclei than edges (An 50–10). They usually show oscillatory and patchy zoning which is more frequent in tonalites and granodiorites.

4. Analytical methods

45 K-Ar analyses on 31 samples were performed (Table 2). Their geological location is shown in Fig. 1. Most of the rock samples (weighing approx. 20 kg) were collected in quarries to prevent surface alteration. Samples were obtained by trituration with steel-jaws, sieving (250, 350 and 500 m), selection of the best fraction (usually the 350–500 µm), washing and separation of the different minerals by magnetic methods (Isody-

namic Frantz® separator), heavy liquids (bromoform and methylen iodate) and hand picking.

The K content of each mineral separate was measured by flame spectrophotometry. Duplicated analyses of 100 mg of sample were carried out. Analytical precision was better than 2%.

25 to 30 mg samples were degased under vacuum at approx. 100°C for ten to twelve hours before analysis to reduce atmospheric contamination. Argon was extracted during complete sample fusion in a Mo crucible heated by a radiofrequence furnace and mixed with a known amount of ^{38}Ar spike and purified in a pyrex glass extraction line.

Measurements were done in static mode with an AEI MS-10 mass-spectrometer with a permanent magnet of 4.1 kG and connected to a computer for data processing. Analytical precision on ^{40}Ar and ^{38}Ar peak heights was higher than 0.5%; and 1% on ^{36}Ar. The Ar/Ar analysis followed the analytical techniques described in Cosca and O'Nions (1994).

K-Ar analyses were done at the Département de Minéralogie, Université de Genève (Switzerland). The Ar-Ar analyse was performed at the Institut de Minéralogie, Université de Lausanne (Switzerland). Ages were calculated using the constants recommended by Steiger & Jäger (1977). All errors are quoted at one sigma level.

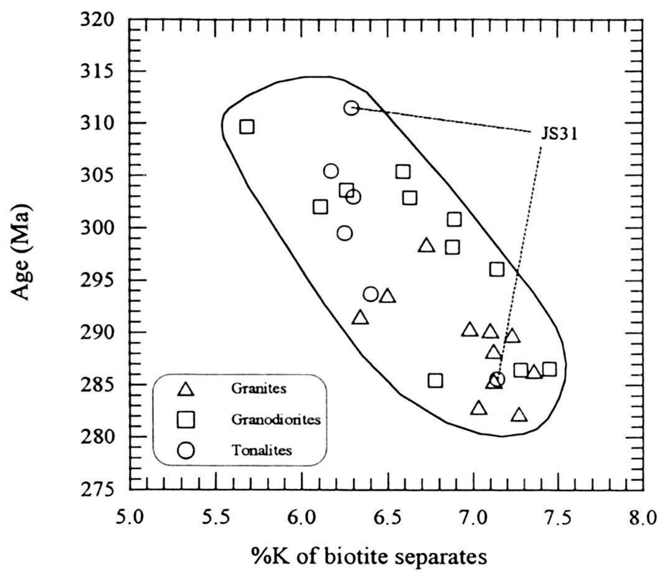

Fig. 2. Correlation between age and potassium content in biotite separates. An increase in age is observed as potassium content decreases. The plotted lines connect two separates of the same sample JS31 with different chlorite content.

5. Results and discussion

5.1. *K-Ar analyses of amphiboles*

The two amphibole separates analysed, belong to a quartz-diorite (JS62) and a hornblende-gabbro (ES27). Both amphiboles gave the same age: 293 ± 7 Ma. This age is higher than the biotite age of intemediate-acid rocks, but taking error into account, it is analytically indistinguishable from these, as discussed below. Field evidence suggested that these rocks may represent the oldest intrusions in the area, but this assumption can not be proved with these data. Moreover, the K-Ar age in amphiboles has a blocking temperature of about 500°C (McDougall & Harrison 1988) and should be slightly older (Cosca and O'Onions, 1994).

5.2. *K-Ar analyses of biotites*

28 biotite separates (Table 2) containing variable amounts of chlorite were obtained from leucocratic granites, granodiorites, tonalites and quartzdiorites. Their apparent ages cover a considerable time span from 282 to 311 Ma. There is no relationship between geographical situation or composition of the rocks and the ages of biotite separates. Even samples belonging to the same intrusion may have different ages. However, a plot of the potassium content of biotite separates versus their apparent ages (Fig. 2) clearly shows that the age decreases statistically with increasing K content.

Biotite microprobe analyses (Solé 1993) of all sampled rock types indicate potassium contents between 7.5 and 8.1%. K values from separates below 7.5% do not correspond to pure biotite compositions; the low K content is due to the presence of other minerals. These contaminating minerals must contain extraneous argon (in the sense defined by McDougall & Harrison 1988, p. 11). If not, they would act only as diluent preserving the age (the lack of potassium would be accompanied by a paucity of argon). The XRD spectra of some biotite separates indicate that chlorite is by far the prevailing contaminant mineral (Fig. 3). No weathering mineral phases were found, such as kaolinite, vermiculite, etc., comparable to those reported in Mitchell et al. (1988). A semiquantitative chlorite determination (Fig. 4) was done using the (002)chlorite/((002)chlorite+(001)biotite) values from these spectra (Lo & Onstott 1989; Ruffet et al. 1991). The XRD-measured separates contain between 0.5 and 10% chlorite.

Provided that an inverse correlation between %K content in biotite separates and %chlorite is also observed, the latter could be the main phase responsible for the decrease in potassium in the separates. Thus, the existence of extraneous argon in chlorite seems to be the most probable explanation for the correlation between K concentration and age.

To illustrate this point, two biotite fractions with different chlorite contents were separated from tonalite JS31, in order to control the influence of this mineral on their apparent ages. The first fraction had 6.29%K and an apparent age of 311 Ma. The other had 7.14%K and an age of 286 Ma. If pure biotite contains no extraneous argon, the lower age obtained, that corresponding to the low chlorite-bearing separate, would be a better approach to the cooling age of this rock.

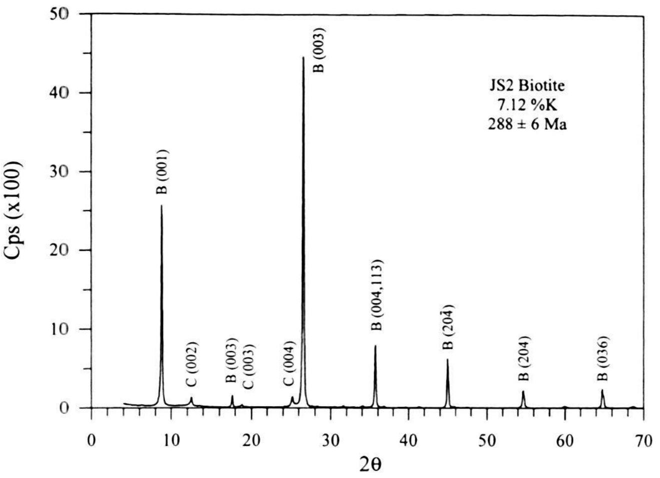

Fig. 3. X-ray spectra of biotite separate JS2. B and C represent the X-ray reflections for biotite and chlorite, respectively.

Similar results were obtained for biotite separates from the main leucogranitic intrusions studied in this area (Fig. 1). These separates also displayed variable apparent ages ranging from 282 to 294 Ma, but the average age obtained from the samples with more than 7% K was 286 ± 6 Ma. The same results are valid for other K-rich biotite separates from granodiorites and tonalites: for example, JS11 (7.45% K, 287 Ma) and JS12 (7.28% K, 286 Ma).

A $^{40}K/^{36}Ar$-$^{40}Ar/36Ar$ isochron (Fig. 5) including all the biotite separates with less than 7%K gave an age of 298 ± 3 Ma. The quality of the isochron given by its low MSWD would suggest that it is a good isochron. However, as has been previously discussed, the presence of extraneous argon in chlorite-bearing samples implies that the age obtained is overestimated. A second $^{40}K/^{36}Ar$-$^{40}Ar/^{36}Ar$ isochron (Fig. 5) including all the most potassium-rich (>7%K) biotite separates (11 samples) gave an age of 284 ± 4 Ma, a value considered to be the best estimate of a common closure biotite age for all the corresponding intrusions.

This conclusion is consistent with the ^{40}Ar-^{39}Ar biotite data obtained by Solé (1993). The Ar/Ar spectra of the JS31 biotite is show in Fig. 6, which gives a total fusion age of 282.2 ± 1.0 Ma and a plateau-age of 286.1 ± 1.4 Ma. A Rb/Sr mineral isochron on tonalite A-13 (equivalent to sample JS31) gives a similar age of 284±7 Ma (Del Moro & Enrique 1996). Also, the only muscovite analysed (JS60), belonging to a leucocratic granite, give an age of 281 ± 6 Ma.

5.3. K-Ar analyses of feldspars

Fourteen K-feldspars were analysed. The ages range from 187 Ma in sample JS2 to 278 Ma in sample JS21 (Table 1). There appears to be no correlation between the feldspar ages with

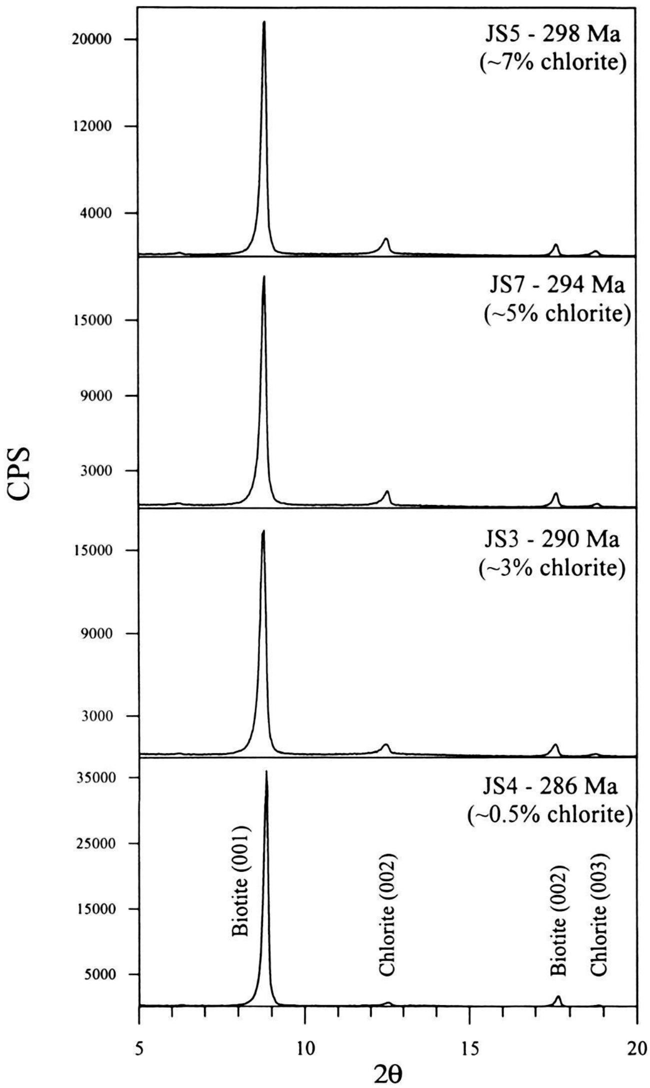

Fig. 4. Comparative X-ray spectra of biotite separates from Céllecs pluton.

the bulk composition of the rock and their geographical location.

Feldspars usually give lower ages than biotites or amphiboles. It could be concluded that they did not retain argon and give ages without geological signification. The first experiments on diffusion in feldspars showed that argon migration in these minerals seems to obey diffusion laws. Experiments have proved that potassic feldspars do not lose argon at ambient temperatures if unaffected by thermal, tectonic or fluid circulation processes (e.g. Foland 1974; Harrison & McDougall 1982).

There are also several theoretical studies regarding argon diffusion in potassic feldspars, which are based on isotopic studies of geologically well known areas (Hart 1964; Hanson &

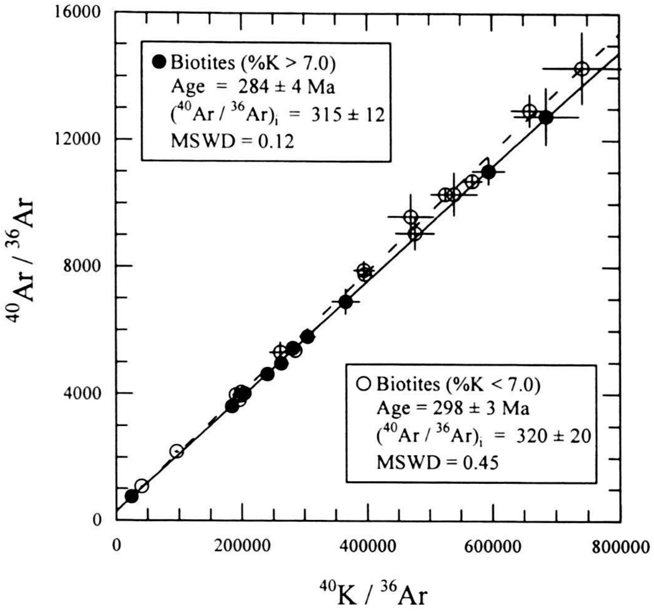

Fig. 5. $^{40}K/^{36}Ar$ – $^{40}Ar/^{36}Ar$ isochrons of biotite separates from granites, granodiorites and tonalites. The error bars are equal to 1σ. Regression lines calculated after York (1969).

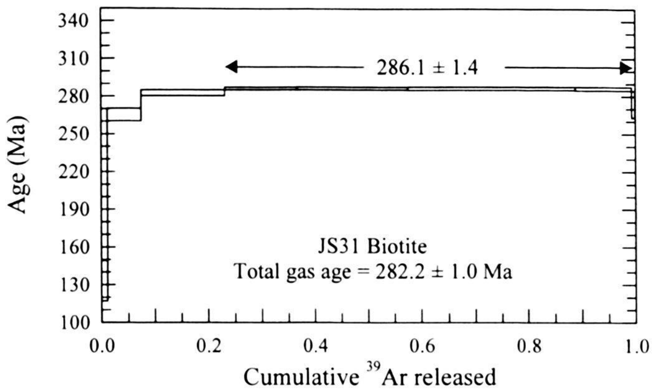

Fig. 6. $^{40}Ar/^{39}Ar$ spectra of biotite from tonalite JS31.

Gast 1967; Harrison & McDougall 1985). The predictions of these studies are consistent with experimental data.

Since the crystalline structure of feldspars is diverse, there is no clear agreement as to which factors affect argon diffusion. It seems that the principal control on argon diffusion is exercised by microporosity (Parsons et al. 1988; Burgess et al. 1992) and by the presence of microperthites (Foland 1974; McDougall & Harrison 1988; Shibata et al. 1994).

Most of the K-feldspars we analysed showed an important argon loss. However, the presence of feldspars (JS21, JS38) with ages close to the biotite ages suggests that they have retained almost all their radiogenic argon (unless these K-feldspars had a late argon excess).

The analysed K-feldspars show a slight turbidity, which has been considered an important cause of argon loss (Parsons et al. 1988). All the samples have a similar degree of turbidity, even the granodiorite JS21 and leucogranite JS38, which have the oldest potassic feldspar ages determined. Therefore we can infer that this turbidity is not the main cause for the age difference among potassic feldspars from the Montnegre Massif, and that turbidity per se does not imply argon loss, as stated by Burgess and Parsons (1994).

Thermal, tectonic or fluid circulation processes, can lead to an Ar loss. (Albarède et al. 1978; Maluski 1978; Parsons et al. 1988). Processes that cause argon loss in Montnegre Massif cannot be mainly due to pervasive fluid circulation since these usually produce great changes in rock mineralogy. The only post-magmatic phenomenon clearly visible is the minor replacement of biotite with chlorite, the turbidity of K-feldspars, and plagioclase sericitization, all of which are of slight intensity (see Table 1). These mineralogical changes could have been produced during the cooling of the massif (i.e. deuteric) without necessarily invoking a late hydrothermal alteration. In addition, K-feldspars and plagioclase do not show petrographic evidence of recrystallization. Tectonic processes can also be ruled out since the Montnegre massif shows no significant post-Hercynian deformation. Alpine deformation (Paleogene) is restricted to a narrow band along the sinistral convergent wrench-fault in the Northern border of the pre-Littoral graben (Guimerà & Álvaro 1994).

A slow cooling rate of the batholith which would allow the temperature to decrease gradually between 284 and 186 Ma ago (the ages of biotites and youngest K-feldspars, respectively) can be excluded because field data prove that these granites were at the surface before Triassic times, since they are locally covered with lower Triassic sediments (Buntsandstein facies). It is now possible to conclude that a slight thermal post-Permian event with only minor fluid circulation was responsible for the resetting of K-feldspar ages. This event is considered slight because it opened the K-Ar K-feldspar system but not the biotite system.

6. Significance of biotite and K-feldspar K-Ar ages

The interpretation of excess argon in chlorite is strongly dependent on the time of chlorite formation. The plot of the data from biotite separates in the ^{40}K-^{40}Ar diagram (Fig. 7) shows a broad alignment with a non-zero intercept. Following Harper's model (Harper 1970), the intercept of this line indicates the mean quantity of excess argon (~1 nmol/g) and has an isochron meaning, its apparent ages being equivalent to the time of incorporation of excess argon. However, in our opinion, this line is only a true isochron in the case of a pseudomorphism of biotite by chlorite in which potassium loss was produced without argon loss.

If we consider a biotite with 7.5%K content at the time of its formation t0 (point A in Fig. 7), point B would represent

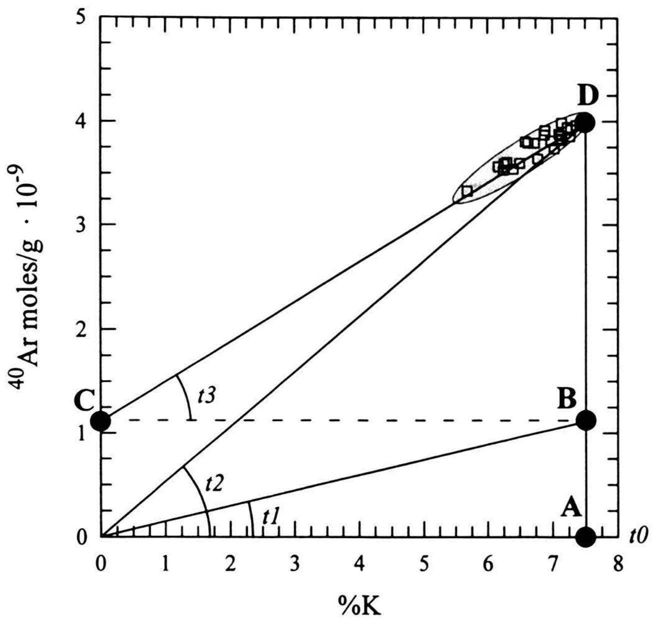

Fig. 7. %K – ^{40}Ar* schematic diagram showing a model for extraneous argon accumulation in chlorite during their formation. Legend: (A) biotite at formation time t0, (B) biotite at time t1, (C) chlorite formed at time t1 and (D) biotite at time t2. See text for detailed explanation.

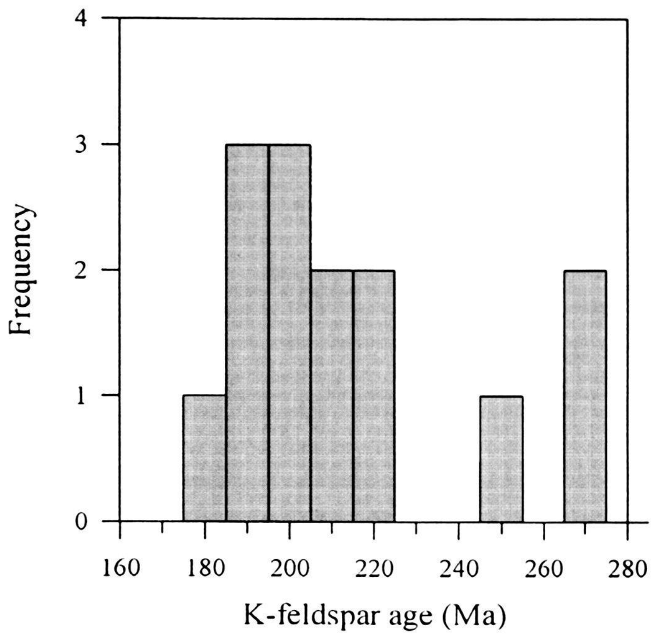

Fig. 8. Histogram of K-feldspar ages.

the radiogenic argon formed in the biotite at time t1. If chlorite formed at this time (t1) by pseudomorphism of biotite without Ar loss, chlorite will be represented by point C. From time t1 to the present time (t2) the radiogenic argon of biotite increases from point B to point D. During the same time chlorite maintains the same argon in point C, since pure chlorite has no potassium. Therefore, the line C–D, defined by the mixtures biotite-chlorite, would represent the isochrone which gives us the age of the chloritization (before present, t3).

In our case, as we deal with mixtures biotite-chlorite, the above model would be applicable if chlorite formed without Ar loss. In this case the poorly defined regression line would indicate an age of about 200 Ma (Fig. 7). However, the spread of the data suggests that another explanation is also possible: chlorite has a deuteric origin (formed during the late stages of cooling of the batholith), and the extraneous argon must have been incorporated in a later event (excess argon). In this case the amount of ^{40}Ar would depend on several factors which include the Ar retention capacity of chlorite and the degree of external overpressure. As this excess argon would be randomly produced, the slope in the biotite-chlorite regression line would have no significant meaning.

In most cases, biotites submitted to hydrothermal processes may suffer from argon loss (e.g. Criss et al. 1982), and biotites submitted to high to moderate pressure often contain extraneous argon (e.g. Brewer 1969; Giletti 1971). However, in Montnegre, epizonal rocks have extraneous argon in their chlorites, but biotite has not been affected. It is important to note that the biotite closure temperature has not been reached in Montnegre samples, as opposed to the case of granites of Central Swiss Alps (Dempster 1986) and eastern Taiwan (Lo and Onstott, 1989), in which a minimum temperature of 300°C was reached in a greenschist facies metamorphism.

A comparison is possible between the ages of K-feldspars and those of biotites, since two K-feldspar samples (granodiorite JS21 and leucogranite JS38) have ages of 278 ± 6 Ma and 277 ± 6 Ma, respectively, very close to that estimated for biotites with no extraneous argon (284 ± 4 Ma). If these K-feldspars have no argon loss or excess, as we assume because the ages are very closely together, this would imply that the cooling rate of the pluton was at least 20°C/Ma.

As discussed above, the argon loss in K-feldspars must have occurred later than the lower Triassic. The time of this argon loss is not well constrained, but two hypotheses can be put forward: (i) The disturbance was produced at the age registered by the youngest feldspars, when they lost almost all their radiogenic argon; (ii) The disturbance is more recent than the youngest feldspars and they only underwent a partial loss of argon.

In the first case, the event can be dated as Jurassic. In the second case, the age could be related to the late Mesozoic rifting stages (Salas & Casas 1993) or to the Paleogene compressive faults that formed the present-day Catalan Coastal Ranges. We favour the first alternative, since a thermal event responsible for argon loss happening about 30 to 40 Ma ago (Alpine compression) would probably produce a scattering of

Tab. 2. K-Ar analyses. Biotites are ordered by decreasing potassium content. K-feldspars are ordered by increasing age. 40Ar* is the radiogenic argon corrected for atmospheric contamination and mass fractionation.

Sample	Rock	%K	$^{40}Ar^*$ moles/g x 10^{-9}	$^{40}Ar^*$ %	$^{40}K/^{36}Ar$ x 10^3	$^{40}Ar/^{36}Ar$	Age (± 1σ)
Amphiboles							
JS62	Quartz-diorite	2.05	1.129 ± 0.019	90.8	157 ± 3	3198 ± 25	293 ± 7
ES27	Hornblende-gabbro	0.315	0.174 ± 0.001	70.3	38 ± 0.4	996 ± 4	293 ± 5
Muscovites							
JS60	Granite	8.12	4.275 ± 0.051	97.6	680 ± 25	12292 ± 349	281 ± 6
Biotites							
JS11	Granodiorite	7.45	4.013 ± 0.026	95.7	366 ± 22	6903 ± 387	287 ± 6
JS4	Granite	7.36	3.961 ± 0.026	94.9	305 ± 12	5796 ± 189	286 ± 6
JS12	Granodiorite	7.28	3.920 ± 0.023	97.3	595 ± 26	11025 ± 413	286 ± 6
JS9	Granite	7.27	3.852 ± 0.024	94.0	263 ± 11	4964 ± 175	282 ± 5
JS3	Granite	7.23	3.942 ± 0.030	92.7	204 ± 10	4022 ± 169	290 ± 6
JS31	Tonalite	7.14	3.832 ± 0.033	93.6	241 ± 9	4625 ± 140	286 ± 6
ES22	Granodiorite	7.14	3.985 ± 0.022	60.6	24 ± 1	751 ± 2	296 ± 6
JS1	Granite	7.12	3.818 ± 0.028	91.8	184 ± 5	3608 ± 73	285 ± 6
JS2	Granite	7.12	3.860 ± 0.024	97.7	686 ± 51	12758 ± 900	288 ± 6
JS29	Granite	7.10	3.877 ± 0.026	94.6	281 ± 7	5441 ± 86	290 ± 6
JS6	Granite	7.03	3.735 ± 0.023	94.5	285 ± 7	5365 ± 51	283 ± 5
Biotites + Chlorites							
ES7	Granite	6.98	3.814 ± 0.025	97.2	569 ± 16	10713 ± 193	290 ± 6
JS34	Granodiorite	6.89	3.912 ± 0.026	97.1	526 ± 15	10296 ± 187	301 ± 6
ES4	Granodiorite	6.88	3.869 ± 0.024	96.2	396 ± 9	7765 ± 82	298 ± 6
JS10	Granodiorite	6.78	3.637 ± 0.022	92.3	196 ± 5	3816 ± 60	285 ± 5
JS5	Granite	6.73	3.788 ± 0.024	97.9	742 ± 61	14291 ± 1133	298 ± 6
JS32	Granodiorite	6.63	3.792 ± 0.023	97.7	660 ± 29	12945 ± 506	303 ± 6
ES9	Granodiorite	6.59	3.803 ± 0.021	92.6	190 ± 5	3978 ± 64	305 ± 6
JS7	Granite	6.50	3.594 ± 0.023	97.1	540 ± 37	10297 ± 674	294 ± 6
JS62	Quartz-diorite	6.40	3.540 ± 0.058	92.5	197 ± 6	3951 ± 64	294 ± 7
ES11	Granite	6.34	3.479 ± 0.024	96.7	477 ± 31	9063 ± 505	292 ± 6
JS15	Tonalite	6.30	3.604 ± 0.052	94.4	261 ± 17	5301 ± 314	303 ± 7
JS31	Tonalite	6.29	3.709 ± 0.025	96.9	470 ± 37	9585 ± 718	311 ± 6
JS35	Granodiorite	6.26	3.590 ± 0.022	96.3	395 ± 16	7888 ± 279	304 ± 6
JS36	Tonalite	6.25	3.531 ± 0.062	92.7	199 ± 8	4060 ± 134	300 ± 7
ES1	Tonalite	6.17	3.561 ± 0.022	72.4	40 ± 0.8	1072 ± 3	305 ± 6
JS21	Granodiorite	6.11	3.479 ± 0.017	96.1	377 ± 8	7490 ± 64	302 ± 6
ES10	Tonalite	5.68	3.328 ± 0.018	86.5	96 ± 2	2180 ± 22	310 ± 6
K-Feldspars							
JS2	Granite	10.50	3.582 ± 0.021	96.6	729 ± 24	8611 ± 219	187 ± 4
JS7	Granite	11.00	3.844 ± 0.032	95.3	515 ± 27	6329 ± 297	191 ± 4
JS9	Granite	11.70	4.145 ± 0.001	92.6	313 ± 12	4017 ± 198	194 ± 5
JS3	Granite	10.80	3.849 ± 0.023	96.9	786 ± 28	9679 ± 285	195 ± 4
JS1	Granite	7.56	2.774 ± 0.018	49.9	24 ± 0.5	590 ± 1	200 ± 4
JS34	Granodiorite	8.53	3.149 ± 0.048	90.2	221 ± 6	3026 ± 13	201 ± 5
JS4	Granite	10.80	4.100 ± 0.031	98.1	1183 ± 48	15285 ± 520	207 ± 4
JS35	Granodiorite	11.30	4.505 ± 0.052	97.0	702 ± 20	9673 ± 156	216 ± 4
JS31	Tonalite	7.13	2.867 ± 0.022	95.0	417 ± 13	5918 ± 138	218 ± 4
JS5	Granite	11.50	4.735 ± 0.032	96.4	581 ± 15	8312 ± 116	223 ± 4
JS6	Granite	11.20	4.693 ± 0.032	95.2	420 ± 10	6167 ± 80	227 ± 5
JS12	Granodiorite	10.40	4.861 ± 0.056	95.6	406 ± 11	6658 ± 79	251 ± 6
JS38	Granite	9.53	4.941 ± 0.033	92.1	163 ± 4	3131 ± 10	277 ± 6
JS21	Granodiorite	11.10	5.681 ± 0.031	95.1	331 ± 8	5975 ± 69	278 ± 6

feldspar ages distributed over a long period of time, between 280 and 30 Ma. Although it is difficult to constrain the age at which the K-feldspars lose their argon, there is a grouping of ages in the Lower Jurassic (Fig.8).

Geological constraints indicate that the thickness of sediments above the Montnegre Massif during Mesozoic times was probably small. Thus, because a regional disturbance does exist (affecting at least 500 km2), with a heterogeneous geographical distribution (since very close samples give different ages), and there is no evidence of significant signs of alteration in these rocks, we propose a thermal heating induced by hydrothermal fluids circulating through a close-spaced fracture system as the main cause of this disturbance. These fluids would alter only narrow bands of rock along the fractures leaving the rest of the intrusive bodies unaltered.

These fractures were probably originated in late-Hercynian times since they show the same trending as the prominent granitic dyke swarm that cuts the batholith (Enrique 1990). The origin of the fluids may be related with the Mesozoic rifting in the Catalan Range (Salas & Casas 1993).

The apparent ages of the youngest analysed feldspars are similar to other significant geological ages found in the Iberian Peninsula. In the Pyrenees, Montigny et al. (1983) studied the magmatism (ophites and lavas) associated with Triassic sediments and obtained ages of 195 ± 8 and 197 ± 7 Ma for the ophites and 210 ± 6 and 215 ± 7 Ma for the lavas. In the Los Pedroches Batholith (Central Iberian System) Halliday & Mitchell (1984) found ages of 210–230 Ma for the mineralizations related to the Opening of the Atlantic. Recent work using the $^{40}Ar/^{39}Ar$ technique (Sebai et al. 1991) has confirmed that the great basic dyke of Plasencia shows ages around 200 Ma. This magmatism has been related to the opening of the Central Atlantic Ocean during Liassic times, associated to a major rifting. The period around 180–190 is known to have been an important time of fault reactivation which was probably linked to increased Kimmerian tectonic activity (Schaltegger et al. 1995). The rifting tectonics associated with these phenomena was the same that formed the Iberian Basin and this is consistent with the apparent ages of K-feldspars, as mentioned above.

7. Conclusions

The Montnegre Massif gave a K-Ar biotite age of 284 ± 4 Ma for leucogranites, granodiorites and tonalites. The amphiboles, from gabbros and quartz-diorites, gave an age of 293 ± 7 Ma. Thus, cooling in the range of 300–500°C probably occurred between 293 and 284 Ma ago.

The presence of chlorite in biotite separates increases the apparent age more or less proportionally to the biotite/chlorite ratio. This implies the existence of extraneous argon located in chlorite. The most accurate ages are those from separates with high potassium contents (i.e., those with a minimum amount of chlorite).

The extraneous argon in chlorite can be attributed to either post-Hercynian chloritization without significant Ar loss or, capture of radiogenic argon by chlorite.

Most of the potassic feldspars underwent argon loss, which was probably caused by a slight regional, but inhomogeneously distributed, thermal event with minor fluid circulation. This disturbing event probably occurred during Mesozoic times.

Some potassic feldspars have preserved nearly the same ages as those of the biotites, indicating that K-feldspar can preserve its radiogenic Ar for a long period of time, as long as it remains unaffected by subsequent processes of recrystallization or overheating.

The K-Ar system in pristine biotites of Montnegre Massif was not opened. Therefore, both argon loss in K-feldspars and the origin of the extraneous argon in chlorite occurred below the closure temperature of biotite.

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