K-Ar dating of illites for time constraint on tectonic burial metamorphism of the Jurassic Nampo Group (West Korea)

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ABSTRACT: The timing of tectonic burial metamorphism of the early Mesozoic Nampo Group (western Korea) related to the Jurassic Daebo orogeny was first constrained by K-Ar dating of illite in sedimentary rocks. Two shale samples show a clear decreasing trend of both the amount of detrital component and K-Ar age of illites with decreasing grain size from >4 μ m to 4–2 μ m, and to <2 μ m fractions. This observation most likely reflects a binary mixture of old detrital 2M₁ and young authigenic 1M_d illites, usually seen in argillaceous sedimentary rocks. In this sense, the latest illitization attributable to the crustal loading can be estimated to be 157–140 Ma, the extrapolated illitic age inferred from a linear regression between the amount of detrital component and apparent K-Ar age of different size fractions. This result suggests that the maximum tectonic burial metamorphism of the Nampo Group has occurred in the last stage of the Daebo orogeny.

Key words: K-Ar dating, illite, tectonic burial metamorphism, Nampo Group, Jurassic Daebo orogeny

1. INTRODUCTION

Radiometric dating of authigenic illite in sedimentary and metamorphic rocks is a useful and powerful tool to constrain the latest diagenetic and low-grade metamorphic ages closely associated with burial heating and hydrothermal alteration (e.g., Clauer et al., 1993; Dong et al., 1995; Onstott et al., 1997; Uysal et al., 2006). Hoffman et al. (1976) also suggested that such illite dating can help to interpret the timing of regional overthrusting. The Korean Peninsula experienced a large-scale crustal thickening and shortening during the Jurassic Daebo orogeny, which was triggered by subduction of the paleo-Pacific plate beneath the East Asian continent (e.g., Chang, 1995; Kim, 1996). This peninsulawide orogeny eventually facilitated (i) Jurassic basins formation and their filling and (ii) subsequent tectonic burial metamorphism attributable to basement overthrusting and loading on the basin fills (Egawa and Lee, 2008, 2009). It was suggested that the overthrusting was initiated in early Middle Jurassic time just after the deposition of the basin fills (Koh, 2006; Egawa and Lee, 2008), but the timing of tectonic burial metamorphism due to crustal loading remains poorly constrained.

This study deals with K-Ar illite dating of the Nampo Group, a Lower to Middle Jurassic nonmarine basin-fill of the Chungnam Basin located in the central western Korean Peninsula (Fig. 1a) (Egawa and Lee, 2009 and references therein), to infer the age of diagenesis or low-grade metamorphism induced by postdepositional crustal loading. The Nampo Group in the Ocheon area comprises the Hajo, Amisan, Jogyeri, and Baegunsan formations with decreasing age and is structurally overlain by pre-Jurassic basement rocks (Choi et al., 1987; Egawa and Lee, 2006, 2008). Petrographic observation of sandstones and X-ray diffraction (XRD) analysis on less than 2 µm fraction reveal that the Nampo Group underwent significant compaction and low-grade metamorphism, respectively, and the evidence of a down-sequence increasing degree of thermal alteration is well preserved in the Ocheon area compared with the other two areas where the Nampo Group occurs (Fig. 1a) (Egawa and Lee, 2008).

2. MATERIALS AND ANALYTICAL METHODS

Two samples were selected from black to dark-grayish shale of the Amisan Formation in the Ocheon area. Sampling locations, sites 1 and 2, are positioned in the middle part of the Amisan Formation, where site 2 is located ca. 80 m above site 1 in the stratigraphic position (Fig. 1). The shale samples were powdered using an agate mortar and pestle, and calcite was removed using 1N HCl, whose treatment does not affect the K-Ar ages according to the experimental study by Clauer et al. (1993). Three different size fractions of the illitic clay minerals (>4, 4–2, and <2 μ m) were separated by high-speed centrifugation, and were air-dried.

For measurement of illite crystallinity, the oriented samples of different size fractions were run on a XRD through

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Fig. 1. (a) Map of the southern Korean Peninsula showing the major tectonic provinces and the study area (boxed). CB = Chungnam Basin, GB = Gyeongsang Basin, GM = Gyeonggi massif, OB = Okcheon Belt, TB = Taebaeksan Basin, YM = Yeongnammassif. (b) Simplified geological map and sampling locations in the Ocheon area of the Chungnam Basin where the JurassicNampo Group was deposited. (c) Schematic stratigraphic column of the Nampo Group in the Ocheon area. (b) and (c) were modified after Egawa and Lee (2008).

 $3-10^{\circ}2\theta$ with a scan speed of $0.5^{\circ}2\theta$ /min at operating conditions of 40 kV/30 mA, using a Rigaku Model D/Max-3c diffractometer with Co-filtered CuK α radiation. The obtained Kübler Index (KI) values, the most often-used index of illite crystallinity, were standardized using the Crystallinity Index Standard scale of Warr and Rice (1994).

To identify illite polytypism (cf. Grathoff and Moore, 1996), the oriented samples were analyzed on a XRD between 16 and 40°20 with a scan speed of $0.5^{\circ}20$ /min. The illite in argillaceous sedimentary rocks nearly always contains a mixture of detrital (2M₁) and authigenic (1M_d) components. The 2M₁ polytype is expected for the detrital illite or micas derived from the basement rocks including slates, schists, and phyllites, while the 1M_d polytype generally grows in clastic sedimentary rocks (Peaver 1999). The measured XRD patterns reveal polytype-specific peaks of illite at ~35°20, which are generally strong in a clay fraction

having high $2M_1$ -illite content as reported by Grathoff and Moore (1996, 2002). The amount of $2M_1$ illite of each clay fraction was quantified using best-fit analysis of the obtained XRD curves with the experimental XRD patterns based on a mixture of endmember $1M_d$ illite and $2M_1$ muscovite samples as authigenic and detrital components, respectively (Haines and van der Pluijm, 2008). Such $2M_1$ polytype quantification can estimate the apparent proportion of $1M_d$ illite.

K-Ar dating was carried out at Korea Basic Science Institute (KBSI) and Commonwealth Scientific and Industrial Research Organisation (CSIRO), Australia. K-Ar dating is usually done with the branched decay of ⁴⁰K to both ⁴⁰Ca and ⁴⁰Ar, the values of whose decay constants and isotopic abundance ratio are after Steiger and Jäger (1977): λ^{40} K = 5.543×10^{-10} /yr (λ^{40} Ca = 4.962×10^{-10} /yr and λ^{40} Ar = 0.581 $\times 10^{-10}$ /yr) and ⁴⁰K/K = 1.167×10^{-4} atom%, respectively. The total ⁴⁰K decay constant corresponds to a half-life of 1.25×10^9 yr and K-Ar age can be calculated by the following equation (Steiger and Jäger, 1977): age (yr) = $1/\lambda^{40}$ K × ln (40 Ar^{*}/ 40 K × λ^{40} K/ λ^{40} Ar + 1), where 40 Ar^{*} is radiogenic 40 Ar generated by 40 K decay.

The foregoing K-Ar ages of separated illites themselves do not constrain the timing of actual illitization during diagenesis and low-grade metamorphism because a mixture of detrital and authigenic components of illite in common argillaceous sediments results in the measured ages older than the latest authigenic illitic age (e.g., Clauer et al., 1993, 1995; Grathoff and Moore, 1996). It is known that perfect removal of detrital 2M1 illite/mica from clay sample for authigenic 1M_d illite dating is impossible even grain separation as small as <0.02 µm (Clauer et al., 1997), but it is also known that the finer fraction typically tends to increase in authigenic illite at the expense of detrital one. In this study we determined the timing of the latest illitization in the Nampo Group by illite age analysis, an alternative illite dating technique that determines apparent authigenic illite age by a linear relationship between percentage detrital 2M1 polytype and K-Ar ages measured from different clay fractions (Pevear, 1992; Grathoff and Moore, 1996; Haines and van der Pluijm, 2008).

3. RESULTS

3.1. Illite Crystallinity

The analyzed illite-rich clay samples of >4, 4–2, and <2 μ m present KI values of 0.19, 0.20, and 0.19° Δ 20 at site 1, respectively, and 0.21, 0.23, and 0.24° Δ 20 at site 2, respectively, all of which range in the thermal grade of epizone (Table 1) (Kübler, 1967; Blenkinsop, 1988).

3.2. Illite Polytypism

Figure 2 shows the measured XRD patterns of polytypespecific illite peaks: $1M_d$ polytype peaks at $24-34^\circ 2\theta$, $2M_1$ polytype peaks at $23-32^\circ 2\theta$, and 2.58\AA band which is common to both polytypes peaks at $35^\circ 2\theta$. All of these peaks of the analyzed samples reveal a weakening trend with claysize fining although $1M_d$ peaks do not show any clear trend. According to illite polytype quantification (Fig. 3), the analyzed illite-rich clay samples of >4, 4–2, and <2 µm present the amount of $2M_1$ illite of 31, 25, and 13% at site 1, respectively, and of 35, 27, and 10% at site 2, respectively (Table 1). All these results are strongly suggestive of the presence of mixed illites of decreasing detrital ($2M_1$) and

 Table 1. Illite crystallinity, K-Ar ages, and illite polytype of illite-rich fractions separated from black shales of the Amisan Formation in the Ocheon area

Sample	Size	KI	K	⁴⁰ Ar*		Isotopic age	Illite polytype	
	(µm)	$(^{\circ}\Delta 2\theta)$	(wt%)	(10^{-8}ccSTP/g)	(%)	(Ma)	2M ₁ (%)	$1M_{d}(\%)$
Site 1	>4	0.19	2.89	2143.48	95.5	181.9 ± 3.5	31	69
	4–2	0.20	3.44	2439.13	94.8	173.9 ± 3.3	25	75
	<2	0.19	4.39	2992.37	95.5	167.4 ± 3.2	13	87
Site 2	>4	0.21	3.14	1987.22	96.3	155.9 ± 3.2	35	65
	4–2	0.23	3.28	1952.81	94.8	147.0 ± 3.0	27	73
	<2	0.24	3.54	2080.64	95.3	145.2 ± 2.9	10	90

The clay samples of sites 1 and 2 were dated at KBSI and CSIRO, respectively.

The Kubler Index (KI) values and illite polytype ratios of shales were estimated from the same samples as those used for K-Ar dating. 40 Ar* is radiogenic 40 Ar generated by 40 K decay.



Fig. 2. XRD patterns of the three different illite size fractions of site 1 (a) and site 2 (b) samples. Open squares = $1M_d$ (diagentic or authigenic) illite, closed circles = $2M_1$ (detrital) illite, open triangle = 2.58Å band of $1M_d$ and $2M_1$.



Fig. 3. An example of XRD pattern (black) and best match (grey) for >4 μ m fraction of illite-rich clay from site 2. Estimation of best fit curve is after Haines and van der Pluijm (2008). Closed circles = 2M₁ (detrital) illite.

increasing authigenic $(1M_d)$ origins in shale samples with grain-size fining.

3.3. Illite Dating

The obtained radiometric ages are listed in Table 1. K-Ar date of the illitic material of >4, 4–2, and <2 μ m fractions was ca. 181.9, 173.9, and 167.4 Ma at site 1, respectively, and ca. 155.9, 147.0, and 145.2 Ma at site 2, respectively. The younging radiometric ages match well with decreasing size fractions and related weakening polytype-specific illite



Fig. 4. Extrapolation of detrital and authigenic K-Ar age of illiterich clay samples of the Amisan Formation from sites 1 and 2. See Table 1 for illite polytype quantification and K-Ar dating of different size fraction. The percentage detrital component on illite age analysis plot is plotted against the function $\exp^{\lambda t} -1$, where λ is potassium decay constant and t is apparent age. The depositional age of the Baegunsa Formation, the uppermost unit of the Nampo Group, is after Koh (2006).

peaks (Fig. 2), whose relationships can support adequacy of illite age analysis in this study (cf. Grathoff and Moore, 1996). The extrapolation from illite age analysis plots of sites 1 and 2 presents 157 and 140 Ma for the apparent age of authigenic endmember illite, respectively (Fig. 4).

4. DISCUSSION AND INTERPRETATION

The analyzed illitic clays in this study display both decreasing 2M₁ polytype and K-Ar age with decreasing clay size (Fig. 4 and Table 1), which strongly suggests a mixture of authigenic and detrital illite component. Here note that all of the experimented illitic clays show significantly low KI values corresponding to epizone (Table 1). It has been reported that 2M₁ polytype is most stable among illite polytypes and can authigenically grow in high temperature of >280 °C, whereas the authigenic growth of $1M_d$ is thought to form at lower temperature, generally below approximately 200 °C (Haines and van der Pluijm, 2008 and references therein). The temperature of anchizone ranges from 175 to 320 °C according to the standardized clay mineral crystallinity data suggested by Warr (1996). If authigenic 2M₁ illite were formed in sedimentary basins, it would be unlikely that the XRD measurement shows decreasing 2M₁ ratio with clay-size fining. In this sense, it is more likely that the Amisan Formation originally experienced thermal heating not in epizonal but in anchizonal conditions, and that mixing of 1M_d illite with detrital 2M₁ illite having relatively low KI value consequently resulted in enhancing apparently high illite crystallinity of the measured samples. Thus, the maximum burial temperature of the Amisan Formation is thought to be below 280 °C, which is in agreement with the sandstone textural observation suggesting the absence of ductile deformation in the middle part of the Amisan Formation (Egawa and Lee, 2008).

The extrapolated K-Ar age of illite in the Amisan shale presents 157 and 140 Ma at sites 1 and 2, respectively (Fig. 4), which is younger than the depositional age of the Nampo Group (~170 Ma; Koh, 2006). Thus, it is interpreted that the authigenic illite started to form 13 million years after the deposition of the Nampo Group and continued its formation for at least 17 million years. It is known that the Daebo orogeny ended at ca. 135 Ma ago (Kim, 1996), just 5 Ma later than the latest illite ages we dated, and thus it appears that the calculated illitic age corresponds to the age of lowgrade metamorphism attributable to basement overthrusting in the late stage of the Daebo orogeny (Egawa and Lee, 2008). It is very unlikely that the analyzed epizonal shales were hydrothermally altered by post-Jurassic granite intrusion. According to Schlegel et al. (2007), the isotopic age of hydrothermal overprinting is expected either when the K-Ar age of 2M₁ illites was totally reset due to hydrothermal heating, or when separation of the pure $1M_d$ illite precipitated from hydrothermal fluids was successful. Neither cases seem to be applicable to this study by the following lines of evidence: (i) younging radiometric ages of illites with decreasing clay size (Table 1), and (ii) no post-Jurassic granite intrusion in and around the study area (Egawa and Lee, 2008). Given that the Nampo Group in the Ocheon area escaped from hydrothermal overprinting, we conclude that the timing of illitization probably corresponds to that of the maximum tectonic burial metamorphism.

5. CONCLUSION

The Jurassic Nampo Group located in western Korea was deposited by Middle Jurassic time, and subsequently underwent tectonic burial and related low-grade metamorphism attributable to crustal loading. Based on the relationships between decreasing detrital illite component with decrease in K-Ar illitic age and clay size, the authigenic illitization in the Nampo Group is inferred to have occurred at 157–140 Ma, which corresponds to the last stage of the Daebo orogeny.

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