Protracted Storage for Calc-Alkaline Andesitic Magma in Magma Chambers: Perspective from the Nageng Andesite, East Kunlun Orogen, NW China

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Abstract: Calc-alkaline andesitic rocks are a major product of subduction-related magmatism at convergent margins. Where these melts are originated, how long they are stored in the magma chambers, and how they evolved is still a matter of debate. In this study, we present new data of whole-rock elemental and Sr-Nd-Pb isotope compositions, and zircon U-Pb-Th isotopes and trace element contents of Nageng (basaltic-)andesites in the East Kunlun Orogen (NW China). The similar age and whole-rock elemental and Sr-Nd-Pb isotope contents suggest that the Nageng andesite and basaltic andesite are co-magmatic. Their low initial $^{87}$Sr/$^{86}$Sr (0.7084–0.7086) but negative $\varepsilon$Nd(t) values ($-10.61$ to $-9.49$) are consistent with a magma source from the juvenile mafic lower crust, possibly related to the mantle wedge with recycled sediment input. The U-Pb age gap between the zircon core (ca. 248 Ma) and rim (ca. 240 Ma) reveals a protracted magma storage (~8 Myr) prior to the volcanic eruption. When compared to the zircon rims, the zircon cores have higher Ti content and Zr/Hf and Nb/Ta ratios, but lower Hf content and light/heavy rare earth element ratios, which suggests that the parental magma was hotter and less evolved than the basaltic andesite. The plagioclase accumulation likely resulted in Al$_2$O$_3$-enrichment and Fe-depletion, forming the calc-alkaline signature of the Nageng (basaltic-)andesites. The magma temperature, as indicated by the zircon saturation and Ti-in-zircon thermometry, remained low (725–828 °C), and allowed for the magma chamber to survive over ~8 Myr. The decreasing $\varepsilon$Hf(t) values from zircon core (avg. 0.21, range: $-1.28$ to $1.32$) to rim (avg. $-3.68$, range: $-7.30$ to $-1.13$), together with the presence of some very old xenocrystic zircons (268–856 Ma), suggest that the magma chamber had undergone extensive crustal contamination.

Keywords: antecrystic zircon; juvenile mafic lower crust; thermal history; Sr-Nd-Pb-Hf isotopes; zircon trace elements

1. Introduction

Calc-alkaline rocks are generally considered to have formed at convergent margins, and they contain higher Al$_2$O$_3$ and lower FeO/FeO$^+$ ratios than tholeiitic rocks [1–5]. Calc-alkaline andesitic rocks are extensively distributed on Earth and they represent an important constituent of the continental crust [6,7]. Hence, understanding the formation mechanism of calc-alkaline andesites is important in unravelling the continental crust evolution [8], which has remained a key debate in igneous petrology. Many models were proposed to explain the formation of these melts: (1) the partial melting of hydrous mantle [6,9]; (2) fractional crystallization of mantle-derived basaltic magma [10]; and, (3) mixing
between mafic and felsic magmas [4,11,12]. These models are built up around the fundamental question of whether the calc-alkaline signature is inherited from the source region or induced by magmatic processes, including magma fractionation, crustal contamination, and mixing with Si-rich, Fe-poor crustal materials [13,14].

Constraining the duration of arc magma is crucial in understanding how magmas are transported, stored, and evolved in the crust, and what may trigger volcanic eruptions [15,16]. Many studies have suggested a timescale of hundreds of thousands to millions of years in crustal-level magma chambers [16–20]. Based on whole-rock geochemistry and Sr-Nd-Pb isotopes, in combination with zircon U-Pb geochronology, trace element geochemistry, and zircon saturation and Ti-in-zircon thermometry on the Nageng (basaltic-) andesites from the East Kunlun Orogen (EKO, NW China), we propose that the magma can reside in the magma chamber for ~8 Myr prior to eruption, and undergo extensive crystal accumulation in order to attain its calc-alkaline character.

2. Geological Setting and Petrography

2.1. Geological Setting

The EKO extends E–W for 1500 km along the northern margin of Bayanhar Terrane (Figure 1A). It is separated from the West Kunlun Orogen by the sinistral Altyn Tagh fault on the west and adjoins the Elashan Orogen by the Wenquan fault on the east (Figure 1B). The formation of the EKO involves the closure of Paleo-Tethyan A’neymaqen Ocean and multistage subduction-accretion-collision between the Qaidam and Bayanhar terranes [21]. The orogen comprises four major tectonic units, i.e., (from north to south) the North Qimantagh belt, the Central EKO, the South EKO, and the Bayanhar Terrane, which are separated by the North-East-Kunlun, Central East-Kunlun, and South East-Kunlun faults, respectively [21,22]. The study area is located in the easternmost segment of the Central EKO (Figure 1B). This segment was extensively intruded by three pluton generations during the Ordovician–Silurian, Early Devonian, and Late Permian–Triassic [21], among which the latter predominates and includes four sub-phases at ca. 270–250 Ma, 250–240 Ma, 240–220 Ma, and 220–200 Ma, which are interpreted to have been related to oceanic plate subduction, subducted slab break-off, mantle underplating, and island-arc extension, respectively [21]. Abundant mafic micro-granular enclaves (MMEs) that suggest magma mingling characterize these Late Permian–Triassic graniteoids [23–25].

The Nageng (basaltic-)andesites are exposed as numerous lava domes in NE Nageng (Figure 1C). Previous works classified them as the Upper Triassic Elashan Formation [26–29], but we reclassified them as the Middle Triassic Naocangjiangou Formation according to their final emplacement age (ca. 240 Ma) here [24]. The (basaltic-)andesites are underlain and overlain by the Paleoproterozoic Jinshuikou Group and Upper Triassic Elashan Formation, respectively. The Jinshuikou Group mainly comprises biotite-(amphibole)-plagioclase gneiss, quartz-mica schist and hornblende schist. The Elashan Formation mainly contains rhyolites and their pyroclastic equivalence (Figure 1C).
Figure 1. Schematic tectonic maps of (A) China and (B) East Kunlun Orogen (modified after Ref. [26]); and, (C) Simplified geologic map of the Nageng area, showing the distributions of (basaltic-)andesites.

2.2. Sampling and Petrography

Eight representative samples (AD1-4 and BA1-4) were collected for whole-rock geochemical and isotopic analyses from outcrops at Nageng (Figure 1C). The samples AD2 and BA2 were also zircon U-Pb dated. The samples can be classified as andesite and basaltic andesite based on their petrographic features.

The andesite samples contain phenocrysts of amphibole (5–10 vol.%), and plagioclase (25–30 vol.%), K-feldspar (<5 vol.%) and quartz (<1 vol.%) in a cryptocrystalline groundmass (Figure 2A–D). Amphibole phenocrysts are chlorite- and carbonate-altered (Figure 2C–D). The basaltic andesite samples have coarser phenocrysts of amphibole (10–15 vol.%), plagioclase (35–40 vol.%), and pyroxene (<5 vol.%) in a cryptocrystalline groundmass (Figure 2E–G). Sporadic pyroxene phenocrysts were chlorite- and carbonate-altered with their cracks being filled with rutile (Figure 2H).
Figure 2. Hand-specimen and thin-section (micro)photographs of the Nageng (A–D) andesite and (E–H) basaltic andesite: (A) Andesite hand-specimen with phenocrysts of plagioclase and amphibole; (B) Major plagioclase and minor K-feldspar phenocrysts; (C) Sericite-altered plagioclase and carbonate-chlorite-altered amphibole phenocrysts; (D) Carbonate-chlorite-altered amphibole with sporadic quartz phenocrysts; (E) Basaltic andesite hand-specimen with phenocrysts of plagioclase and amphibole; (F) Plagioclase phenocrysts; (G) Carbonate-chlorite-altered amphibole phenocrysts; and, (H) Carbonate-chlorite-altered pyroxene phenocrysts with cracks filled with rutile. B–D,F: cross-polarized light; G,H: plane-polarized light. Abbreviations: Cab: Carbonate; Chl: Chlorite; Kfs: K-feldspar; Pl: Plagioclase; Px: Pyroxene; Qtz: Quartz; Amp: Amphibole; Ser: Sericite; Rt: Rutile.
3. Analytical Methods

3.1. Zircon U-Pb-Hf Isotope and Trace Element Analyses

Zircon grains were separated while using the conventional heavy-liquid and magnetic separation technique, and then hand-picked under a binocular microscope. Representative grains were mounted in epoxy resin, polished, and gold-coated. The internal structure of the zircon grains was observed under transmitted and reflected light, and cathodoluminescence (CL) imaging using a TESCAN MIRA3 field-emission scanning electron microscope (FE-SEM) at the Sample Solution Analytical Technology (SSAT) Co. Ltd. (Wuhan, China). The LA-ICP-MS zircon U-Pb dating and trace element analyses were simultaneously performed with an Agilent 7700e ICP-MS instrument (Agilent Technologies Inc., Palo Alto, CA, USA) at SSAT. The detailed analytical procedures followed those were described by Ref. [30]. Excel-based ICPMSDataCal 8.3 and GLITTER 4.0 (Macquarie University, Sydney, New South Wales, Australia) software were used to conduct off-line selection and integration of background and analyzed signals, time-drift correction, and quantitative calibration [30,31]. Zircon U-Pb concordia diagrams and weighted average ages were calculated while using the Isoplot 3.0 software [32]. Tables S1 and S2 present the analytical results. The U-Pb dating results have been filtered for any potential inclusions and fractures by checking the locations and time-resolved signals of all analysis spots.

In-situ zircon Hf isotopic compositions were also determined at the SSAT while using a Neptune Multi-Collector (MS)-ICP-MS (Thermo Fisher Scientific Inc., Waltham, MA, USA) connected to a Geolas HD excimer ArF laser ablation system. Hafnium isotopic measurements were made on the same age domains of the U-Pb-dated zircon grains, using a spot size of 44 μm. The zircon 91500, GJ-1 and Termora 2 were used as the standards. Detailed analytical procedures are as described in [33]. Off-line selection and integration of analyte signals, and mass bias calibrations were performed using ICPMS DataCal [34]. The initial $^{176}\text{Hf}/^{177}\text{Hf}$ ratios and $\varepsilon_{\text{Hf}}(t)$ values were calculated with the measured U-Pb ages with reference to the present-day Chondritic reservoir $^{176}\text{Hf}/^{177}\text{Hf} = 0.282772$ and $^{176}\text{Hf}/^{177}\text{Hf} = 0.0332$ [35]. A decay constant of $1.865 \times 10^{-11}$ per year for $^{176}\text{Lu}$ was adopted for the $\varepsilon_{\text{Hf}}$ calculations of Ref. [36]. Two-stage model ages were calculated using $^{176}\text{Lu}/^{177}\text{Hf} = 0.015$, $^{176}\text{Hf}/^{177}\text{Hf} = 0.28325$, and $^{176}\text{Yb}/^{177}\text{Hf} = 0.0384$ [37].

3.2. Whole-Rock Major and Trace Element Analyses

Bulk-rock geochemical analyses were performed at the Beijing Research Institute of Uranium Geology (BRIUG), Beijing, China. For major element analysis, whole-rock powder (0.5 g) of each sample was digested by HF + HNO$_3$, and then analyzed with an AxiosMAX X-ray fluorescence spectrometer (XRF) (Malvern Instruments Inc., Westborough, MA, USA) with detection limits of 0.015 wt.% for Al$_2$O$_3$, SiO$_2$, MgO, and Na$_2$O; 0.01 wt.% for CaO, K$_2$O, TiO$_2$, and FeO$^t$; and, 0.005 wt.% for MnO and P$_2$O$_5$. The FeO content was determined by chemical titration with a detection limit of 0.1 wt.%. Trace element contents were determined using a NexION300D high-resolution ICP-MS (PerkinElmer Inc., Waltham, MA, USA). Approximately 50 mg of powder was dissolved at 150 °C in high-temperature Teflon bombs while using a HF + HNO$_3$ mixture. Rhodium was used as an internal standard to monitor signal drift during the analysis. The Chinese national rock standard GBW-1 and GBW-2 were used to calibrate the elemental concentrations of the measured samples. The analytical uncertainty for both major and trace elements was generally better than 5%.

3.3. Whole-Rock Sr-Nd-Pb Isotope Analyses

The analyses were performed on an IsoPROBE-T Thermal Ionization Mass Spectrometer (TIMS) (Isotopx Ltd., Swettenham, Cheshire, UK) at the BRIUG. Detailed analytical procedures are as described in Ref. [38]. The measured Sr and Nd isotope ratios were nor-
malized to natural $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$ and $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$, respectively. Repeated analyses yielded an average $^{86}\text{Sr}/^{88}\text{Sr} = 0.710236 \pm 0.000007$ ($2\sigma$, $n = 6$) for Sr standard NISTSRM 987 and $^{146}\text{Nd}/^{144}\text{Nd} = 0.511864 \pm 0.000003$ ($2\sigma$, $n = 6$) for the Nd standard JNd-1 [39]. The total chemical Sr and Nd blanks are <200 pg and <100 pg, respectively. The $^{208}\text{Pb}/^{206}\text{Pb}$, $^{207}\text{Pb}/^{206}\text{Pb}$, and $^{206}\text{Pb}/^{204}\text{Pb}$ values of the Standard NBS981 analyzed in this study are 2.164940 ± 15 ($2\sigma$), 0.914338 ± 7 ($2\sigma$), and 0.0591107 ± 2 ($2\sigma$), respectively. The measured results of the NBS981 exhibit mass fractionation approximately 0.1%/au relative to the recommended values. The total blank levels are below ±2%. Two-stage Nd depleted-mantle model ages (TDM2-Nd) were calculated using the same assumption formulation as Ref. [40].

4. Results

4.1. Zircon U-Pb Ages and Trace Element Compositions

Tables S1 and S2 (Supplementary Materials) and Figure 3 present the zircon U-Pb isotope and trace element data and representative CL images, respectively. The zircons from the andesite and basaltic andesite samples mostly show clear magmatic oscillatory zoning and they can be classified into two types: Type A zircons have similar core-rim U-Pb ages (~240 Ma); Type B zircons have different core-rim U-Pb ages (core: ~248 Ma; rim: ~240 Ma) (Figure 3). Type A zircons have similar sizes (150–200 μm) and length/width ratios (1.5–2.0) (Figure 3), whereas type B zircons have varying sizes (200–300 μm) and length/width ratios (2–4) (Figure 3).

Notably, the core and rim of type A zircons both have similar U-Pb ages to those of the rim of type B zircons (Tables S1 and S2). Thus, we integrated the rim and core data of type A zircons with the type B zircon rim data to facilitate the discussion, and the following discussion is mainly focused on type B zircons.

For the andesite samples, thirteen zircon-rim spots yielded $^{206}\text{Pb}/^{238}\text{U}$ ages of 235.2–245.0 Ma, with a weighted mean $^{206}\text{Pb}/^{238}\text{U}$ age of 239.5 ± 1.8 Ma (MSWD = 1.01). Six spots on the zircon cores yielded $^{206}\text{Pb}/^{238}\text{U}$ ages of 245.7–251.3 Ma, with a weighted mean $^{206}\text{Pb}/^{238}\text{U}$ age of 249.0 ± 2.8 Ma (MSWD = 0.33). For the basaltic andesite sample, twelve spots on the zircon rims yielded $^{206}\text{Pb}/^{238}\text{U}$ ages of 236.8–243.7 Ma, with a weighted mean $^{206}\text{Pb}/^{238}\text{U}$ age of 240.4 ± 1.8 Ma (MSWD = 1.18). Five spots on the zircon cores yielded $^{206}\text{Pb}/^{238}\text{U}$ ages of 245.7–249.7 Ma, with a weighted mean $^{206}\text{Pb}/^{238}\text{U}$ age of 247.5 ± 2.0 Ma (MSWD = 0.51).

The Th/U ratios of all zircons are 0.13–1.63, which suggests a magmatic origin (>0.1) [41]. Zircon rims of andesite and basaltic andesite samples have relatively high Hf (8329–12,486 and 10,127–11,761 ppm) contents and LREE/HREE (0.01–0.12 and 0.06–0.24) ratios, and low Ti (1.15–4.37 and 0.90–4.89 ppm) contents and Zr/Hf (39.72–55.95 and 48.98–58.92), Nb/Ta (0.73–3.08 and 2.23–3.59) ratios (Figure 4A–D). Comparatively, the zircon cores have lower Hf (9260–11,908 and 8052–11,351 ppm) contents and LREE/HREE (0.01–0.19 and 0.01–0.04), but higher Ti (1.35–11.08 and 3.21–13.54 ppm) contents and Zr/Hf (41.65–53.56 and 43.70–61.60), Nb/Ta (0.72–3.02 and 2.23–3.59) ratios (Figure 4A–D).
Figure 3. (A, B) Linearized probability density plots and (C, D) weighted mean zircon ages of the Nageng andesite and basaltic andesite samples; (E, F) Zircon U-Pb concordia diagrams and representative zircon cathodoluminescence (CL)-images from andesite and basaltic andesite; (G, H) Weighted mean ages of zircon rims and cores from andesite and basaltic andesite.
Figure 4. Boxplots of zircon (A) Zr/Hf, (B) Nb/Ta, (C) LREE/HREE ratios, and (D) Ti content for the Nageng andesite and basaltic andesite.

4.2. Whole-Rock Major and Trace Elements

Table S3 (Supplementary Materials) lists the major and trace element compositions of the studied samples. These samples can be divided into andesite and basaltic andesite based on the Total Alkali-Silica (TAS) diagram (Figure 5A). The andesites have 55.76–56.39 wt.% SiO₂, 16.99–18.25 wt.% Al₂O₃, 3.04–3.50 wt.% MgO, 7.44–8.00 wt.% CaO, 1.92–2.27 wt.% Na₂O, and 0.352–0.423 wt.% K₂O. Comparatively, the basaltic andesites have lower SiO₂ (52.55–54.22 wt.%). These samples have Al₂O₃ (19.71–20.35 wt.%), MgO (2.64–2.71 wt.%), CaO (9.22–9.63 wt.%), Na₂O (1.95–2.07 wt.%), and K₂O (0.210–0.363 wt.%) concentrations. The andesite and basaltic andesite samples have Mg♯ values [molar 100*Mg/(Mg + Fe)] of 56.05–58.48 and 49.44–53.10, respectively, and similar low-K and calc-alkaline character (Figure 5B–D).

In the chondrite-normalized rare earth element (REE) diagram (Figure 6A,B), all of the samples are enriched in light REEs (LREEs) with (La/Yb)N ratios of 6.52–7.20 (andesite) and 3.67–4.05 (basaltic andesite). The andesites have more-distinct Eu anomalies (δEu = 0.74–0.82) than those of the basaltic andesites (δEu = 0.80–0.87). All the samples have flat heavy REE (HREE) patterns and decreasing total HREE abundance with increasing SiO₂. The andesites and basaltic andesites show similar enrichments in certain large-ion lithophile elements (LILEs; e.g., Ba, Rb), and depletions in some high field strength elements (HSFEs; e.g., Nb, Ta, P, and Ti) in the primitive mantle-normalized trace element diagram (Figure 6C,D).
Figure 5. Diagrams of (A) (Na₂O + K₂O) vs. SiO₂ [42], (B) K₂O vs. SiO₂ [43], (C) FeO'/MgO vs. SiO₂ [3], and (D) Alkali-FeO'-MgO (AFM) [2].

Figure 6. (A,B) Whole-rock chondrite-normalized rare earth element (REE) patterns and (C,D) primitive-mantle-normalized multi-elements plots for the Nageng andesite and basaltic andesite. Chondrite and primitive mantle normalizing values are from Ref. [44].
All of the samples in this study are variably altered, as demonstrated by the presence of minor secondary minerals and moderate to high loss on ignition (LOI) (3.41–6.29 wt.%). In order to evaluate the elemental mobility in the Nageng (basaltic-)andesites, we correlated the major element oxide contents with the chemical index of weathering [CIW = molar Al₂O₃/molar(Al₂O₃ + CaO + Na₂O)] [45], and found no discernible correlations for K₂O, Na₂O, MgO, and FeO₆ (Figure S1). This precludes a significant alteration effect on the samples and, thus, the geochemical data can be used for petrogenetic interpretations.

4.3. Sr-Nd-Pb-Hf Isotope Data

Table S4 (Supplementary Materials) lists whole-rock Sr-Nd-Pb isotopic compositions of the (basaltic-)andesites. Initial ⁸⁷Sr/⁸⁶Sr (I), εNd(t), (²⁰⁶Pb/²⁰⁴Pb), (²⁰⁷Pb/²⁰⁴Pb), and (²⁰⁸Pb/²⁰⁴Pb) values are calculated while using the zircon U-Pb age of 239.5 Ma for both andesite and basaltic andesite.

The andesite samples have whole-rock I of 0.7084–0.7086 and εNd (t) of −10.53 to −9.58, which correspond to depleted-mantle model ages (TDM2-Nd) of 1624–1533 Ma. They are characterized by highly radiogenic Pb isotope compositions, with present-day whole-rock ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb, and ²⁰⁸Pb/²⁰⁴Pb ratios of 18.399–18.439, 15.595–15.622, and 38.661–38.745, respectively. The corresponding initial ratios (at 239.5 Ma) are 18.259–18.286, 15.588–15.615, and 38.328–38.441, respectively. The basaltic andesite samples have similar I (0.7085–0.7086) and εNd (t) (−10.61 to −9.49) to those of the andesite samples, which correspond to TDM2-Nd of 1552–1630 Ma. They have present-day whole-rock ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb, and ²⁰⁸Pb/²⁰⁴Pb ratios of 18.408–18.446, 15.603–15.614, and 38.705–39.752, respectively. The corresponding initial ratios (at 239.5 Ma) are 18.280–18.343, 15.598–15.608, and 38.375–38.548, respectively.

Zircon cores of the (basaltic-)andesite samples have ¹⁷⁶Hf/¹⁷⁷Hf ratios of 0.282595–0.282658 and 0.282426–0.282563, depleted εHf(t) values (−7.30 to −2.39 and −5.85 to −1.13), and older TDM2 of 1423–1734 Ma and 1344–1643 Ma characterize zircon rims of the (basaltic-)andesite samples, respectively.

4.4. Crystallization Temperature

The revised Ti-in-zircon thermometer [46] was used to calculate the Ti-in-zircon crystallization temperatures (TzircTi), assigning aSiO₂ (SiO₂ activity) = 1.0, and aTiO₂ (TiO₂ activity) = 0.6. When considering too high aTiO₂ or too low aSiO₂ results in underestimation of temperature, and an error of 0.2 in activity estimates yields an error of ~30 °C [47], which does not affect our main conclusions. Consequently, we obtained TzircTi of 611–717 °C (avg. 652 °C, n = 13) and 594–727 °C (avg. 685 °C) for the andesite and basaltic andesite samples, respectively (Table S1). Accordingly, the TzircTi for zircon cores of andesite and basaltic andesite are 666–807 °C (avg. 713 °C, n = 5) and 690–828 °C (avg. 756 °C, n = 5), respectively (Table S1).

Using the zircon saturation temperatures (Tzircsat) that were based on the whole-rock Zr contents and major element compositions [48], we obtained a temperature of 713–768 °C (avg. 713 °C, n = 5) and 690–828 °C (avg. 756 °C, n = 5), respectively (Table S1).

5. Discussion

5.1. Magma Source

The andesites and basaltic andesites have similar chondrite-normalized REE and primitive mantle-normalized multi-element patterns, which indicates a cognatic relation (Figure 6A–D). This conclusion is further supported by their similar Sr-Nd isotopes (Figure 7A) and a linear Pb isotope trend (Figure 7B).
Figure 7. Plots of (A) ($^{87}$Sr/$^{86}$Sr)$_i$ vs. $\varepsilon$Nd(t) and (B) ($^{207}$Pb/$^{204}$Pb)$_t$ vs. ($^{206}$Pb/$^{204}$Pb)$_t$. Data source: A’nyemaqen MORB and OIB (interpreted Paleo-Tethyan evolution related) are from Refs. [49,50]; Peraluminous S-type gneissic granites from Ref. [51]; Magma-mixing granitoids and their MMEs from Refs. [23,26,52]; Enriched mantle-derived mafic rocks from Refs. [53–57].

The ancient lower crust and enriched mantle (EM) were commonly interpreted to be the magma source for the Late Permian-Triassic igneous rocks in the EKO [23,49,51,53,54,58]. However, the Sr-Nd-Pb isotopes of (basaltic-)andesites are different from both the EKO peraluminous S-type gneissic granites (interpreted ancient lower crust-derived) [49,51,58] and EKO mafic rocks (e.g. gabbro, diabase, and lamprophyre, interpreted EM-derived) [23,53,54,59], excluding a sole contribution from any of these two source regions (Figure 7A,B). Mixing of these two sources may also be ruled out, because the $I_{Sr}$ and $\varepsilon$Nd(t) values of the (basaltic-)andesite rocks do not fall on the mixing array of these two source domains (Figure 7A). We propose that the magma source may have been the juvenile (newly-formed) mafic lower crust because:

1. The Hf model ages (1.2–1.4 Ga) of zircon cores from the studied rocks are much younger than ancient lower crust rocks in the EKO (e.g., Proterozoic gneiss and schist; 1.6–2.2 Ga) [49,51,58], which suggests the involvement of juvenile components.

2. The zircon cores of our samples have lower Hf contents and LREE/HREE ratios, but higher Ti contents and Zr/Hf, Nb/Ta ratios than those of the zircon rims (Figure 4A–D), suggesting that the zircon cores were crystallized in a hotter and less-evolved melt [46,60–62]. In other words, the parental magma contains more mantle-derived components than those of the current zircon host (basaltic andesite). The overlapping of $\varepsilon$Hf(t) values between the zircon core (−2.79 to 1.76) and typical mantle-derived mafic rocks (−2.79 to 1.76) in the EKO (Figure 8) evidence the mantle-derived affinity [23,53,54,59].
Figure 8. Plot of zircon $\varepsilon_{\text{Hf}}(t)$ vs. U-Pb age for the Nageng andesite and basaltic andesite. Data source: Peraluminous S-type gneissic granitoids from Refs. [63,64]; Magma-mixing granitoids and their mafic micro-granular enclaves (MMEs) from Ref. [65]; Enriched mantle-derived mafic rocks from Refs. [55–57].

(3) The studied samples have relatively low Mg $^+$ (49.44–58.48), which precludes a primitive mantle-derived origin, because primitive andesites that are formed by direct extraction from the mantle or subducted oceanic crust have Mg $^+$ > 60 [7,66]. In addition, the $\varepsilon_{\text{Nd}}(t)$ (−10.61 to −9.49) of the studied rocks are consistent with those of subducted crust-derived sediments (−10 to −6), which suggests that their magma source contains components of recycled sediments through subduction or crustal delamination [67,68].

(4) The varying mixing ratios of the juvenile mafic lower crust to EM could simply match the clear linear $^{143}$La vs. $\varepsilon_{\text{Nd}}(t)$ array (Figure 7A) of the Late Permian-Triassic EKO granitoids that contain abundant MMEs (interpreted magma-mixing related) [69–71]. Although we favor a juvenile mafic lower crustal magma source, we do not preclude an EM-type magma source that is isotopically distinct from the previously reported EM-type rocks in the EKO [29,65,72]. Because all EM-type (e.g., EM1 and EM2) sources share a common inheritance from the continental crust, elemental and isotopic difference of the EM-type sources can be induced by the addition of different proportions of recycled lower-/upper-continental crust through subduction or crustal delamination [73–75].

5.2. Magma Chamber Process

Our andesites and basaltic andesites have an overlapping age distribution in ca. 248 and 240 Ma groups (Figure 3) and a general resemblance in mineral assemblage and whole-rock elemental contents (Figure 6), which implies that the two types were crystallized and evolved in the same magma chamber. Zircon U-Pb dating reveals that zircon cores from both rock types have markedly older weighted mean $^{206}$Pb/$^{238}$U ages (249.0 ± 2.8 Ma and 247.5 ± 2.0 Ma) than the zircon rims (239.5 ± 1.8 Ma and 240.4 ± 1.8 Ma). The zircon rims are assumed to be autocrystic and they have crystallized from the host magmas, because the $T_{\text{zirco}}$ (765 °C and 741 °C) of the two rock types are higher than the zircon rim $T_{\text{zirco}}$ (611–717 °C and 594–722 °C), which suggests that the host magmas were Zr-saturated [76]. However, it is essential to distinguish whether the zircon cores are xenocrystic (inherited crystals either from the wallrocks or source region) or autocrystic (recycled from similar magma system) in order to track magma chamber process by zircon chemistry [47,77].
Zircon cores of our samples show relatively homogeneous textures with the absence of a resorbed or truncated surface, ruling out a xenocrystic origin [78,79]. The zircon cores show differentiation-related indices (e.g., lower LREE/HREE ratios and higher Ti contents and Zr/Hf and Nb/Ta ratios), which indicate that they were in equilibrium with and crystallized in a hotter and less-evolved magma, as aforementioned. Given their distinct older ages (ca. 248 Ma) than the zircon rims (ca. 240 Ma), we argue that the zircon cores are antecrystic and recycled from a long-lived magma with progressive growth. This protracted zircon crystallization history was also supported by the elevated MSWDs and linearized probability plots (Figure 3A–D), because MSWD > 1 and an abnormal age distribution preclude the two distinct age populations being induced by analytical uncertainties [80,81]. In addition, the analytical uncertainties (± 1.8–2.8 Ma) are not large enough to encompass the anticipated duration (~8 Myr) of magma storage. Thus, we interpret that the obtained zircon U-Pb ages have recorded the prolonged and continuous crystallization from ca. 248 to 240 Ma for the (basaltic-)andesites.

The studied samples are calc-alkaline, and they have high Al2O3 (16.99–20.35 wt.%) and low FeOt (3.92–4.94 wt.%). The majority of high-alumina arc basalts occur as porphyritic lavas, which are formed by the prior accumulation of plagioclase phenocrysts in the (basaltic-)andesite magmas [82,83]. This crystal accumulation process results in the observed Al2O3-enrichment and Fe-depletion, which defines the calc-alkaline trend [8]. Among the mineral phases in the high-alumina basalt/andesite, only plagioclase has positive Eu anomalies, which implies that positive Eu anomalies can indicate plagioclase accumulation in these lavas [82,84]. However, plagioclase accumulation can also be indicated by the minor or lack of Eu anomalies of (basaltic-)andesite in our case (Figure 6A and C). This is because the groundmass (representing the residual melt) has commonly negative Eu anomalies, which may have been (partially) cancelled-out by plagioclase accumulation [82,84]. Combined with the abundance and increasing plagioclase phenocryst contents from early andesite to late basaltic andesite (as suggested by field observation), we consider that the calc-alkaline signature of Nageng (basaltic-)andesite reflects extensive plagioclase accumulation.

Despite the Nageng andesites being isotopically and chemically similar to the basaltic andesites, there are still some discernible differences in their petrological features, chondrite-normalized REE patterns, and zircon trace element geochemistry. For example, the andesites have lower plagioclase and amphibole phenocryst contents, which suggests that they contain more interstitial melts in the magma chamber [85,86]. The andesites also have higher LREEs and lower HREEs (Figure 6), together with higher zircon Hf contents, but lower zircon Zr/Hf and Nb/Ta ratios (Figure 4), suggesting that they were located in the shallower part of the magma chamber with more interstitial melts prior to their eruption [46,55–57]. Taking their similar T_{ZircTi} (Figure 6) into account, these distinctions indicate that the Nageng (basaltic)andesites were successively extracted from a common magma chamber with a downward elevation of crystal accumulation.

The zircon core T_{ZircTi} (666–828 °C, avg. 735 °C) are only slightly higher than those of zircon rim (594–727 °C, avg. 667 °C), which suggests a very slow secular cooling rate (avg. 8.5 °C/Myr) for the Nageng magma chamber. Because these T_{ZircTi} values are below the solidus (~850 °C), “cold storage” is suitably applied to depict their melt-present history [87,88]. Cathodoluminescence images of zircons show an undisturbed oscillatory zoning pattern, indicating a (nearly) continuous crystallization. Abrupt magma recharge events can be precluded for the absence of zircon with increasing Ti from core to rim. However, the prolonged ~8 Myr storage of the andesitic magma requires an “open” architecture, where continuous mild magma replenishment offers heat to sustain extended melt-present lifespan [89]. We infer that these mild magma recharge events only aided in buffering the thermal dissipation, but they were insufficiently recorded by the zircon chemistry. Moreover, the ε_{Hf(t)} values of the zircon rims (~7.30 to −1.13) are more negative than those of the zircon cores (~1.28 to 1.32) (Figure 8), which indicates that the magma chamber had
undergone crustal contamination, as also supported by the presence of some xenocrystic zircons with very old ages (268–856 Ma).

Common mechanisms that can induce volcanic eruption include (1) the destabilization of magma chamber by tectonic interruption [90] and (2) thermal turbulation by an abrupt injection of hotter mafic magmas [86]. However, the latter mechanism is often accompanied by zircon resorption and overgrowth, and/or increased Ti content from the zircon core to rim [91–93], which is not found in our samples. Hence, when considering the coeval tectonic setting, the Nageng andesitic volcanism may have been triggered by seismic events pertaining to the closure of Paleo-Tethyan A’neymaqen Ocean and the related continent-continent collision at ca. 240 Ma [29,64,94].

6. Conclusions

1. Zircon geochronological data indicate that the zircon core (ca. 248 Ma) and rim (ca. 240 Ma) of antecrystic zircons from the Nageng (basaltic-)andesites have a pronounced age gap, which indicates that the main emplacement of the continental arc magma could have built up over protracted magma storage (~8 Myr) prior to the eruption.

2. Protracted magma storage at convergent margins, e.g., at Nageng, was generally accompanied by extensive plagioclase accumulation, which dictates igneous differentiation along the calc-alkaline trend.

3. The zircon trace element and Sr-Nd-Pb-Hf isotope features indicate that the Nageng andesites and basaltic andesites were successively extracted from a common magma chamber, which were continuously replenished by magmas that were derived from the juvenile mafic lower crust.

Supplementary Materials: The following are available online at www.mdpi.com/2075-163X/11/2/198/s1, Table S1: LA-ICP-MS zircon U-Pb isotope data of the Nageng andesites and basaltic andesites, Table S2: Zircon trace elements (ppm) of the Nageng andesites and basaltic andesites, Table S3: Whole-rock major (wt.%) and trace (ppm) element compositions of the Nageng andesites and basaltic andesites, Table S4: Whole-rock Sr-Nd-Pb isotope data of the Nageng andesites and basaltic andesites, Table S5: LA-(MC)-ICP-MS zircon Hf isotope data of the Nageng andesites and basaltic andesites, Figure S1: Selected major elements vs. CIW variation diagrams assessing the alteration effect on major elements.

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