Heavy mineral composition and geochemistry of the Weald facies from the Maestrat Basin (Spain): Provenance implications for Late Jurassic-Early Cretaceous rifting stage

Composición y geoquímica de los minerales pesados de las facies Weald de la Cuenca del Maestrazgo (España): Implicaciones para la procedencia durante la etapa de rifting Jurásico Superior-Cretácico Inferior

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RESUMEN

Las areniscas en facies Weald de la Cuenca del Maestrazgo fueron depositadas durante la etapa de rift Jurásico superior-Cretácico inferior. Una característica de las cuencas intracratónicas desarrolladas durante periodos de rifting es la presencia de varias potenciales áreas fuentes que pueden aportar sedimentos a la vez. Este hecho complica la interpretación de la procedencia usando los modelos clásicos a escala tectónica. En este trabajo, la geoquímica de roca total se relaciona con el análisis químico en minerales pesados y con el análisis modal para llegar a discriminar la procedencia de areniscas syn-rift.

La composición modal y los análisis de roca total de las areniscas estudiadas indican una composición arcósica-subarcósica. Las areniscas de la Fm. Mora, en la base, presentan la mayor abundancia en minerales pesados y el contenido más bajo en feldespatos (menor contenido en Rb). Por el contrario, las areniscas de la Fm. Camarillas, en el techo, reflejan un incremento en Rb relacionado con un aporte mayoritario de feldespato-K. La composición química de las turmalinas analizadas sugiere una procedencia mixta, a partir de granitos (turmalinas ricas en Fe) y rocas metamórficas (turmalinas ricas en Mg) para todas las unidades estudiadas. Esta interpretación es consistente con la mayor abundancia de fragmentos de roca metamórficos observados en las formaciones Mora y El Castellar y la presencia de abundante feldespato-K y fragmentos de roca plutónicos en la Fm. Camarillas.

La sedimentación de las areniscas de la Fm. Camarillas coincidió con el levantamiento y erosión del Macizo Ibérico. Para este momento, el área fuente principal estaba constituida por rocas plutónicas, lo que implica una dilución significativa de las áreas fuentes metamórficas, que fueron dominantes durante la sedimentación de las areniscas inferiores de las formaciones Mora y Castellar.

Key words: provenance, sandstones, heavy minerals, geochemistry, Lower Cretaceous, Iberian Chain

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Introduction and geological setting

One of the most typical characteristics of intracratonic basins developed during *rifting* stages is the occurrence of several source areas, which can provide detrital sediments at the same time and therefore complicate provenance discrimination. Frequently, the application of classical tectonic scale models in this geological setting is not enough discriminating of provenance. Whole-rock geochemical analyses are usually applied to provenance studies because of the rapid acquisition of a large number of variables with high precision. Moreover, it is an useful tool due to the high discrimination capacity between source rocks of contrasting composition (e.g., McLennan, 1989; Welje and von Eynatten, 2004). Single-grain electron microprobe chemical analyses in heavy minerals are applied to the study of compositional differences between mineral assemblages introduced by provenance source rocks (von Eynatten and Gaupp, 1999). In the present study, we apply whole-rock geochemistry and chemical analyses in heavy minerals in order to discriminate provenance of Weald facies sandstones deposited during the Late Jurassic-Early Cretaceous *rifting* stage.

The study area is located in the Iberian Peninsula (Spain), in the eastern part of the Iberian Chain (Fig. 1-A), which correspond to the Maestrat Basin (Fig. 1-B). The development of this intracratonic basin is related to the opening of the Tethys in the eastern part of the Iberian Peninsula. Maestrat Basin contains up to 5.8 km of Mesozoic sediments, dominantly carbonates. Two main rifting stages took place in this basin (Salas and Casas, 1993; Salas *et al*., 2001). The first stage during Triassic age produced high angle normal faults in the Paleozoic basement. The second stage, during the Upper Jurassic-Lower Cretaceous is related to the opening of the North Atlantic, which created an extensional syn-sedimentary fault system, which divided the Maestrat Basin into several sub-basins (Salas and Guimerà, 1996). This paper is focused in the western most sub-basins (Aliaga, Galve and Penyagolosa) where siliciclastic sediments

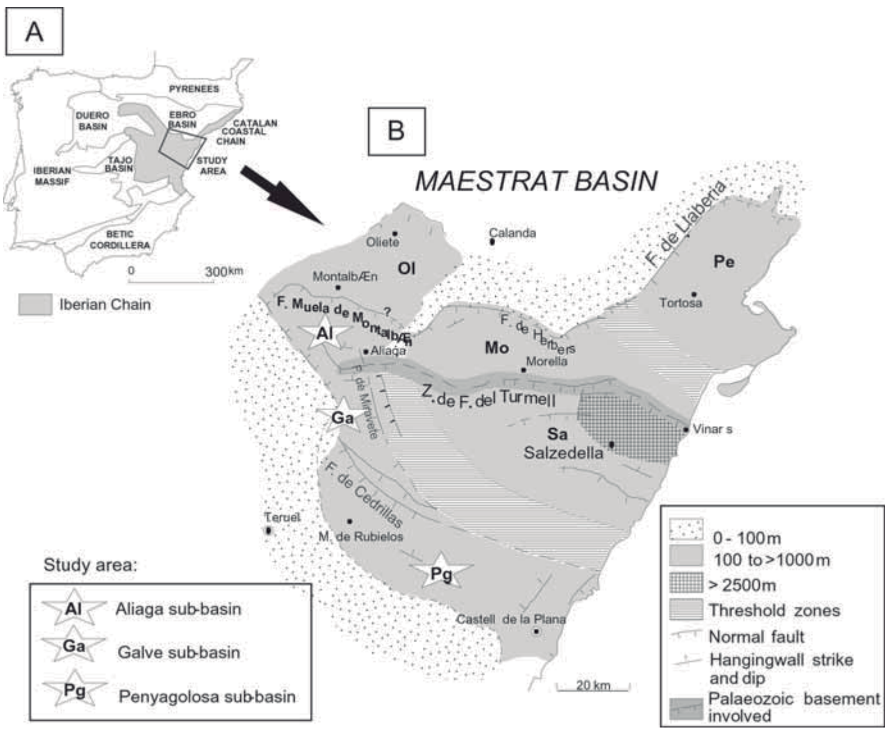

Fig. 1.- (A) Simplified map of the Iberian Peninsula showing the main structural units. (B) Detail corresponding to the Maestrat Basin located in the Iberian Chain (modified from Salas *et al.*, 2001). Note the seven sub-basin in which Maestrat Basin have been divided: Oliete (Ol), Morella (Mo), Perelló (Pe), Salzedella (Sa), Penyagolosa (Pg), Galve (Ga) and Aliaga (Al). The last three sub-basins are the study area.

Fig. 1.- (A) Mapa simplificado de la Península Ibérica mostrando las principales unidades estructurales. (B) Detalle de la zona de estudio, con la localización de la Cuenca del Maestrazgo dentro de la Cadena Ibérica (modificado de Salas* et al.*, 2001). Quedan señaladas las siete subcuencas en que ha sido dividida la Cuenca del Maestrazgo: Oliete (Ol), Morella (Mo), Perelló (Pe), Salzedella (Sa), Penyagolosa (Pg), Galve (Ga) and Aliaga (Al). Las tres últimas corresponden al área de estudio.

are dominant. These sub-basins were filled by fluvial sediments with intercalations of lacustrine carbonates, evolving towards the top to marine carbonates (Salas, 1987). The boundaries between the studied stratigraphic units (Mora, El Castellar and Camarillas) are regional unconformities resulting in three depositional sequences. Major facies changes and stacking pattern of parasequences reveal the presence of three 2nd-order sequences (1-3, Fig. 2).

Methodology

A total of 125 sandstones from Mora, El Castellar and Camarillas formations were sampled in 10 stratigraphic sections. Modal analyses were performed in 49 fine to medium size sandstones (0.17 to 0.35 mm) following the «classical or genetic» approach and counting up to 300 to 400 points. Whole-rock chemical composition analyses were performed in 18 sandstones samples selected based on the high content in heavy minerals and the absence of carbonate cements. Analyses were performed in ACTLABS (Canada) following the «WRA+trace 4Lithoresearch» routine by ICPAES for major elements and ICP-MS for trace (REE) elements. B and Li were analyzed in 10 samples by PGNAA (Pulsed Gamma Neutron Activation Analysis). Tourmaline chemical composition was analyzed in 10 thin section carbon coated with JEOL JXA-8900 M electron microprobe equipment (15 Kv of current, 20nA of intensity and 5im of diameter beam).

Petrography and geochemistry

The chemical classification of Lower Cretaceous sandstones based on Fe_2O_3/K_2O *vs* SiO_2/Al_2O_3 ratios (Herron, 1988) correspond to arkoses and sub-arkoses, which is similar to the petrographic classification (Caja, 2004).

Feldspars in Mora Fm. sandstones are exclusively plagioclase (0.9-13%), with a pure albite composition, without twined and showing variable alteration to illite and chlorite. Plagioclase grains can be also partially to completely replace by calcite. K-feldspar grains are not present. El Castellar Fm. sandstones have moderate to low content of feldspars (1.2-9.4%), mainly idiomorphic plagioclase with or without twining and detrital K-feldspars are present in very low abundances (~1-2%). Camarillas Fm. sandstones have abundant K-feldspar (<21.2%) and plagioclase with and without twined (<10.9%). K-feldspar grains occur from well preserved to illite and kaolin altered (epimatrix) and calcite replacement. It is important to note that K-feldspar abundance in the studied sandstones increase toward the uppermost formation (Camarillas). In contrast, plagioclase is more abundant in the lower Mora Fm. sandstones decreasing toward Camarillas Fm.

Low and medium grade metamorphic rock fragments are common in Mora Fm. sandstones (<5.4%). Occasionally, scarce micritic grains are present. In El Castellar

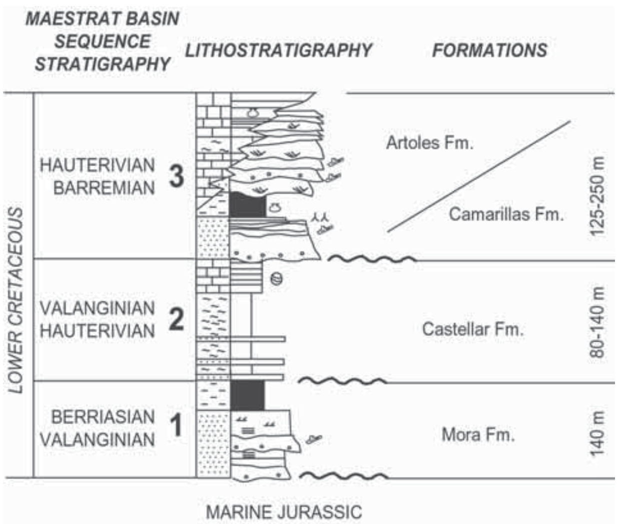

Fig. 2.- Schematic stratigraphic column of the Lower Cretaceous studied formations.

Fig. 2.- Columna estratigráfica sintética para las formaciones del Cretácico Inferior estudiadas.

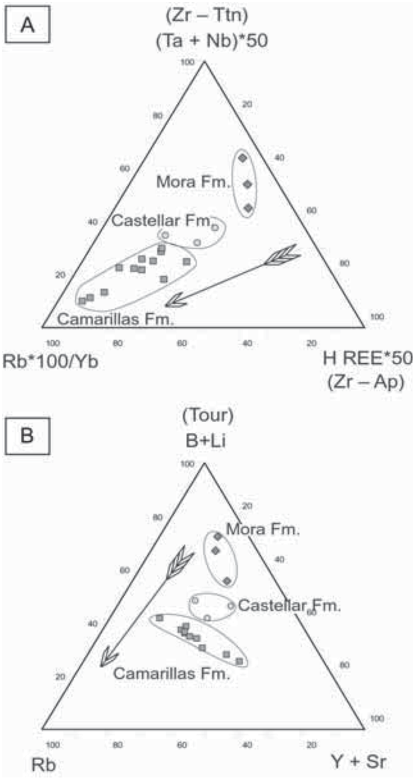

Fig. 3.- Relationships between selected trace elements (included rare earth elements, REE) based on their contribution to the heavy minerals observed assemblage and to the feldspar modal abundance (which is proportionally to Rb content). A) Zr: Zircon; Ap: Apatite; Ttn: Titanite. B). Tour: Tourmaline.

Fig. 3.- Relaciones entre elementos traza (incluidos las tierras raras, REE) seleccionados en función de su presencia en la paragénesis mineral observada y la abundancia modal de feldespatos (la cual es proporcional al contenido en Rb). A) Zr: Circón; Ap: Apatito; Ttn: Titanita. B) Tour: Turmalina.

Fm. sandstones clay-grains are very abundant up to 9.9%. Medium grade metamorphic rock fragments (up to 3%) and plutonic rock fragments (<1.9%) occur in low amounts. In Camarillas Fm. sandstones plutonic rock fragments are very abundant (<8%) compared with low and medium grade metamorphic rock fragments (<4.2%). In summary, rock fragment modal abundance in the studied sandstones reflect that low and medium grade metamorphic rock fragments are very common in Mora and El Castellar fms. However, plutonic rock fragments are dominant in Camarillas Fm.

Heavy minerals assemblage in Lower Cretaceous sandstones is made up of tourmaline, zircon, titanite and apatite. Tourmaline is the most abundant heavy mineral up to 2.2% in Mora Fm., <0.3% in El Castellar Fm. and <0.6% in Camarillas Fm. Tourmaline grains are characterized by coarse sizes (<200µm), high angularity, sub-idiomorphic shapes, poor sorting and brown to green pleochroism. Respect to the other heavy minerals of the assemblage, zircon, titanite and apatite are present in amounts less than 1%. Zircon and apatite have small sizes (less than 30 µm) and titanite appears frequently disaggregated.

The highest abundances in most of the trace elements (e.g., Y, Zr, Nb, Sn, La, Ce, Pr, Nd, Sm, Eu, Gd, Dy, Er, Tm, Yb, Lu, Hf, Ta, Th and U) are present in Mora Fm. sandstones compared with El Castellar and Camarillas sandstones (Caja *et al*., 2004). Trace element content (included rare earth elements, REE) have been plotted in a ternary diagram (Fig. 3), which takes into account the modal abundance of heavy minerals (zircon, titanite, apatite and tourmaline) and reflect a discrimination of the studied sandstones from the three formations. In this ternary diagram, Rb has been selected as a pole because it is considered as indicative of the K content discriminating the K-feldspar modal abundance. The relationship between Rb content and modal K-feldspar abundance is consistent with the high correlation index, up to 0.96, in Rb *vs* total feldspar modal % (Fig. 4). Therefore, Mora Fm. sandstones display the lowest Rb contents due to the absence of K-feldspar. In the other two poles of the ternary diagram, have been selected different trace and REE elements, which are mainly hosted in the observed heavy mineral assemblage (Rollinson, 1993; Bea, 2001).

Chemical composition of tourmalines ranges from dravite (Mg-endmember) to schorl (Fe-endmember) (Fig. 5). Both endmembers are well represented in the three studied formations. B and Li abundances have slightly variations between analyzed tourmalines of the different formations. Tourmaline chemical compositions in Mora Fm. sandstones have the maximum values in Mg content and the lowest in Fe. This pattern is also observed in Camarillas Fm. Compositions of El Castellar Fm. tourmalines are characterized by a group with high Mg content and another group with high Fe content.

Discussion and conclusions

Minor and trace (REE) elements are accepted to be transported in invariable proportions from the parent rock to the sedimentary basin (McLennan *et al*., 1993). Therefore, provenance information can be extracted using different relationships between trace and REE elements (Rollinson, 1993; Bea, 2001; McLennan, 2001). In the analysed rocks, the selected relationships between REE elements plotted in a ternary diagram (Fig. 3) allow a clear differentiation of petrofacies, which are compositional different in terms of type of feldspar present (K-feldspar and/or plagioclase) and heavy mineral assemblage, and relative modal abundance of each heavy mineral. On one hand, Rb is indicative of the K content and therefore, Rb is a discriminating element of the feldspar modal abundance. On the other hand, modal abundance of heavy mineral assemblage is consistent with bulk-rock chemical analyses in the sandstones. Zircon presence is related to Zr, Hf, Ta and Nb; titanite with Ti, Nb, V, Y and Sr; apatite with P and Y; and tourmaline with B and Li. These trace (REE) elements are hosted mainly in the referred heavy minerals, which can be useful for provenance discrimination. Therefore, the highest REE abundance is related with the highest heavy mineral modal abundance, e.g. Mora Fm. sandstones (Caja *et al*., in press).

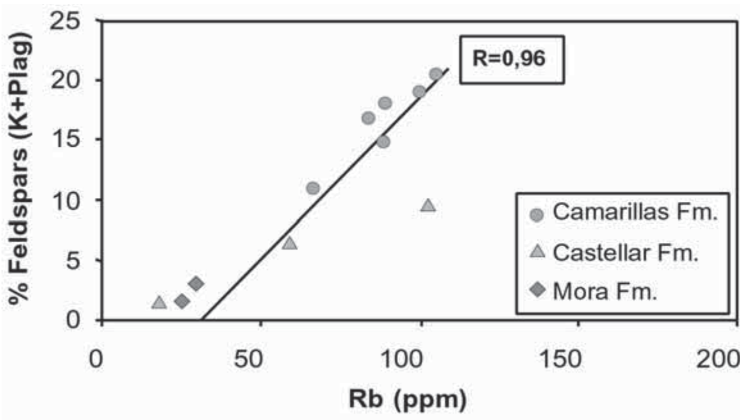

Fig. 4.- Rb versus modal feldspar abundance. Note the high correlation between both parameters.

Fig. 4.- Abundancia de feldespatos frente al contenido en Rb. Observar la alta correlación entre ambos parámetros.

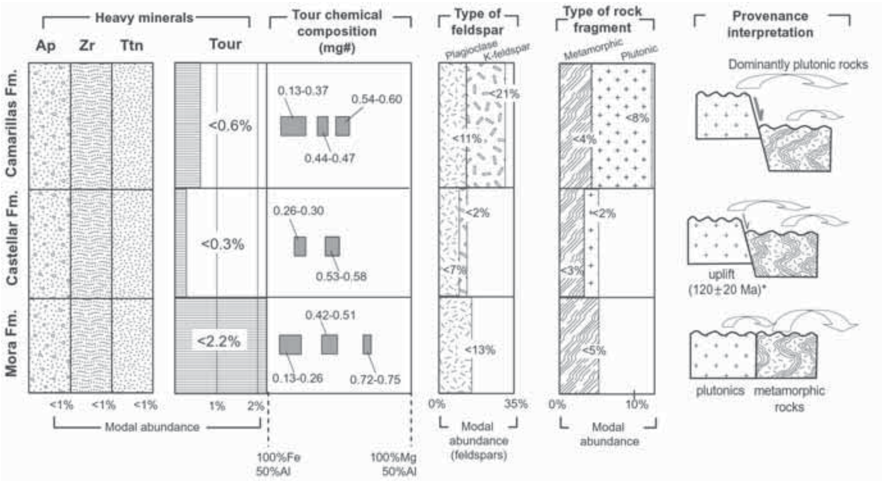

Fig. 5.- Synthesis of the type and heavy minerals modal abundance, tourmaline chemical composition (showing the main variation ranges; mg#=Mg/Mg+Fe), type and feldspar modal abundance and the provenance interpretation linking petrological observations and geochemical data for the Lower Cretaceous syn-rift sandstones in the Maestrat Basin. *(after Bruijne and Andriessen, 2000).

Fig. 5.- Síntesis del tipo y abundancia modal de minerales pesados, de la composición química en turmalinas (quedan reflejados los principales rangos de variación; mg#=Mg/Mg+Fe), el tipo y abundancia modal de feldespatos y la interpretación de la procedencia integrando datos petrográficos y geoquímicos para las areniscas syn-rift *de la Cuenca del Maestrazgo. *(tomado de Bruijne y Andriessen, 2000).*

In addition to bulk-rock chemical data, it is accepted that tourmaline chemistry reflects the source area composition (Henry and Guidotti, 1985). High concentrations of Al, Fe and Li are typical from granites and pegmatites, and high Mg concentration is typical of metasediments (von Eynatten *et al*., 1999). Chemical composition of the analyzed tourmalines suggest a mixed provenance from granitic (Fe-tourmaline) and metamorphic (Mg-tourmaline) source rocks, as reflect the mg# parameter (mg#= Mg/Mg+Fe; Fig. 5). If analyzed tourmaline composition is compared with tourmaline from possible source areas it is possible to rule out a provenance supply from calc-alkaline Lower Permian volcanic rocks of the Iberian Chain (Lago *et al*., 1993) because of their highest Fe-Tourmaline composition. However, tourmalines from granites of the Spanish Central System (Andonaegui, 1992) have Fe and Mg ranges comparable to the tourmaline analyzed in the Lower Cretaceous sandstones. Therefore, source areas supplied tourmaline minerals with different chemical composition (typical from granites and metamorphic rocks) and with variable modal abundances as suggest the higher contents in Mora Fm (Fig. 5).

Lower Cretaceous sandstones of the Maestrat Basin, based on petrological observations linked to geochemical data, are mainly derived from the mixing of metamorphic and granitic supplies (Caja, 2004). Source areas provided both types of rocks at the same time, although Camarillas Fm. sedimentation coincided with a generalised erosion stage of the Iberian Massif. This erosion event has been characterised by apatite fission-track analyses, dating an important cooling stage for the Mesozoic (120±20 Ma), which is related to the uplifting and erosion of the Iberian Massif due to the Neocimmerian tectonic stage (Bruijne and Andriessen, 2000). However, the major supply of plutonics during Camarillas sedimentation do not imply the dissapparition of metamorphic source areas (eg., Paleozoic shales) due to the fact that plutonic source areas supply more sand size fragments than metamorphic ones. Therefore, the mixing of both source areas implies a significant dilution of metamorphic rocks (Palomares and Arribas, 1993).

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