

Repeated large-scale eruptions from a single compositionally stratified magma chamber: An example from Aso volcano, Southwest Japan

Katsuya Kaneko ^{a,*}, Hiroki Kamata ^a, Takehiro Koyaguchi ^b,
Masako Yoshikawa ^{c,d}, Kuniyuki Furukawa ^{a,1}

^a Graduate School of Human and Environmental Studies, Kyoto University, Sakyo-ku, Kyoto 606-8501, Japan

^b Earthquake Research Institute, University of Tokyo, 1-1-1 Yayoi, Tokyo 113-0032, Japan

^c Institute of Geothermal Sciences, Kyoto University, Noguchibaru, Beppu, 874-0903, Japan

^d The Institute for Frontier Research on Earth Evolution, Japan Agency for Marine-Earth Science and Technology (JAMSTEC), 2-15 Natsushima-Cho, Yokosuka, Kanagawa 237-0061, Japan

Received 13 January 2006; received in revised form 8 March 2007; accepted 10 May 2007

Available online 21 May 2007

Abstract

Aso volcano has the largest caldera (18×25 km) in the southwestern Japan Island Arc, which was formed as a result of four gigantic pyroclastic-eruption cycles. The latest eruption cycle (Aso-4 cycle, 90 ka) produced the most voluminous deposits (>600 km³) among the four eruption cycles. We investigated pre-eruptive magma chamber configurations and processes in compositionally stratified Aso-4 pyroclastic eruptions based on whole-rock chemical, phenocryst mineralogical, and Sr isotope analyses of juvenile clasts. Previous stratigraphical studies have suggested that the Aso-4 cycle is divided into two subcycles, in both of which eruption of voluminous silicic is followed by eruption of mafic magma. Geochemical and mineralogical results indicate that the pre-eruptive magma chamber was composed of three layers: silicic, hybrid, and mafic magma layers from the top to the bottom. The silicic magma layer was relatively homogenous, whereas the mafic magma layer was compositionally zoned where magma became more silicic with height. The hybrid magma was generated by mixing the silicic magma and the silicic end of the zoned mafic magma layer (i.e. the uppermost part of the mafic magma layer). The first subcycle began with evacuation of the upper silicic magma and successively proceeded to tap the lower hybrid and mafic magmas. The eruption of the first subcycle stopped before the whole magma chamber evacuated. During this subcycle, the hybrid magma and the upper part of the mafic magmas layer were selectively withdrawn and mixed with the overlying silicic magma. The second subcycle eruption started with the new hybrid magma, and followed by eruption of the mafic magma.

© 2007 Elsevier B.V. All rights reserved.

Keywords: Aso volcano; caldera; magma mixing; pyroclastic flow; zoned magma chamber

* Corresponding author. Tel.: +81 75 753 6874; fax: +81 75 753 6872.

E-mail addresses: katsuya@gaia.h.kyoto-u.ac.jp (K. Kaneko), kamata@gaia.h.kyoto-u.ac.jp (H. Kamata), tak@eri.u-tokyo.ac.jp (T. Koyaguchi), masako@bep.vgs.kyoto-u.ac.jp (M. Yoshikawa), kfuru@aichi-u.ac.jp (K. Furukawa).

¹ Present address. Aichi University, 10-31 Tsutsui 2-chome, Higashi-ku, Nagoya 461-8641, Japan.

1. Introduction

Contemporaneous eruptions of silicic and mafic magmas have been widely reported from large ignimbrite deposits (e.g., [Ritchey, 1980](#); [Hildreth, 1981](#)).

Their petrological features give important information about the magma plumbing system beneath the volcanoes. This study is concerned with a large ignimbrite eruption at Aso volcano, Southwest Japan.

Aso volcano has the largest caldera (18×25 km) in the southwestern Japan Island Arc (Fig. 1). It was formed by four pyroclastic-eruption cycles over a period of about 300 kyr (Ono and Watanabe, 1985). “Cycle”, which corresponds to “eruption” defined by Fisher and Schmincke (1984), means that an eruption event is not a single eruptive pulse but consists of many eruptive pulses without long time gaps between them. Although many geological, petrological, and geochemical studies have clarified the general features of eruption history and petrology of Aso volcano (e.g. Matsumoto, 1943; Ono et al., 1977; Watanabe, 1978; Ono and Watanabe, 1985; Kamata, 1997; Hunter, 1998; Furukawa, 2006), detailed physical conditions and processes within the magma plumbing system for each large eruption cycle are not fully understood.

In this paper, we focus on the most recent (90 ka) and largest (>600 km³) pyroclastic-eruption cycle, the Aso-4 cycle, among the four eruption cycles. The eruptions of the Aso-4 cycle are complex; it has six units, deposits of which are characterized by different lithological

features. The stratigraphical relationships of these units suggest that this eruption cycle is divided into two eruptive subcycles that are characterized by a progression from silicic to mafic magma. The purpose of this study is to clarify pre-eruptive magma chamber configurations and physical processes that result in these two subcycles in the Aso-4 cycle on the basis of results of whole-rock chemical, phenocryst mineralogical, and Sr isotope analyses of juvenile clasts. We believe that the example of the Aso-4 eruption provides an important constraint about the mechanisms of large-scale pyroclastic eruptions, such as the condition of termination of eruption.

2. Geology of the Aso-4 cycle

Aso volcano is located in central Kyushu, a large island in Southwest Japan (Fig. 1). The basement rocks in central Kyushu consist of Paleozoic to Mesozoic metamorphic rocks, Cretaceous granites, and Miocene volcanic and plutonic complexes. Activities of Aso volcano are characterized by four gigantic eruption cycles separated by time intervals between 100 ky to several 10 ky: Aso-1 (eruption volume 50 km³, age 266 ± 14 ka), Aso-2 (50 km³, 141 ± 5 ka), Aso-3 (>150 km³, 123 ± 6 ka), and

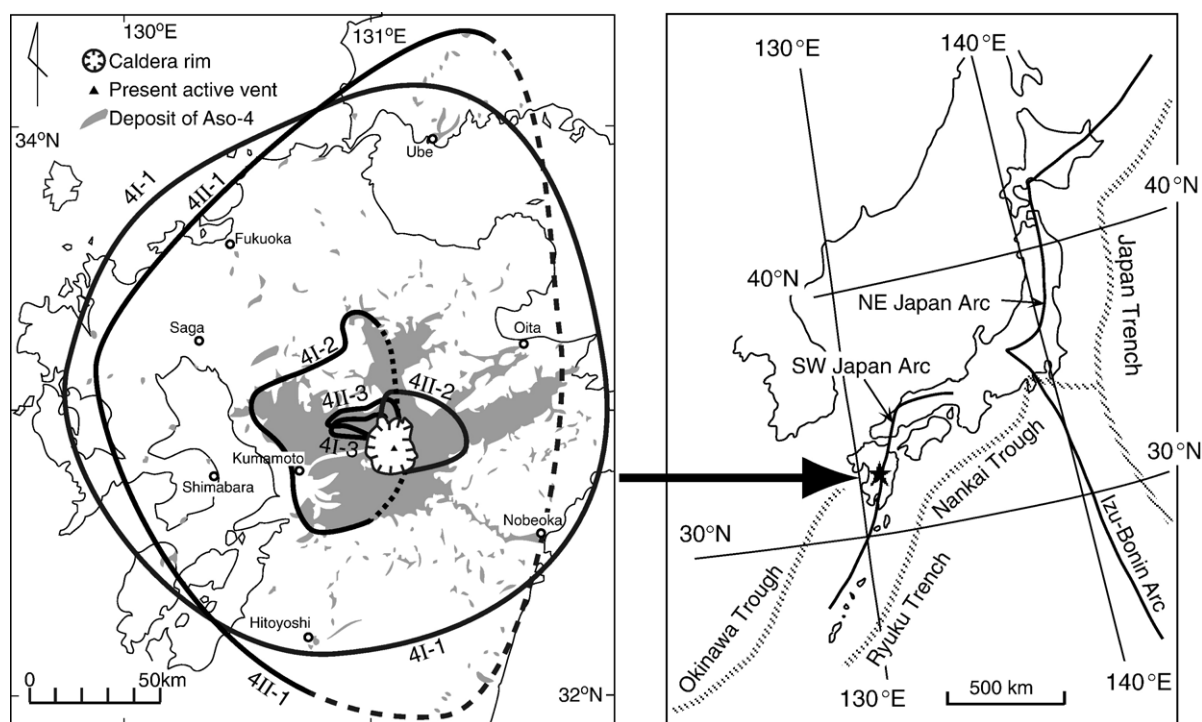


Fig. 1. The locality map of Aso volcano and the distribution of the Aso-4 cycle flow deposits (after Ono et al., 1977; Watanabe, 1978; Suzuki-Kamata and Kamata, 1990; Kamata, 1997). Distribution area of deposit of each unit is presented (solid line). The distribution of 4II-1P deposit is found only sporadically in the eastern area (Suzuki-Kamata and Kamata, 1990) (dashed line).

Aso-4 ($>600 \text{ km}^3$, $90 \pm 4 \text{ ka}$), and many minor eruptions between the large eruption cycles (Ono et al., 1977; Matsumoto et al., 1991; Committee of catalog of Quaternary volcano in Japan, 1999). After the most recent gigantic eruption cycle, the Aso-4 cycle, volcanic activities at Aso formed many relatively small stratified or monogenetic cones inside the caldera (Ono and Watanabe, 1985). Detailed geology and eruption history of Aso volcano in the eastern area, northern area, western area outside the caldera, and inside and around the caldera are summarized in Ono et al. (1977), Kamata (1997), Watanabe (1978), and Ono and Watanabe (1985). We review previous works related to the Aso-4 cycle here.

The Aso-4 cycle consists of ignimbrite deposits and co-ignimbrite ash fall deposits. The ignimbrite deposits widely cover northern Kyushu, and co-ignimbrite ash fall deposits more than 10 cm thick are present in Hokkaido at a distance of 1600 km from Aso caldera (Ono and Watanabe, 1983; Machida et al., 1985). The total volume of Aso-4 ejecta involving co-ignimbrite ash fall is estimated at more than 600 km^3 (Machida and Arai, 2003).

The pyroclastic-flow deposits of the Aso-4 cycle are divided into two subcycles that are characterized by a progression from silicic to mafic magma (Watanabe, 1978; Ono and Watanabe, 1983) (Table 1). Stratigraphic terminology for the Aso-4 cycle differs regionally. Moreover, we regrouped some units into one unit on the basis of petrological features in this paper. For these reasons, we newly define more systematical stratigraphic units for the Aso-4 cycle, which are consistent with the concept of eruption “sub-cycle”. A comparison between our units and the units by previous studies is shown in Table 1.

The first subcycle is divided into three units, 4I-1, 4I-2, and 4I-3, in stratigraphic order. Unit 4I-1 is the most voluminous ($>60 \text{ km}^3$) rhyolitic pumice flows among the

units of the Aso-4 cycle. It is widely distributed around the caldera (Fig. 1). 4I-1 occurs in various degree of welding: non-welded, highly vesiculated grey-white pumice facies to densely-welded tuff facies (Lipman, 1967; Ono et al., 1977; Watanabe, 1978). The welded facies are as a result of valley filling, and the non-welded facies are in the upper part of the deposits (Ono et al., 1977; Watanabe, 1978). 4I-1 in this paper corresponds to Oyatsu white pumice-flow, Koei ash-flow, Hatobira pumice-flow, and Yame pumice-flow in Watanabe (1978) and 4A in Ono et al. (1977) and Kamata (1997). Watanabe (1978) have divided the 4I-1 in this paper into the four units on the basis of field occurrence and petrographical features. However, differences of petrological features are very slight as presented in later section so that we consider the four units by Watanabe (1978) to be from the same magma and call them 4I-1 as a single unit. Unit 4I-2 is nonwelded, highly vesiculated grey pumice flow with small amount of scoria and banded pumice. Eruption volume is estimated at more than 4 km^3 (Watanabe, 1978). This unit is distributed in the western area outside the caldera. It corresponds to Motoigi pumice-flow in Watanabe (1978). Unit 4I-3 is non- to weakly-welded scoria flow with small amount of pumice and banded pumice. This unit is small ($\sim 0.5 \text{ km}^3$) and is distributed in the northwestern to western limited area near the caldera. The uppermost part of 4I-3 is slightly weathered, indicating that the activity of the Aso-4 cycle shortly reposed after the eruption of 4I-3 (Watanabe, 1978). This unit corresponds to Benri scoria-flow in Watanabe (1978) and part of 4T in Kamata (1997).

The second subcycle is divided into three units, 4II-1, 4II-2, and 4II-3, in stratigraphic order. Unit 4II-1 is non-welded, lithic-rich pumice flow. It corresponds to Tosu orange pumice-flow in Watanabe (1978) and part of 4T in Kamata (1997). This unit is the second largest ($\sim 10 \text{ km}^3$) in the Aso-4 cycle. This flow is characterized

Table 1

Stratigraphy and eruption volumes of pyroclastic flows in the Aso-4 cycle based on the previous works (Watanabe, 1978, 2001)

Subcycle	Units	Volume (km^3)	Corresponding units in previous works
Second subcycle	4II-3 Non- to weakly welded scoria flow with banded scoria	~ 0.2	Kunomine scoria-flow in W; part of 4T in K
	4II-2 Densely-welded silicic pyroclastic flow	>5	4B in O and K
	4I-1 Non-welded orange pumice flow (very altered)	~ 10	Tosu orange pumice-flow in W; part of 4T in K
First subcycle	4I-3 Non- to weakly welded scoria flow with pumice and banded pumice	~ 0.5	Benri scoria-flow in W; part of 4T in K
	4I-2 Non-welded pumice flow with banded pumice and scoria	>4	Motoigi grey pumice-flow in W
	4I-1 Non- to densely-welded pumice flow	>60	4A in O and K; Oyatsu white pumice-flow, Koei ash-flow, Hatobira pumice-flow, and Yame pumice-flow in W

The volume of Unit-4B is roughly estimated by us on the basis of geological studies by Ono et al. (1977) and Kamata (1997). In the column of corresponding units in previous works, O, W, and K refer to Ono et al. (1977), Watanabe (1978), and Kamata (1997), respectively.

Table 2

Whole-rock compositions of juvenile fragments in the Aso-4 cycle

Sample	TK44	TK08	NJ01	ON08	910901	PM04	910901	910901	303250		911208	911208
Unit and Type	4I-1P	4I-1P	4I-1P	4I-1P	4I-1P	4I-1P	4I-1P	4I-1P	4I-1P	YG01	08-1	08-2
										4I-2P	4I-2P	4I-2S
<i>Major element (wt.%)</i>												
SiO ₂	67.21	69.00	67.24	67.18	67.41	67.49	69.03	69.18	68.34	65.70	67.71	55.34
TiO ₂	0.54	0.51	0.53	0.53	0.54	0.53	0.51	0.51	0.52	0.57	0.58	0.98
Al ₂ O ₃	15.85	15.52	15.77	15.74	15.76	15.93	14.94	14.94	15.44	17.06	15.76	17.66
FeO*	2.61	2.15	2.49	2.51	2.50	2.45	2.39	2.38	2.41	2.67	2.79	8.02
MnO	0.13	0.11	0.13	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.18
MgO	0.94	0.69	0.92	0.93	0.91	0.87	0.88	0.90	0.88	0.89	1.12	3.38
CaO	2.07	1.29	1.95	2.06	2.00	1.90	1.89	1.90	1.90	1.85	2.35	6.71
Na ₂ O	4.67	4.33	4.68	4.58	4.75	4.65	4.50	4.53	4.59	4.20	4.30	4.01
K ₂ O	4.05	5.21	4.19	4.16	4.08	4.21	3.83	3.83	4.02	4.13	4.42	2.00
P ₂ O ₅	0.13	0.08	0.12	0.13	0.12	0.12	0.12	0.12	0.12	0.17	0.15	0.41
Total	98.19	98.87	98.01	97.94	98.19	98.26	98.22	98.41	98.33	97.36	99.30	98.70
<i>Trace element (ppm)</i>												
Rb	135.1	195.4	140.0	133.8	135.8	137.2	129.4	131.1	134.2	140.5	133.9	58.2
Sr	385.7	218.1	365.3	379.5	369.8	354.8	353.9	357.5	356.2	304.6	382.6	776.8
Y	33.4	31.7	31.3	31.6	31.8	31.7	29.9	30.7	31.2	34.4	31.9	30.9
Zr	268.5	296.6	275.3	268.1	265.6	270.5	255.0	258.3	264.4	323.2	271.5	155.5
Nb	11.6	13.7	11.9	11.5	11.8	11.3	12.2	10.8	11.1	13.6	11.6	6.6
Ba	714.5	824.6	705.3	691.0	718.4	730.1	691.6	702.8	716.5	882.1	688.7	386.0
V	35.0	23.8	31.9	29.2	31.7	32.1	31.0	28.5	30.3	23.1	29.0	173.6
Cr	0.0	0.9	3.1	0.8	0.0	1.0	0.0	0.0	0.5	5.2	0.8	11.8
Ni	8.4	6.3	10.0	12.0	10.6	8.9	11.0	9.9	9.4	10.7	11.9	7.1
Pb	12.8	22.7	16.4	20.5	17.2	18.1	17.9	19.4	18.8	17.8	20.7	6.8
Sample	KC01p	KC01s	KC05	910928	910928	910928	910928			910928	910929	
Unit and Type	4I-3P	4I-3S	4I-3S	01	02b-5/8	02b-7/8	03	TK41	NJ03	07	07	KC03
				4I-3S	4I-3S	4I-3S	4I-3S	4II-2P	4II-2P	4II-3S	4II-3S	4II-3S
<i>Major element (wt.%)</i>												
SiO ₂	65.21	55.34	56.08	55.79	53.03	53.11	51.52	67.92	65.87	49.07	49.28	52.71
TiO ₂	0.68	0.98	0.85	0.98	1.07	1.08	1.13	0.54	0.56	1.33	1.28	1.15
Al ₂ O ₃	16.65	17.66	18.36	18.14	18.01	18.49	18.28	16.36	16.37	19.18	19.15	20.20
FeO*	3.76	8.02	7.76	7.86	9.16	8.87	9.31	2.45	2.73	10.91	10.65	8.10
MnO	0.13	0.18	0.13	0.18	0.19	0.19	0.20	0.09	0.13	0.22	0.21	0.12
MgO	1.45	3.38	2.96	3.06	3.81	3.53	4.07	0.86	0.98	4.55	4.49	4.13
CaO	3.38	6.71	7.32	5.92	7.66	7.50	8.12	2.09	1.96	7.60	7.71	6.75
Na ₂ O	4.20	4.01	3.54	3.94	3.72	3.81	3.76	4.69	4.66	3.50	3.60	3.76
K ₂ O	3.46	2.00	2.28	2.18	1.61	1.60	1.47	4.65	3.92	1.12	1.15	1.25
P ₂ O ₅	0.20	0.41	0.32	0.38	0.44	0.54	0.61	0.13	0.13	0.73	0.72	0.85
Total	99.12	98.70	99.62	98.40	98.69	98.74	98.45	99.79	97.30	98.20	98.24	99.02
<i>Trace element (ppm)</i>												
Rb	117.1	60.0	73.1	65.7	47.0	46.5	41.1	172.7	134.2	28.6	29.1	35.3
Sr	476.1	719.3	622.1	658.7	747.1	830.7	857.5	394.8	320.5	747.5	765.3	632.0
Y	31.1	31.0	22.2	31.4	30.1	32.6	32.8	27.4	33.9	36.1	35.3	38.6
Zr	244.0	156.4	168.2	170.7	135.1	142.1	134.3	261.9	286.3	125.5	124.9	114.5
Nb	10.6	5.8	7.4	7.4	5.6	4.8	5.2	11.4	11.4	5.3	5.0	8.1
Ba	678.2	426.1	454.8	493.7	381.4	349.0	345.0	741.9	768.1	301.6	314.3	532.0
V	49.6	179.0	199.4	174.2	222.1	198.7	206.9	35.1	30.4	250.5	249.1	235.6
Cr	6.7	10.0	18.8	11.3	17.1	9.7	12.0	0.2	1.0	17.0	14.7	15.6
Ni	8.9	6.8	4.4	7.8	4.5	5.9	3.9	10.3	11.6	3.7	5.3	4.8
Pb	17.9	5.8	12.2	2.4	6.7	5.2	4.1	21.1	21.0	7.1	6.3	7.0

All the whole-rock data of major elements presented in Fig. 2 are recalculated to 100% volatile-free with all Fe as FeO.

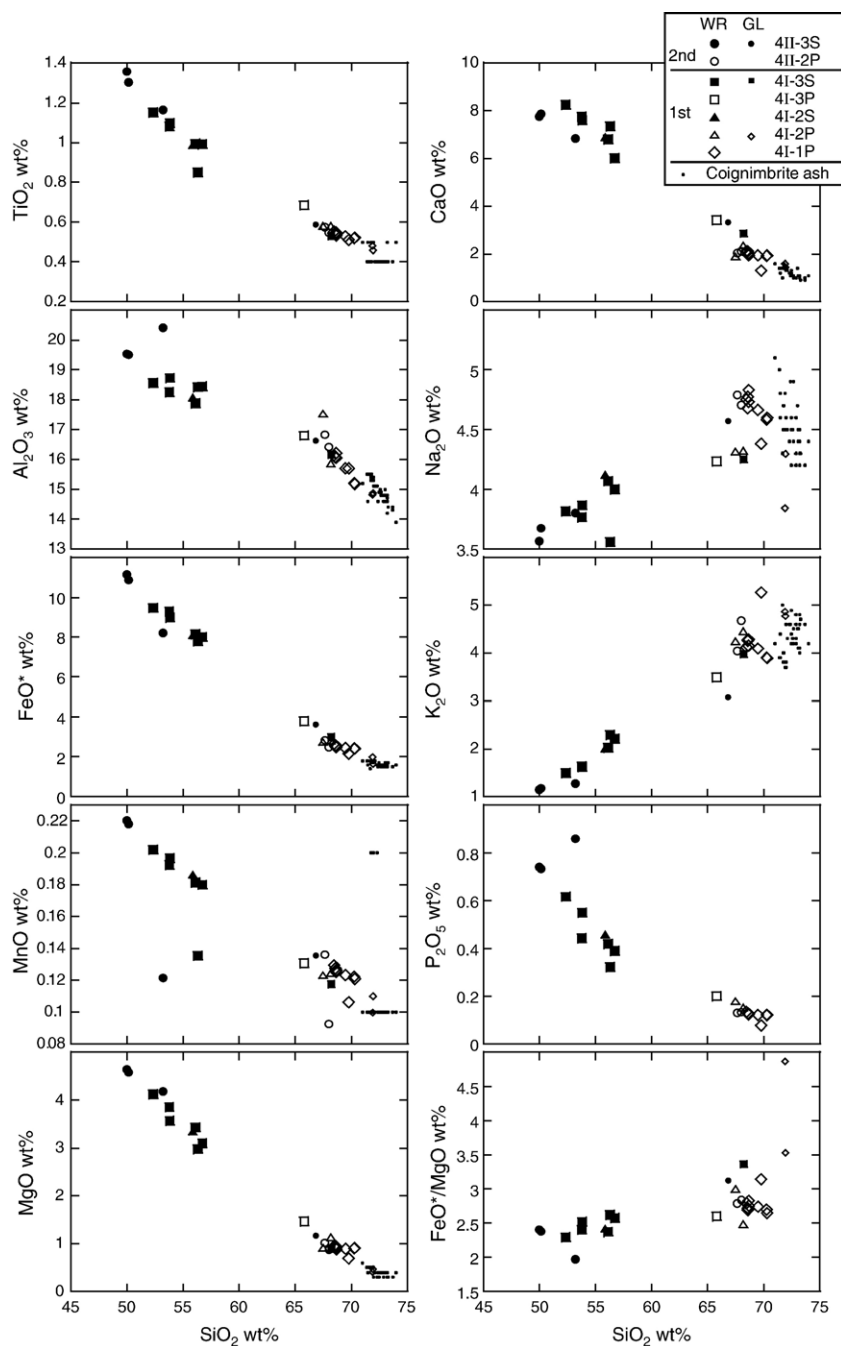


Fig. 2. Oxide-SiO₂ diagrams of the whole-rock and glass chemical composition of the Aso-4 cycle. The data show in Tables 2 and 3. The glass data of co-ignimbrite ash are from Machida and Arai (2003). In glass analyses, P₂O₅ contents are not obtained. Whole-rock (WR) and glass (GL) data are shown as large and small symbols, respectively.

by intensely altered orange pumice and its wide distribution of proximal breccial facies in the northern to eastern area outside the caldera (Watanabe, 1978; Suzuki-Kamata and Kamata, 1990). The distribution of 4II-1P deposit roughly coincides with that of 4I-1P,

although it is found only sporadically in the eastern area (Suzuki-Kamata and Kamata, 1990) (Fig. 1). Unit 4II-2 consists of densely-welded pyroclastic flows. It is equivalent to 4B in Ono et al. (1977) and Kamata (1997). This unit is distributed in the northeastern to

southeastern area outside the caldera. The eruption volume is estimated at more than 5 km³. Unit 4II-3 is non- to weakly-welded scoria flow with small amount of pumice and banded pumice. It corresponds to Kunomine scoria-flow in Watanabe (1978) and part of 4T in Kamata (1997). This unit is the smallest (~0.2 km³) and is distributed in the northwestern to western limited area near the caldera. The co-ignimbrite ash fall deposits with very wide distribution correspond to 4I-1 and/or 4II-1 on the basis of refractory indices of volcanic glass and hornblende and orthopyroxene crystals (Machida et al., 1985).

Pyroclastic flows of the Aso-4 cycle contain large amount of lithic fragments (Ono et al., 1977). Ono et al. (1977), Kamata (1997), and Watanabe (1978) reported lithologies of lithic fragments in the eastern, northern, and western areas outside the caldera, respectively. These descriptions of lithic fragments indicate that lithologies of lithic fragments differ regionally. In the eastern area, for example, pyroxenite and dunite clasts are present that are absent in the other areas. In contrast, granite clasts are not present in the eastern area while they are present elsewhere. The lithologic heterogeneity of lithic fragments may suggest that the magmas of the Aso-4 cycle were erupted from either a ring fissure vent or multiple vents.

3. Analytical method and sample

Whole-rock compositions were determined by X-ray fluorescence spectrometers, Rigaku RIX2100 at Fuji Tokoha University (Sano, 2002). All the whole-rock data (Table 2) presented in Fig. 2 are recalculated to 100% volatile-free with all Fe as FeO. Mineral and glass

compositions were determined by electron probe microanalysis at Earthquake Research Institute, the University of Tokyo (JEOL8800) and Kobe University (JEOL8900), respectively. We used a focused beam for determination of mineral composition. We obtained glass compositions as average compositions of 12 to 32 analytical points by defocused beams (about 10 µm in diameter) because of preventing loss of Na (Table 3). Whole-rock data of Strontium isotope (⁸⁷Sr/⁸⁶Sr) were obtained by VG Micromass 54E mass spectrometer at Geological Survey of Japan. In this analysis, we carefully removed lithic fragments that can be recognized from the samples. The ⁸⁷Sr/⁸⁶Sr ratios of the NIST987 standard were 0.710272 ± 0.000034 (2σ). Strontium isotope data of fiamme and volcanic glass were obtained using a Triton mass spectrometer at Institute of Geothermal Sciences, Kyoto University (Shibata et al., 2003; Takahashi and Yoshikawa, 2005). The ⁸⁷Sr/⁸⁶Sr ratios of the NIST987 standard were 0.710258 ± 0.000012 (2σ). All the data have been normalized to NIST987 ⁸⁷Sr/⁸⁶Sr = 0.710278.

All the samples except 4II-2 were used for petrographical observations and chemical analyses are on non- to weakly-welded, vesiculated lumps of pumice or scoria. Ejecta of 4II-2 are densely welded and whole-rock data of fiamme were obtained. We obtained whole-rock data of the samples from 4I-1, 4I-2, 4I-3, 4II-2, and 4II-3. On the other hand, we elected to exclude samples from 4II-1 from bulk analysis because of intense weathering. Phenocrysts minerals in the 4II-1 pumices are fresh enough for chemical analyses. Hereafter, we refer to pumices of 4I-1, 4I-2, 4I-3, and 4II-1 as 4I-1P, 4I-2P, 4I-3P, and 4II-1P, fiamme of 4II-2 as 4II-2P, and scoriae of 4I-2, 4I-3, and 4II-3 as 4I-2S, 4I-3S, and 4II-3S, respectively.

Table 3
Glass compositions of 4I-1P, 4I-3S and 4II-3S

Sample	NJ01		4AFDI		KC01		KC03	
Unit and type	4I-1P		4I-1P		4I-3S		4II-3S	
No. of analysis points	26		32		23		12	
	Ave	Stdev	Ave	Stdev	Ave	Stdev	Ave	Stdev
SiO ₂	71.95	0.63	71.91	1.14	68.20	2.00	66.83	0.82
TiO ₂	0.46	0.06	0.48	0.12	0.53	0.08	0.59	0.04
Al ₂ O ₃	14.86	0.25	14.82	0.20	16.16	0.83	16.64	0.41
FeO*	1.64	0.23	1.99	0.59	3.00	0.62	3.65	0.32
MnO	0.11	0.03	0.10	0.02	0.12	0.03	0.14	0.02
MgO	0.47	0.07	0.41	0.07	0.89	0.44	1.17	0.27
CaO	1.44	0.14	1.60	0.23	2.86	0.94	3.34	0.46
Na ₂ O	4.30	0.46	3.84	0.27	4.25	0.36	4.57	0.30
K ₂ O	4.77	0.12	4.86	0.38	3.98	0.53	3.08	0.45
Analytical total	97.57	0.74	97.49	0.78	95.76	2.73	98.21	0.57

Averaged compositions (Ave) in analysis points by EPMA are recalculated to 100% volatile-free with all Fe as FeO. Standard deviations (Stdev) are also shown.

4. Whole-rock and glass chemistry

Fig. 2 shows major element variations of whole-rock and glass compositions (Tables 2 and 3). The data of co-ignimbrite ash glass are from Machida and Arai (2003). In the whole-rock compositional variations, there is a gap of SiO_2 content between 57 and 66 wt.%. All the mafic scoriae (4I-2S, 4I-3S, and 4II-3S) have SiO_2 wt.%=50–57, whereas the other ejecta (pumices or fiammes of 4I-1P, 4I-2P, 4I-3P, and 4II-2P) are more silicic (SiO_2 wt.%=66–70). The SiO_2 content of the scoriae of the first subcycles (4I-2S and 4I-3S) vary from 53 to 57 wt.% and that of the second subcycles (4II-3S) vary from 50 to 55 wt.%, and they collectively form a continuous single trend; exceptionally, the scoriae of the second subcycle have slightly lower CaO than those of the first subcycles in the diagram of CaO vs SiO_2 . In the

other ejecta, 4I-1P erupting at first in the Aso-4 cycle is the most silicic and uniform (within the range of 68–70 wt.% SiO_2). Other pumices and fiammes (4I-2P, 4I-3P, and 4II-2P) are less silicic than 4I-1P and they are on a line connected between the silicic end of the mafic scoriae and 4I-1P.

Glass compositions of 4I-1P, 4I-3S and 4II-3S are more silicic than the corresponding whole-rock compositions. The glasses of 4I-3S and 4II-3S are remarkably silicic and their compositions are within the whole-rock compositional range of the pumice samples. The whole-rock compositions of the scoriae are largely affected by high phenocryst abundance (see Section 5 for modal compositions). The glass compositions of 4I-1P are equivalent to those of co-ignimbrite ash. Na_2O contents of 4I-1P glass and some co-ignimbrite ash glasses are systematically lower than the whole-rock compositions

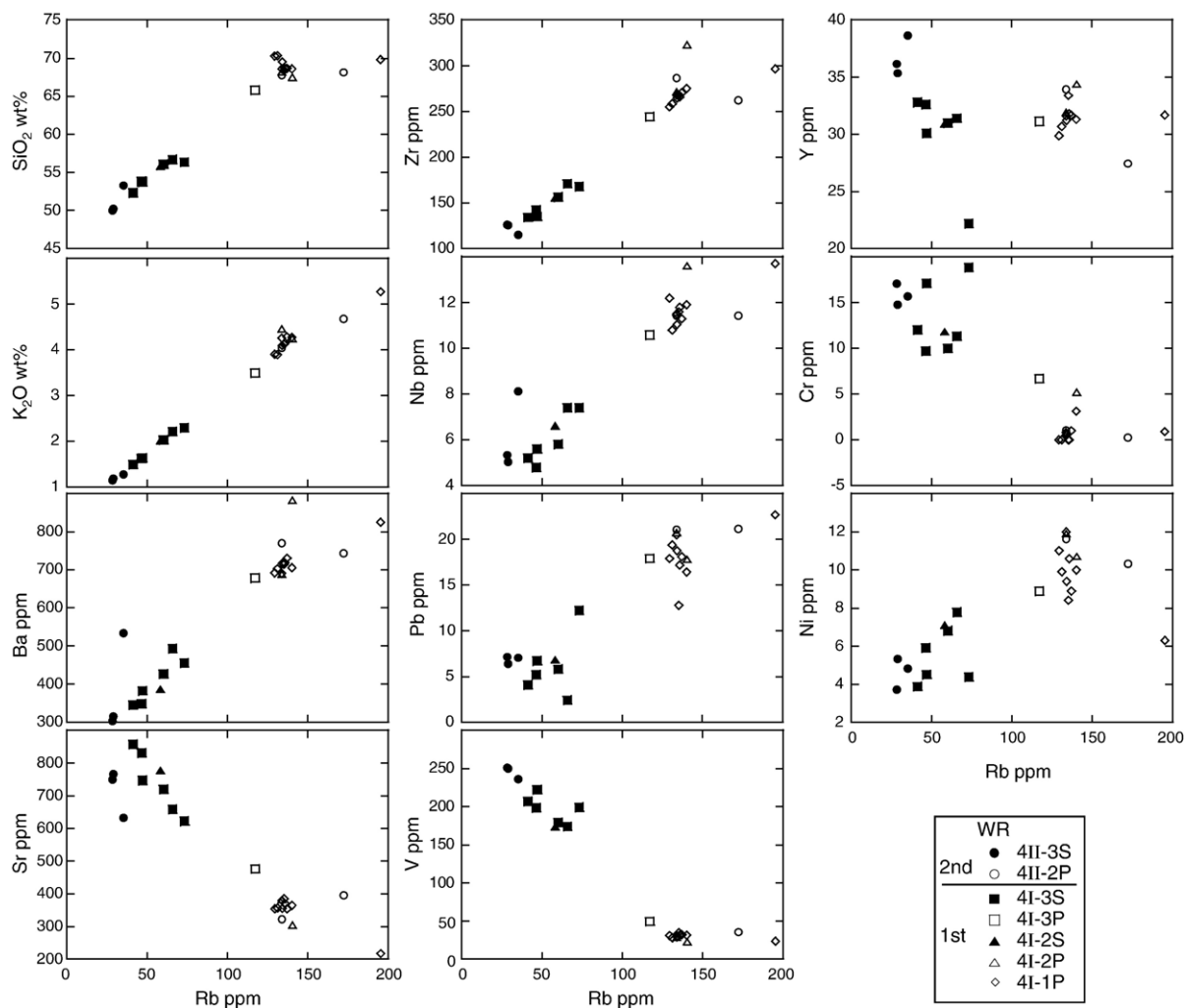


Fig. 3. SiO_2 , K_2O , and trace elements vs Rb diagrams of the whole-rock chemical composition of the Aso-4 cycle. The data show in Table 2.

of the 4I-1P. We consider that this difference may be due to loss of Na during electron microprobe analyses for glass compositions.

Fig. 3 shows trace element variations of whole-rock compositions (for data source see Table 3). Contents of highly incompatible elements (K_2O , Ba, Zr, and Nb) as well as SiO_2 content linearly increase with increasing Rb. Contents of elements which are compatible with plagioclase (Sr) and mafic and/or oxide minerals (Cr, V) linearly decrease with Rb except that Ni contents is higher in the pumices and fiammes than the mafic scoriae. Y content does not show clear correlation with Rb; it is higher in the most mafic samples (4II-3S) than the other samples. Analytical results of $^{87}Sr/^{86}Sr$ (Table 4 and Fig. 4) show that $^{87}Sr/^{86}Sr$ values of all the whole-rock samples and the glass of 4I-1P are within the $2\sigma_{\text{mean}}$ analytical uncertainty (0.74090–0.74115).

5. Petrography and chemistry of phenocryst minerals

5.1. Petrographical features

Petrographical features of juvenile ejecta are summarized in Table 5. Petrographical features of groundmass are described as follows. Individual clasts of 4I-1P have homogenous groundmass of highly vesiculated colorless glass. Although some of pumices, scoriae, and banded pumices coexist at the same stratigraphic horizons in the units of 4I-2, 4I-3, and 4II-3, individual lumps of pumice and scoria without banding have homogenous groundmass. Groundmasses of 4I-2P and 4I-3P are highly vesiculated, colorless glass, of 4I-2S and 4I-3S moderately vesiculated, brown glass, and of 4II-3S highly vesiculated, brown glass. 4II-1P has groundmass of highly vesiculated, much altered, orange glass and it is difficult to judge whether the groundmass is homogenous or not, but obvious heterogeneities in the groundmass are not observed at least. 4II-2 is a densely-welded pyroclastic-flow deposit and has a eutaxitic texture. Groundmasses within fiammes (4II-2P) are homogenous.

Table 4
Strontium isotope ratios with $2\sigma_{\text{mean}}$ errors

Sample	Unit and type	Analysis	$^{87}Sr/^{86}Sr$	
KC03	4II-3S	WR	0.704115	+/-12
91092907	4II-3S	WR	0.704104	+/-15
NJ03	4II-2P	WR	0.704098	+/-08
KC01s	4I-3S	WR	0.704077	+/-13
NJ01	4I-1P	WR	0.704097	+/-13
NJ01	4I-1P	Glass	0.704091	+/-06
ON08	4I-1P	WR	0.704090	+/-14

WR stands for whole-rock composition.

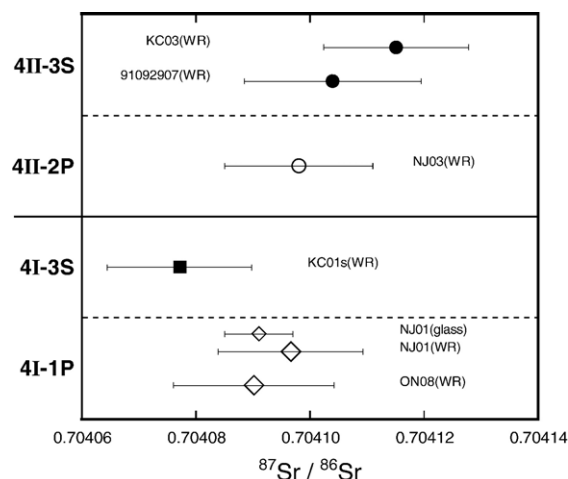


Fig. 4. Strontium isotope ratio ($^{87}Sr/^{86}Sr$). Error bars show $2\sigma_{\text{mean}}$ errors.

4I-1P contains orthopyroxene, plagioclase, hornblende, magnetite, and ilmenite as phenocrysts and very small amount of clinopyroxene and apatite phenocrysts. All the other samples contain olivine, clinopyroxene, orthopyroxene, plagioclase, hornblende, magnetite, ilmenite, and very small amount of apatite as phenocrysts. Phenocryst abundances in the juvenile clasts are obviously different for scoriae (4I-3S and 4II-3S) and the others (4I-1P, 4I-2P, 4II-1P, and 4II-2P) as pointed out by Watanabe (1979). Modal compositions of total phenocrysts of the scoriae are 10 to 37 vol.%, while in the other ejecta about 5 to 10 vol.%.

Chemical features of phenocryst minerals of seven kinds of the samples (4I-1P, 4I-2P, 4I-3P, 4I-3S, 4II-1P, 4II-2P, and 4II-3S) will be described below. We consider that the chemical features of phenocryst minerals of 4I-2S are identical to those of 4I-3S on the basis of whole-rock composition and petrography under microscope. We point out in advance that 4I-2P, 4I-3P, 4II-1P, and 4II-2P have many common features of phenocryst minerals. Their SiO_2 contents are lower than 4I-1P in whole-rock composition although compositional gap is not clear in whole-rock variation diagrams (Fig. 2). When we show their common features, we refer to them as “the pumices with intermediate composition”.

5.2. Olivine, clinopyroxene, and orthopyroxene

Olivine phenocrysts are present except 4I-1P. They occur as anhedral to subhedral crystals with up to 1 mm in diameter. Mg-number ($Mg\# = 100 \times Mg/(Mg + Fe^*)$ in mole) of olivine phenocrysts ($Mg\# = 72-75$), modal composition (tr. ~0.5), and maximum size (1 mm) hardly change throughout all the rocks.

Table 5

Petrographical summary of major juvenile fragments of the units in the Aso-4 cycle

Unit and type	Phases	Form	Mode, vol. %	Maximum size, mm	Zoning	Groundmass
4I-1P	pl	an/eu	4–8	3	basically normal and oscillatory	Highly vesiculated, colorless glass in non-welded parts.
	cpx	sub/eu	tr.	0.2	unzoned	
	opx	sub/eu	0.2–0.8	1.2	reverse	
	hb	sub/eu	0.1–0.5	2	unzoned	
	opq	an/eu	0.3–0.8	0.5 (mt & il)	unzoned	
4I-2P	pl	an/eu	~6	2	unzoned–normal and oscillatory	Highly vesiculated, colorless glass.
	ol	an/sub	tr.	1	unzoned	
	cpx	sub/eu	tr.	0.8	reverse	
	opx	sub/eu	~0.5	0.8	reverse	
	hb	sub/eu	~0.5	2	unzoned	
	opq	an/eu	~0.3	0.5 (mt) 0.2(il)	unzoned	
4I-3S	pl	sub/eu	8–25	2	unzoned–slightly normal and oscillatory	Moderately vesiculated, brown glass
	ol	an/sub	tr.	1	unzoned	
	cpx	an/eu	0.5–1.5	1	reverse	
	opx	sub/eu	0.5–2	0.5	reverse	
	hb	an/eu	0.7–4	1.5	increase of Al ^{IV} to rim	
	opq	an/eu	0.5–4	0.5 (mt) 0.2(il)	unzoned in mt. rarely il with high X _{il} core	
4II-1P	pl	an/eu	~7	2.5	basically normal and oscillatory	Highly vesiculated, much altered glass.
	ol	an/sub	tr.	1	unzoned	
	cpx	sub/eu	0.2	1.2	reverse	
	opx	sub/eu	0.6	1	reverse	
	hb	sub/eu	1.5	3	unzoned– Increase of Al ^{IV} to rim	
	opq	an/eu	0.6	0.5 (mt) 0.2 (il)	unzoned	
4II-2P	pl	an/eu	4–8	2	unzoned–normal and oscillatory	Densely welded, eutaxitic texture. Fiamme is glassy like obsidian.
	ol	an/sub	tr.	0.5	unzoned	
	cpx	an/eu	0.2–1	0.8	reverse	
	opx	sub/eu	0.5–1	1.2	reverse	
	hb	sub/eu	0.5–1.3	3	unzoned– Increase of Al ^{IV} to rim	
	opq	an/eu	~0.5	0.8 (mt) 0.4 (il)	narrow lamellae in mt	
4II-3S	pl	an/eu	10–20	2	unzoned–slightly normal and oscillatory	Highly vesiculated, brown glass.
	ol	an/sub	tr.–0.5	1	unzoned	
	cpx	an/eu	0.3–4	1	reverse	
	opx	sub/eu	~2	0.5	reverse	
	hb	an/eu	0.5–3.5	2	increase of Al ^{IV} to rim	
	opq	an/eu	1–3	0.5 (mt) 0.3 (il)	unzoned in mt. rarely il with high X _{il} core	

Data of mode of phenocrysts except 4II-2P are from Watanabe (1979). Mode of vesiculated pumice and scoriae (vesiculated pumice of 4I-1P and all of 4I-2P, 4I-3S, 4II-1P, and 4II-3S) is determined by “crushing method” proposed by Lipman (1967), (Watanabe, 1979) and mode of fiamme of welded-tuff (welded tuff of 4I-1P and 4II-2P) by point-counting method. In the column of form, “an”, “sub”, and “eu” show anhedral, subhedral, and euhedral, respectively.

In the most silicic pumice (4I-1P), only one sample (TK44) contains a small amount of small clinopyroxene phenocrysts (up to 0.2 mm), whose Mg# range between 70 and 80. Compositional zoning is weak. Clinopyroxene phenocrysts in the other rocks have common features in some aspects. They have a compositional range of Mg#=70–80 with the highest abundance

around Mg#=77–80 (Fig. 5). Clinopyroxenes with Mg#=77–80 show reverse zoning of up to 4 in Mg#. The maximum crystal sizes are 0.8–1.2 mm in length.

Orthopyroxene phenocrysts are commonly contained by all the rocks. They occur as subhedral to euhedral crystals with up to 0.8–1.2 mm in length. They occasionally show glomero-porphyritic texture with

plagioclase and magnetite crystals. Fig. 6 shows frequency diagrams of Mg# of orthopyroxene phenocrysts. Orthopyroxenes of the most silicic pumice (4I-1P) have a narrow compositional range of Mg# = 72–75 with a histogram peak at Mg# = 73–74. Individual crystals show reverse zoning of up to 2 in Mg#. Orthopyroxenes of the mafic scoriae (4I-3S and 4II-3S) have a compositional range of Mg# = 72 to 77 with a histogram peak at Mg# = 75–76, and individual crystals show reverse zoning of up to 4 in Mg#. Orthopyroxenes of the pumices with intermediate compositions (4I-2P, 4I-3P, 4II-1P and 4II-2P) have a compositional range of Mg# = 72–77 with a histogram peak at Mg# = 73–75. Individual crystals show reverse zoning of up to 4 in Mg#. One sample of 4II-1P contains a small amount of orthopyroxenes with low Mg# down to 67, which are not observed in the other samples.

5.3. Plagioclase

Plagioclase phenocrysts occur in all the ejecta. Oscillatory zoning and honeycomb structure are com-

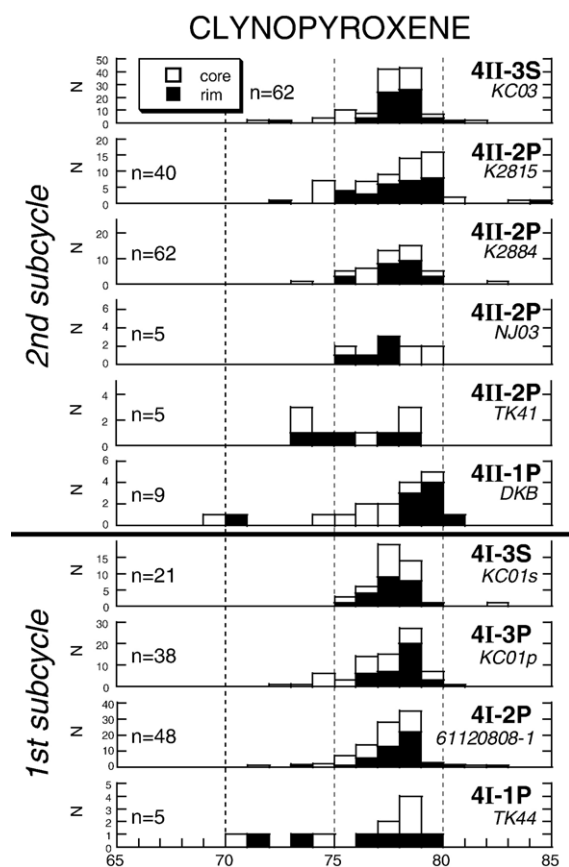


Fig. 5. Frequency diagrams of the Mg-number of clinopyroxene phenocrysts. *n* shows number of analyzed crystals.

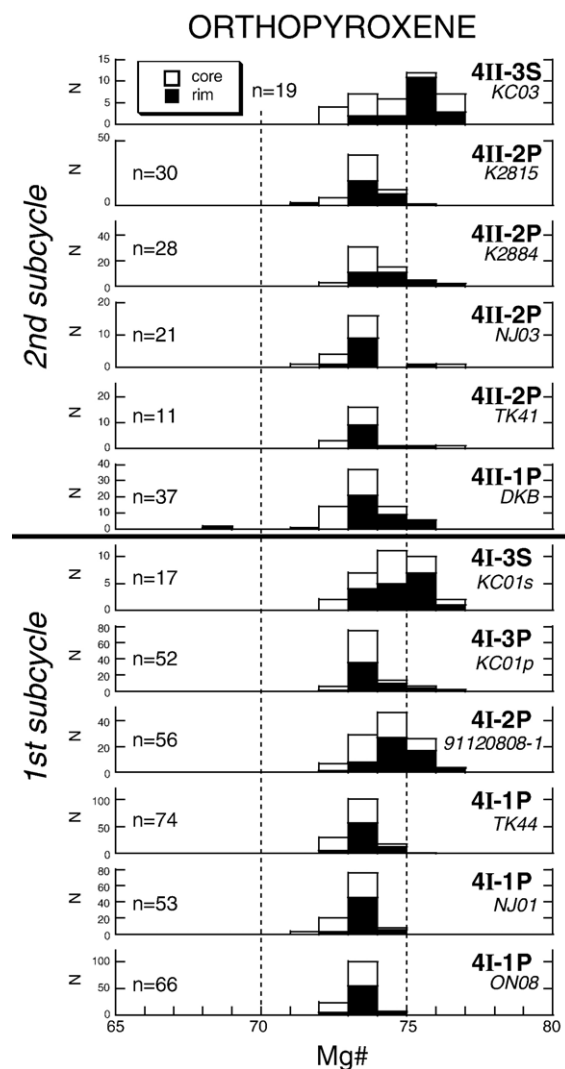


Fig. 6. Frequency diagrams showing the Mg-number of orthopyroxene phenocrysts. *n* shows number of analyzed crystals.

monly observed. Fig. 7 shows variations of anorthite content of plagioclase phenocrysts ($An = 100 \times Ca / (Ca + Na + K)$ in mole).

Plagioclase phenocrysts of the most silicic pumice (4I-1P) occur as anhedral (corroded texture) to euhedral crystals with up to 3 mm in length. A bimodal distribution of An content is obvious; histogram peaks are at An 35 and An 45. A minor proportion (~15% of analyzed crystals) of plagioclase phenocrysts has calcic cores (up to An 68). Compositional zonations tend to be normal with oscillatory zoning of amplitude of up to 10 in An.

Plagioclase phenocrysts of the mafic scoriae (4I-3S and 4II-3S) occur as subhedral to euhedral crystals with up to 2 mm in length. Anhedral crystals are hardly observed. They have distinct compositional features from

those of 4I-1P. An contents of plagioclase in the scoriae vary between An 34 and An 94. Three histogram peaks of An content at An 53, An 62, and An 90 are present, while 4II-3S additionally has a histogram peak at An 45. Compositional zonation is weaker than 4I-1P; compositional zonations of plagioclases with An content less than about 70 tend to be normal with oscillatory zoning of amplitude of up to 5 in An, and those of plagioclases with An content more than 70 are very weak.

Plagioclase phenocrysts of the pumices with intermediate compositions (4I-2P, 4I-3P, 4II-1P and 4II-2P) have common features. They occur as anhedral (corroded texture) to euhedral crystals with up to 2–

2.5 mm in length. They show trimodal distributions; two major peaks are at An 35 and An 45 that are the peaks of 4I-1P, and 4I-3P, 4II-1P, and 4II-2P have a minor peak and 4I-2P have a major peak at An 52–56 that is the peak of 4I-3S and 4II-3S. Small amount of plagioclase have high An contents up to 76. In particular, 4I-3P has very calcic plagioclase up to An 96. Compositional zonations of plagioclases with An content less than 68 tend to be normal with oscillatory zoning of amplitude of up to 10 in An, while those of plagioclases with An content more than 68 are very weak. In summary, plagioclase phenocrysts of the pumices with intermediate compositions (4I-2P, 4I-3P, 4II-1P and 4II-2P) have both features of those of the most silicic pumice (4I-1P) and the mafic scoriae (4I-3S and 4II-3S).

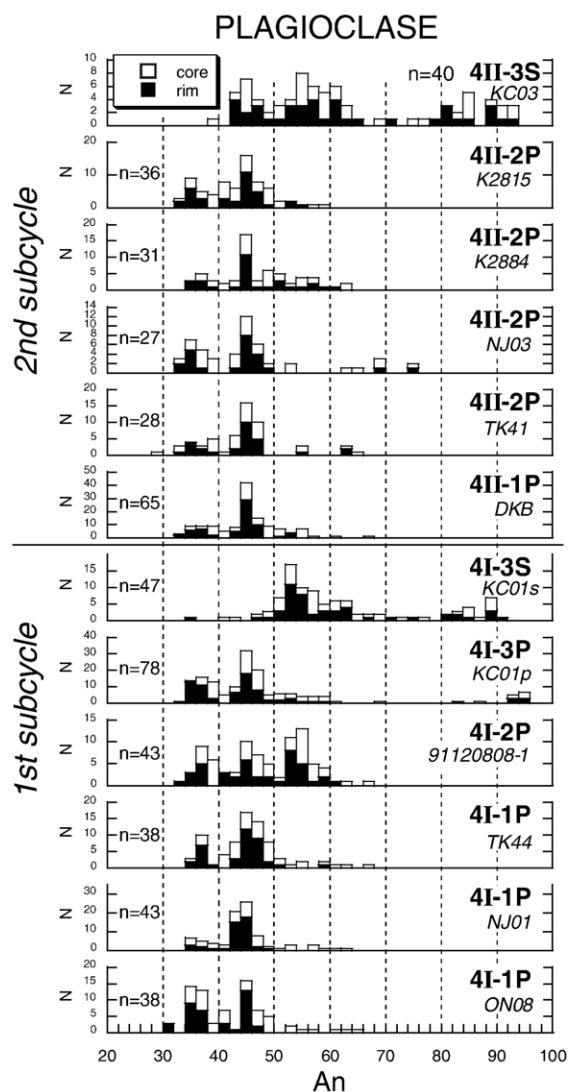


Fig. 7. Frequency diagrams showing the anorthite content ($An/(An+Ab+Or)$) of plagioclase phenocrysts. n shows number of analyzed crystals.

5.4. Hornblende

Fig. 8 shows the variations of aluminum in tetrahedral site of hornblende (Al^{IV}) on the basis of 23 oxygens. Distributions of cations to sites are calculated by the method of Leake et al. (1997). In this method, the distribution of cations cannot be uniquely determined so that the average values of the maximum and minimum estimation of Al^{IV} are shown in Fig. 8. Error is ± 0.05 of Al^{IV} .

Hornblende phenocrysts of the most silicic pumice (4I-1P) occur as subhedral to euhedral crystals with up to 2 mm in length. Compositional range is narrow, varying mainly from 1.5 to 1.7 in Al^{IV} . Zoning is weak.

Hornblende phenocrysts of the mafic scoriae (4I-3S and 4II-3S) occur as anhedral to euhedral crystals with up to 1.5–2 mm in length. Rim compositions (mainly 1.66–1.84) are more restricted than those of cores (1.50–1.90). Zoning where Al^{IV} increases towards rim is dominant. Reverse zoning of up to 2 in Mg# is also common.

Hornblende phenocrysts of the pumices with intermediate compositions (4I-2P, 4I-3P, 4II-1P and 4II-2P) occur as subhedral to euhedral crystals up to 2 mm (4I-2P and 4I-3P) and 3 mm (4II-1P and 4II-2P) in length. Compositional ranges of cores and rims are wide ($Al^{IV}=1.40$ – 1.94), which cover both the compositional ranges of 4I-1P and the scoriae. Zoning is weak, but Al^{IV} contents of some crystals increase from core to rim.

5.5. Opaque oxides

Magnetite phenocrysts are present in all the samples of the Aso-4 cycle, which occur as anhedral to euhedral crystals with up to 0.5–0.8 mm in diameter. Individual crystals of magnetite phenocrysts in all the samples except 4II-2P (welded tuff in the second subcycle) are compositionally homogeneous and range between 0.25 and 0.30 in

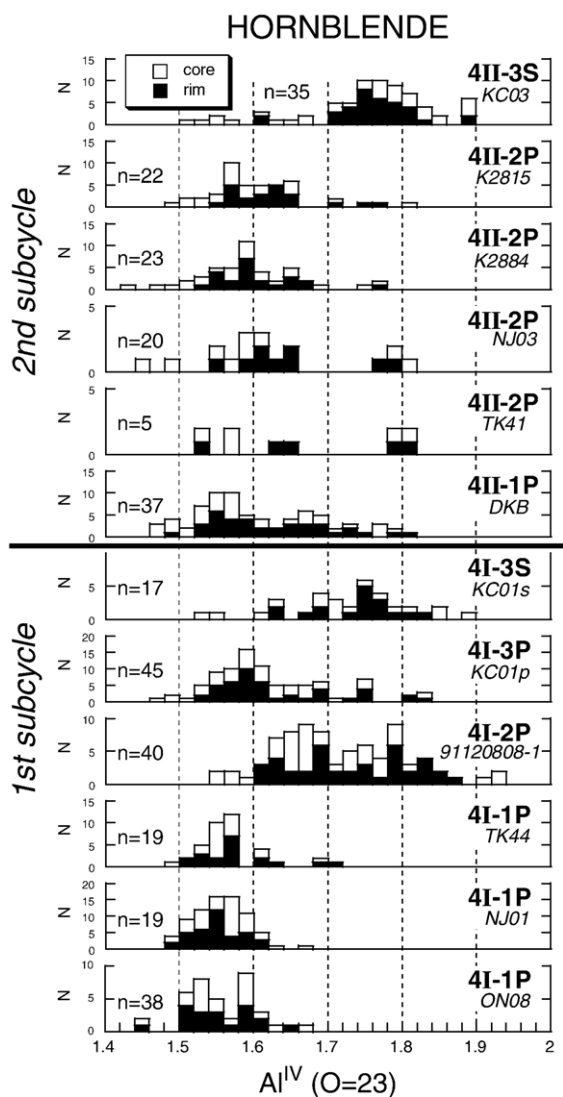


Fig. 8. Frequency diagram showing the tetrahedral-site aluminum (Al^{IV}) content of hornblende phenocrysts on the basis of 23 oxygens. n shows number of analyzed crystals.

ulvospinel content (ulvospinel/(ulvospinel+magnetite) in mole). Magnetite phenocrysts in 4II-2P have exsolution lamellae and their compositions are mainly between 0.25 and 0.3 with an overall range of 0.15 to 0.40 in ulvospinel content. Relationships of Mg and Mn contents of magnetite phenocrysts show distinct features between the most silicic pumice (4I-1P) and the mafic scoriae (4I-3S and 4II-3S); magnetite phenocrysts in the scoriae has higher Mg and low Mn content than those in the most silicic pumice (Fig. 9). The compositional range of magnetite phenocrysts in the pumices with intermediate compositions (4I-2P, 4I-3P, 4II-1P and 4II-2P) covers both the areas of 4I-1P and the scoriae in this diagram.

Fig. 10 shows frequency diagrams of ilmenite contents ($X_{\text{il}} = \text{ilmenite}/(\text{ilmenite} + \text{hematite})$ in mole) of ilmenite phenocrysts. Ilmenite phenocrysts of the most silicic pumice (4I-1P) occur as anhedral to euhedral crystals with up to 0.1 mm in diameter. A narrow and unimodal distribution with peak at $X_{\text{il}} = 0.77\text{--}0.79$ exists. Ilmenite phenocrysts of the mafic scoriae (4I-3S and 4II-3S) occur as anhedral to euhedral crystals with up to 0.3 mm in diameter. A narrow and unimodal distribution with a peak at 0.73–0.74 exists. Ilmenite phenocrysts of the pumices with intermediate compositions have different features between the first and second subcycles; those of the first subcycle (4I-2P and 4I-3P) have the same features as the mafic scoriae, whereas those of the second subcycle (4II-1P and 4II-2P) show bimodal distribution in X_{il} . The histogram peaks of the bimodal distribution are the same as those of the most silicic pumice (4I-1P) and the mafic scoriae (4I-3S and 4II-3S).

5.6. Summary of petrological features of phenocryst

Petrological features of phenocryst minerals of juvenile fragments in the Aso-4 cycle are summarized as follows.

- (1) Phenocryst minerals of the mafic scoriae of the first and second subcycles (4I-3S and 4II-3S) have

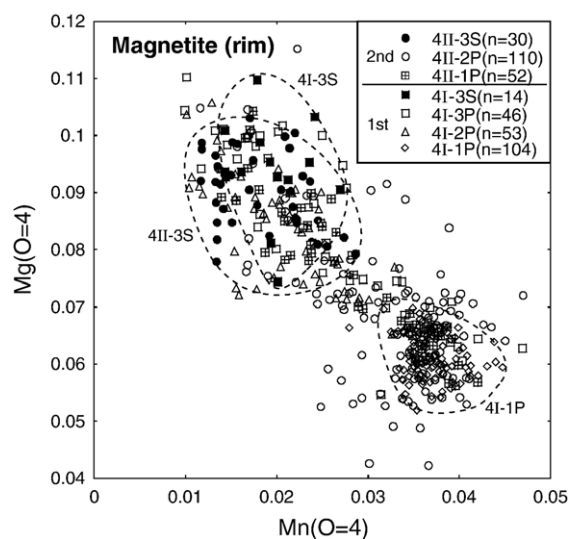


Fig. 9. Mg vs Mn diagram of rim composition of magnetite phenocryst. Mg and Mn contents are calculated on the basis of 4 oxygens. Pots of 4I-1P, 4I-3S, and 4II-3S are limited within areas surrounded by dashed lines, respectively. On the other hands, those of 4I-2P, 4I-3P, 4II-1P, and 4II-2P distribute in both areas of 4I-1P and the scoriae. Number of data is shown (n in legend).

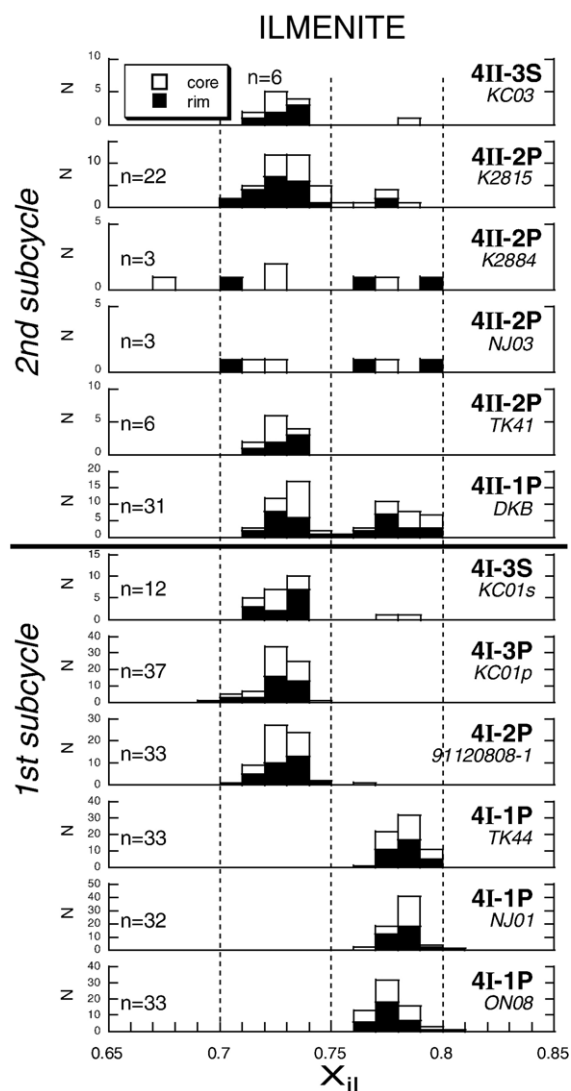


Fig. 10. Frequency diagram showing the ilmenite content (X_{il}) of ilmenite phenocrysts. n shows number of analyzed crystals.

the same petrological features except that the histogram peak of plagioclase at An 45 is observed only in the scoriae of the second subcycle (4II-3S).

- (2) Phenocryst minerals of the mafic scoriae (4I-3S and 4II-3S) have distinct petrological features from those of the most silicic pumice (4I-1P).
- (3) Phenocryst minerals of pumices with intermediate compositions (4I-2P, 4I-3P, 4II-1P and 4II-2P) have both the features of the mafic scoriae (4I-3S and 4II-3S) and the silicic pumice (4I-1P), except for ilmenite phenocrysts. Ilmenite phenocrysts in the second subcycle (4II-1P and 4II-2P) have both the features of ilmenites of the scoriae and 4I-1P,

whereas those in the first subcycles (4I-2P and 4I-3P) are the same as the mafic scoriae.

6. Estimation of pre-eruptive magma conditions

We estimate physico-chemical conditions (temperature, redox state, water content of melt, density and viscosity) of the Aso-4 magmas on the basis of rim compositions of phenocryst minerals and glass compositions of the most silicic pumice (4I-1P), and the mafic scoriae (4I-3S, and 4II-3S). We assume that the glass compositions of these samples represent melt compositions of the Aso-4 magmas. Magmatic pressure is required for estimations of temperature, water content of melt, and density, although there is no petrological or geophysical constraint on the pressure of the Aso-4 magma chamber. We tentatively assume that the pressure of the Aso-4 magma chamber ranges 100–500 MPa in these estimations.

6.1. Temperatures and redox states

Temperatures and redox state of pre-eruptive magmas are estimated from the rim compositions of oxide and pyroxene phenocrysts in the 4I-1P pumice and the scoriae (4I-3S and 4II-3S) using the Fe–Ti-oxide thermometry (Andersen and Lindsley, 1988; Andersen et al., 1993) and the two-pyroxene thermometry (Lindsley, 1983; Andersen et al., 1993).

The rims of magnetite and ilmenite phenocrysts in the 4I-1P pumice and the scoriae have narrow compositional ranges. We applied the Fe–Ti-oxide thermometry to the magnetite–ilmenite pairs that meet the equilibrium criterion based on Mg–Mn partitioning (Bacon and Hirschmann, 1988). Most of magnetite and ilmenite crystals in 4I-1P (more than 20 crystals of both magnetite and ilmenite) and all the ilmenite crystals and at least four magnetite crystals with relatively low Mg/Mn ratio in 4I-3S and 4II-3S meet the equilibrium criterion. Estimated temperatures of 4I-1P, 4I-3S and 4II-3S are 810–850 °C, 830–900 °C, and 830–910 °C, respectively (Fig. 11). Oxygen fugacities of the magmas of 4I-1P, 4I-3S, and 4II-3S are on a buffer curve which is higher by 1.8–2.0 log unit than QFM buffer curve. It is considered that these estimates of the Fe–Ti-oxide thermometry are independent of assumed pressure.

We confirmed that the pairs of clinopyroxene and orthopyroxene used for the two-pyroxene thermometry by Lindsley (1983) are in a state of quasi-equilibrium on Fe–Mg partitioning (Nakamura and Kushiro, 1970). Calculated temperature are 900–960 °C, 940–980 °C, and 940–990 °C for 4I-1P, 4I-3S and 4II-3S,

respectively, for pressures ranging from 100–500 MPa. Estimated temperatures for $P=500$ MPa are at most 15 °C higher than those for $P=100$ MPa.

The above results show that the temperatures estimated by the two-pyroxene thermometry are substantially higher than those by the Fe–Ti-oxide thermometry. Similar discrepancies have been commonly reported and the simplest interpretation of these discrepancies is that pyroxenes have a higher closure temperature than oxides (e.g., Matthews et al., 1999; Schmitt et al., 2001). Judging from the large diffusion coefficients magnetite and ilmenite (Freer and Hauptman, 1978; Pownceby et al., 1987), we consider that the temperatures estimated by the Fe–Ti-oxide thermometry represent the magmatic temperatures just before the eruption.

6.2. Water contents

We estimate water contents of melt of 4I-1P and the scoriae (4I-3S and 4II-3S) using the hygrometer by Housh and Luhr (1991). For this estimation, temperature, pressure, plagioclase composition, and melt composition with the ratio of ferric and ferrous irons are needed. We use the values of temperatures by the Fe–Ti-oxide thermometry. The values of pressures hardly affect the results of this hygrometer in the range of 100 to 500 MPa. Melt compositions are estimated from glass compositions. Ferric and ferrous irons in the melts are evaluated from the equation by Sack et al. (1980). Na_2O content of 4I-1P glass composition (sample NJ01 in Table 3) is lower than the whole-rock compositions of 4I-1P probably because of loss of Na during electron microprobe analyses for glass compositions. Loss of Na in glass composition gives apparently low partition coefficient ($K_D = (\text{Ca}/\text{Na})^{\text{pl}} / (\text{Ca}/\text{Na})^{\text{melt}}$) between plagioclase and melt, which causes an underestimation of water content. In order to evaluate this underestimation of water content, we apply the hygrometer to melt compositions with two different Na contents: $\text{Na}_2\text{O}=4.3$ wt.% (EPMA analysis of glass composition) and 5.0 wt.% (extrapolated Na_2O content from the whole-rock compositional trend in Fig. 2) for 4I-1P. The results show that the effect of loss of Na (i.e., the estimate using $\text{Na}_2\text{O}=4.3$ wt.%) results in only a slight underestimate of water content based on the Ab calibration (at most 0.2 wt.% H_2O).

In the case of the Aso samples, wide compositional variations of rims of plagioclase phenocrysts (more than a few tens wt.% in An content) cause a large uncertainty of water content estimation (see Fig. 7). Accordingly, the compositions of plagioclase are estimated from the

temperatures based on the Fe–Ti-oxide thermometry and the amphibole–plagioclase thermometer by Holland and Blundy (1994). From the amphibole–plagioclase thermometer, we can obtain the relationship between temperature and An content of plagioclase which is in equilibrium with the rims of hornblende phenocrysts (Fig. 12a and c). Compositional ranges of plagioclase that were in equilibrium with the rims of hornblende and two-oxide phenocrysts are estimated from this relationship and the temperatures based on the Fe–Ti-oxide thermometry to be An 30–38, An 35–49, and An 43–57 for 4I-1P, 4I-3S, and 4II-3S, respectively. The estimated compositions of plagioclase are within the ranges of the variation of observed rim compositions, although it does not always coincide with the dominant compositions (see Fig. 12a and c).

The water contents of melts that were in equilibrium with these minerals are estimated from the above compositional ranges of plagioclase, the melt compositions and the temperatures based on the Fe–Ti-oxide thermometry; those are 4.1–5.7 wt.%, 3.8–7.5 wt.%, and 4.1–7.7 wt.% for 4I-1P, 4I-3S, and 4II-3S, respectively (Fig. 12 b and d). The hygrometer by Housh and Luhr (1991) allows for two independent estimations of water content, based on An and Ab exchange, respectively. The two estimations agree fairly well in Fig. 12b and d, which justifies the assumption that the rims of plagioclase, hornblende and two oxides were in equilibrium with the melt. From the minimum estimation of water content (about 4.0 wt.%) and the H_2O solubility equation (e.g., Tamic et al., 2001), it is

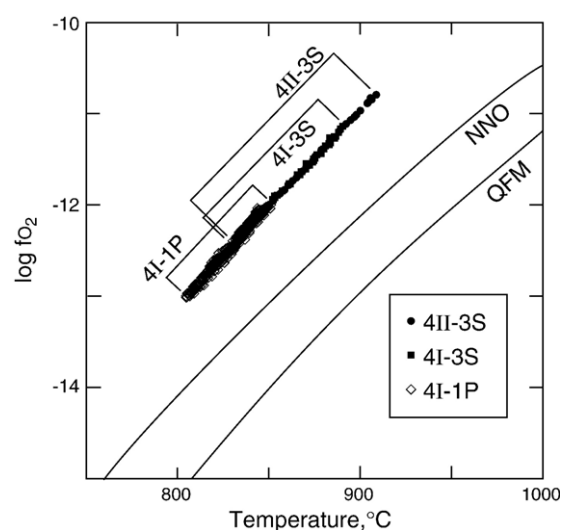


Fig. 11. $\log f\text{O}_2$ vs. temperature diagram estimated by equilibrium between titanomagnetite and ilmenite phenocrysts. Estimations are by the method of Andersen and Lindsley (1988).

inferred that lower bound of magmatic pressure just before the eruption is about 100 MPa.

6.3. Density and viscosity of magmas

Densities and viscosities of the magmas are estimated from melt composition, temperature, pressure, and mode of phenocrysts (Bottinga and Weill, 1972; Shaw, 1972; McBirney, 1984).

Densities of the melts of 4I-1P, 4I-3S, and 4II-3S are estimated at 2110–218, 2060–2200, and 2050–2190 kg m⁻³, respectively for 100 MPa. Estimated melt densities of the scoriae are as small as or even smaller than that of the 4I-1P when the mafic magma contains a large amount of water. On the other hand, bulk densities of the magmas involving phenocryst minerals are estimated at 2180–2240, 2270–2390, and 2260–2380 kg m⁻³ for 4I-1P (10 vol.% phenocryst), 4I-3S (30 vol.% phenocryst), and 4II-3S (30 vol.% phenocryst), respectively, indicating that the magma of 4I-1P is lighter than those of 4I-3S and 4II-3S in bulk density. Estimated densities for 500 MPa are larger than those for 100 MPa by 40–70 kg m⁻³, although the relative magnitude of densities between the magmas does not change depending on assumed pressures.

Viscosities of the melt of 4I-1P, 4I-3S, and 4II-3S are estimated at 4.0–5.2, 2.7–4.7, and 2.4–4.5 log units of viscosity (Pa s), respectively. Bulk viscosities of magma involving crystals can be estimated by

$$\mu_{\text{bulk}} = \mu(1 - 1.35\phi)^{-2.5}$$

where μ_{bulk} , μ , and ϕ are bulk viscosity, melt viscosity, and volume fraction of phenocrysts, respectively (Shaw, 1972). Bulk viscosities of 4I-1P, 4I-3S, and 4II-3S magmas are estimated at 4.2–5.5, 3.2–5.4 and 2.9–5.1 log units of viscosity (Pa s), respectively.

7. Discussion

7.1. Pre-eruptive magma chamber of the Aso-4 cycle

We discuss the pre-eruptive magma chamber on the basis of the petrological features of the juvenile fragments of the first subcycle. The whole-rock compositions of the pumices with intermediate composition (4I-2P and 4I-3P) are plotted on a mixing line between the most silicic pumice (4I-1P) and the silicic end of the mafic scoriae (4I-2S and 4I-3S). The compositional variations of phenocrysts in 4I-2P and 4I-3P are exemplified by those of the most silicic pumice and the mafic scoriae. These features indicate

that the pumices with intermediate compositions originate from a mixture of the most silicic magma and the silicic end of the mafic magmas (this hybrid magma is referred to be as “hybrid magma #1” hereafter). The whole-rock compositions of the intermediate pumices (SiO₂ wt.% = 66–68) are explained by the mixed ratio (the weight fraction of the mafic magma in the mixture) of less than 0.25.

The absence of high- X_{Il} ilmenite phenocrysts in the pumices with intermediate compositions (see 4I-2P and 4I-3P in Fig. 10) may indicate that the high- X_{Il} ilmenite in the silicic magma might dissolve and/or reequilibrate after mixing with the mafic magma. Individual juvenile clasts of the intermediate pumice have homogenous groundmass. These features (reequilibration of phenocrysts and homogeneous groundmass) suggest that this hybrid magma #1 had been generated before the eruptions of the first subcycle. The estimated bulk densities of the magmas (see Section 6.3) and the order of the eruptions in the first subcycle (the silicic, hybrid, and mafic magmas in the stratigraphic order) suggest that the pre-eruptive magma chamber of the Aso-4 cycle was composed of three layers: silicic, hybrid (the hybrid magma #1), and mafic magma layers from the top to the bottom (Fig. 13a).

In contrast with relatively homogenous composition of 4I-1P (within 2 wt.% SiO₂), the mafic scoriae form chemical trends (53–57 SiO₂ wt.%). This fact indicates that the mafic magma layer was compositionally zoned with increasing SiO₂ with height. The hybrid magma #1 is considered to be produced by interfacial mixing of the silicic magma layer and the uppermost part of the mafic magma layer (i.e., the silicic end of the zoned mafic magma layer).

The formation of a compositional gradient in a mafic magma layer underlying a silicic magma layer is supported by experimental results from the viewpoint of physics (e.g., Kaneko and Koyaguchi, 2000). According to the experimental results, when a hot mafic magma is emplaced in a crustal material, compositional convection due to melting and crystallization at a chamber floor, thermal convection due to cooling from above, and precipitation, transportation, and accumulation of crystals simultaneously occur. These complex processes result in a continuous compositional gradient and a wide compositional variation of crystals. Such experimental results would explain the continuous chemical trend in whole rock compositions and the wide compositional variation of plagioclase phenocrysts of the mafic scoriae.

The most silicic pumice (4I-1P) has plagioclase phenocrysts showing a bimodal distribution of An

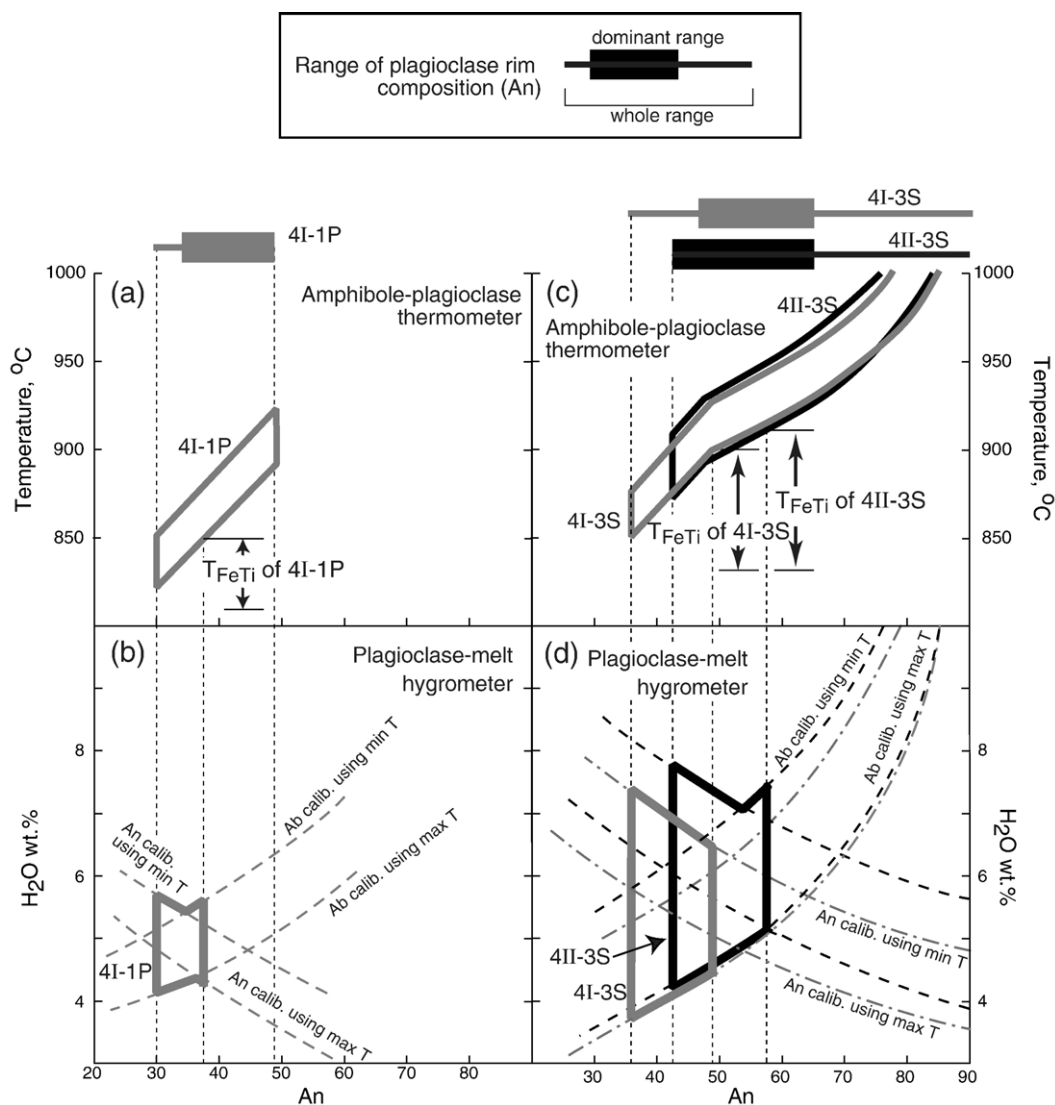


Fig. 12. (a) Temperature vs An content of plagioclase for 4I-1P. Temperature is estimated by the amphibole–plagioclase thermometer by Holland and Blundy (1994). Calculation is carried out on all the pairs of rim compositions of plagioclase and hornblende phenocrysts analyzed by electron microprobe. Area surrounded by grey bold lines represents calculation result. Dominant compositional range (solid box) and whole compositional range (solid line) of plagioclase rim in frequency diagram (Fig. 7) are shown. Temperature range estimated by the Fe–Ti–oxide thermometry by Andersen and Lindsley (1988) is shown in diagram (T_{FeTi}). Assuming equilibrium between magnetite, ilmenite, hornblende, and plagioclase, the compositional range of plagioclase in equilibrium with the other minerals is obtained. The maximum and minimum An contents of plagioclase in equilibrium with the other minerals are determined by the maximum temperature estimated by the Fe–Ti–oxide thermometer and plagioclase with rim composition of the minimum An content in 4I-1P, respectively. (b) Water content of melt (wt.%) vs An content of plagioclase. Water content is calculated by the plagioclase–melt hygrometer by Housh and Luhr (1991). The hygrometer by Housh and Luhr (1991) allows us for two independent estimations of water content, based on An and Ab exchange, respectively. Calculation results by the two independent estimation using the maximum and minimum temperatures estimated by the Fe–Ti–oxide thermometry are shown (grey dashed line). Water estimations using the maximum and minimum temperatures give the lower and upper bounds of water content for plagioclase with a certain composition, respectively. From area restricted by this estimated range of water content and the compositional range of plagioclase (area surrounded by grey bold line), range of water content of melt is estimated at 4.1–5.7 wt.% H₂O for 4I-1P. (c) and (d) are diagrams for water estimation of 4I-3S (grey) and 4II-3S (black).

contents in the range of An 30–50 (see Fig. 7). At present the origin of such disequilibrium features in plagioclase phenocrysts remains unsolved. Neverthe-

less, the fact that all the pumice samples (4I-1P, 4I-2P, 4I-3P, 4II-1P and 4II-2P) contain plagioclase phenocrysts showing the same bimodal distribution in the

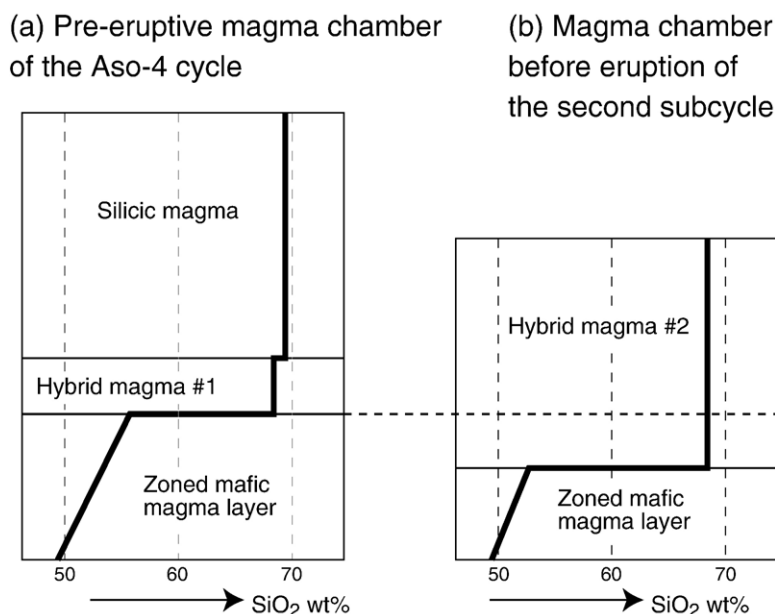


Fig. 13. Schematic sketch of compositional structure of the Aso-4 magma chamber (not to length scale). Schematic vertical profile of whole-rock composition on SiO_2 is presented (bold line). (a) Pre-eruptive magma chamber. (b) Magma chamber before the eruption of the second subcycle.

range of An 30–50 indicates that this petrological feature of 4I-1P characterizes that of the silicic end-member magma.

7.2. Magmas of the second subcycle

The juvenile fragments of the second subcycle are classified into two groups: the pumices with intermediate composition (4II-1P and 4II-2P) and the mafic scoriae (4II-3S). The compositional variations of phenocrysts 4II-1P and 4II-2P are exemplified by those of the most silicic magma (4I-1P) and the mafic scoriae (4I-3S and 4II-3S). The whole-rock composition of 4II-2P is plotted on a mixing line between the most silicic pumice of the first subcycle (4I-1P) and the silicic end of the mafic scoriae of the first subcycle (4I-3S). These features suggest that 4II-1P and 4II-2P also originate from a mixture of the most silicic magma and the silicic end of the mafic magmas (this hybrid magma is referred to be as “hybrid magma #2” hereafter). Although the whole-rock composition of 4II-1P was not obtained due to intensive weathering, it is inferred from the petrological features of the phenocrysts that the origin of 4II-1P is same as 4II-2P. Homogeneous groundmasses of 4II-2P suggest that this hybrid magma #2 was generated before the eruptions of the second subcycle began. Although the hybrid magma #2

has the same end-member magmas as the hybrid magma #1, it has a different compositional variation of ilmenite phenocrysts from that of the hybrid magma #1; high X_{Il} ilmenite phenocrysts that characterize the silicic end-member magma are present only in the hybrid magma #2 (see Fig. 10). This fact may indicate that the two hybrid magmas were produced at different stages.

One possible mechanism to generate the hybrid magma #2 is magma flow accompanied by the eruption of the first subcycle. When a layered magma chamber that has a silicic magma and underlying mafic magma is tapped from its top, the mafic magma can be evacuated into the conduit by viscous drag of silicic magma before all the silicic magma is evacuated (Koyaguchi, 1985; Blake and Campbell, 1986; Freundt and Tait, 1986). During the first subcycle of the Aso-4 cycle, it is reasonable to consider that evacuation of the hybrid magma #1 and the upper part of the mafic magma layer occurred without complete evacuation of the overlying silicic magma. The magma flow during the first subcycle disrupted the interfaces between the magma layers and the hybrid magma #2 was generated by mixing the silicic magma, the hybrid magma #1, and the uppermost part of the mafic magma. Before the second subcycle, the magma chamber had two layers: the layer of the hybrid magma #2 and the underlying zoned mafic magma layer (Fig. 13b).

The mafic scoriae of the second subcycle (4II-3S) have a whole-rock compositional variation of SiO_2 wt.% = 50–54 and form a continuous trend with the mafic scoriae of the first subcycle (see Fig. 2). They have the same compositional features of phenocrysts as the mafic scoriae of the first subcycle except the histogram peak of plagioclase around An 45 (see Fig. 7). These features suggest that the mafic scoriae of the second subcycle originate from the deeper part of the zoned mafic magma layer in the pre-eruptive magma chamber (Fig. 13a).

The different compositional variations of the mafic scoriae between the first and second subcycles may be explained by an alternative model that replenishment of a new mafic magma occurred after the first subcycle and the mafic magma of the second subcycle was the replenished magma. We can recognize that the mafic scoriae of the two subcycles show slightly different trends in the diagram of CaO vs SiO_2 (Fig. 2) and that the mafic scoriae of the second subcycle have higher Y contents than those of the first subcycle (Fig. 3); however, these differences are not so obvious as to eliminate the model that those scoriae were derived from a single zoned magma layer. For example, the high Y contents of the mafic scoriae in the second subcycle are explained by accumulation of apatite, in which Y is strongly compatible. Although we cannot eliminate the alternative model, considering the continuous chemical trends in most of the whole rock variation diagrams and the resemblance of the petrological features (e.g., compositional variations of phenocrysts) between the two subcycles, the model of the single zoned mafic magma layer would be the simplest interpretation.

7.3. Proposed scenario of the Aso-4 cycle

In the above model we have proposed that evacuation of the hybrid magma #1 and the mafic magma occurred without complete evacuation of the overlying silicic magma. In order to discuss conditions under which these processes occur, we should know dimension of the magma chamber. At first, we discuss roof area of the magma chamber. Ono et al. (1993) has reported an area (20 km N–S and 10 km E–W in diameter) with negative Bouguer anomaly (–10 mgal) surrounded by steep gravity gradient inside the caldera, and has proposed that the formation of the Aso caldera occurred by a piston-like collapse of the basement with 500 m subsidence like Long Valley Caldera and Crater Lake (Hildreth and Mahood, 1986; Suzuki-Kamata et al., 1993). Moreover, data of deep drill holes inside the Aso caldera indicate about 500 m subsidence of the basement rocks near the

steep gravity gradient (Hoshizumi et al., 1997), which is consistent with the idea of a piston-like collapse. The caldera was formed by all the four large pyroclastic-eruption cycles. On the other hand, the Aso-4 cycle ($>600 \text{ km}^3$ in eruption volume) is much larger than the total volume of the previous three large pyroclastic-eruption cycles ($>250 \text{ km}^3$ in eruption volume) and dominantly contributed to the formation of the caldera. Here, we assume that the structure beneath Aso volcano proposed by Ono et al. (1993) was formed by the Aso-4 cycle and that the area with the negative gravity anomaly is identical to the roof area of the magma chamber in the Aso-4 cycle, which is estimated at 200 km^2 .

The volume of the hybrid magma #2 of the second subcycle is more than 10^1 km^3 (Table 1) and its layer thickness is estimated at more than 10^2 m deep. Therefore, the silicic magma with the same order of thickness must remain for the single chamber model when the mafic magma was evacuated in the first subcycle. The equations by Blake and Ivey (1986) and the estimated bulk densities and viscosities of the silicic and mafic magmas indicate that magma discharge rate of more than $10^5 \text{ m}^3 \text{ s}^{-1}$ can evacuate the mafic magma leaving the overlying silicic magma with 10^2 m deep. This discharge rate is occasionally estimated as that of large ignimbrite eruption (Cas and Wright, 1987), and it can be stated that the single chamber model is appropriate. In the following, we propose a scenario of the eruptions in the Aso-4 cycle on the basis of the above discussion and geological information.

Before the eruptions, there was a three-layer magma chamber as mentioned above (Fig. 14a). The total volume of the Aso-4 is estimated at more than 600 km^3 , most of which from the 4I-IP (the silicic magma of the first subcycle; $>60 \text{ km}^3$) and co-ignimbrite ash ($>400 \text{ km}^3$) (Table 1). Glass of co-ignimbrite ash has the same composition as that of 4I-IP which is the silicic magma, suggesting that magmas of co-ignimbrite ash basically are identical to the silicic magma. Representative apparent density of 4I-IP is 500 kgm^{-3} (Watanabe, 1978). An apparent density of co-ignimbrite ash for estimation of the volume is more than 1000 kgm^{-3} because its deposits are compacted by deposits after the Aso-4 ash (Machida and Arai, 2003). Therefore, the dense rock equivalent (DRE) volume of the silicic magma exceeds 200 km^3 and the pre-eruptive magma chamber with the roof area of 200 km^2 requires that the silicic magma layer was more than 10^3 m deep. The rock from the hybrid magma #1 of the first subcycle ($>4 \text{ km}^3$ in volume of deposits) has typical apparent density of 500 kgm^{-3} (Watanabe, 1978) so that its DRE volume is about 1 km^3 and its thickness in the chamber is estimated at more than 10^1 m deep.

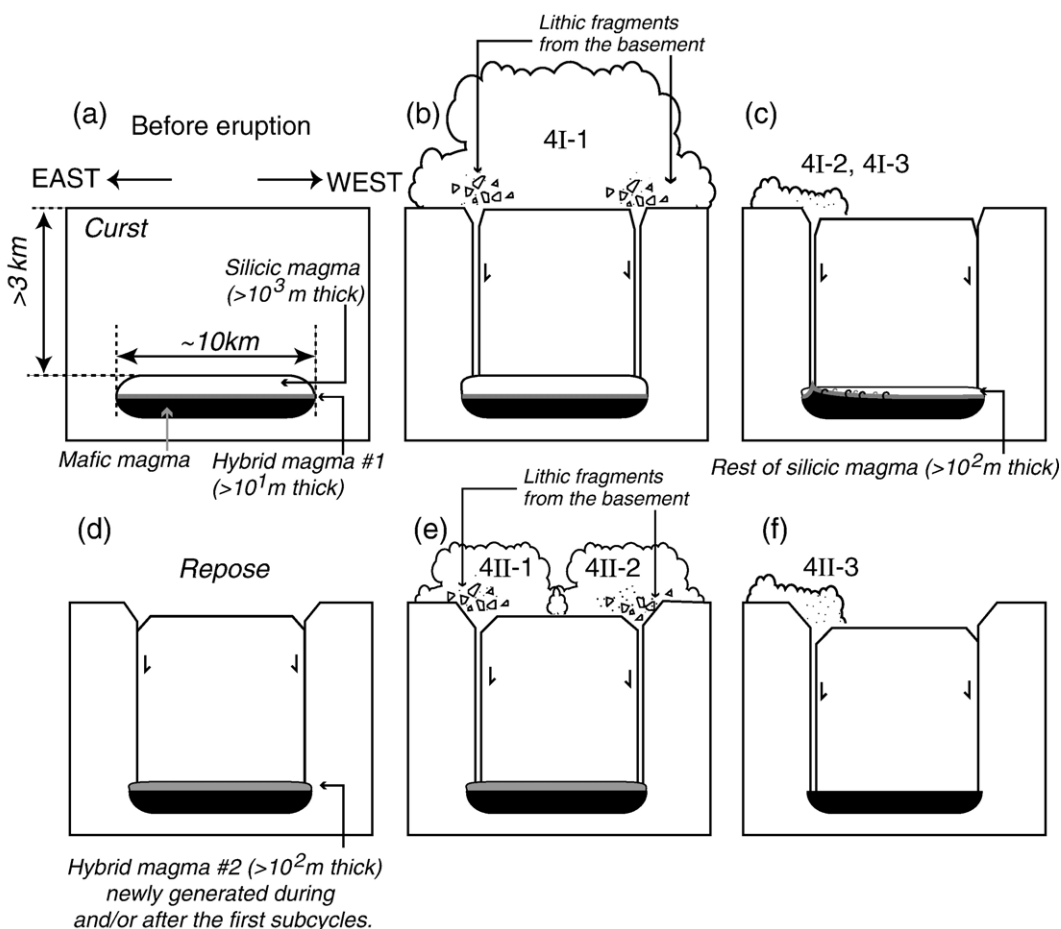


Fig. 14. Schematic illustration showing a proposed model of the pre-eruptive magma chamber and processes during the eruptions in the Aso-4 cycle. Depth of the magma chamber is estimated from the minimum H₂O saturation pressure on 4I-1P.

The eruption in the first subcycle began with the silicic magma (4I-1) at the upper part of the layered magma chamber. The pyroclastic flow of 4I-1 is distributed all around the caldera and was the most voluminous and violent pyroclastic-flow eruption in the Aso-4 cycle, which is larger by more than one order than the other eruptions of the Aso-4 cycle in volume. Most of the co-ignimbrite ash deposits originated from this eruption. This eruption ruptured the cap rocks around a fissure, expanded the fissure, and then created a ring fissure vent, which caused a piston-like collapse (Fig. 14b). The fact that lithologies of lithic fragments are different in areas outside the caldera (Ono et al., 1977; Watanabe, 1978; Kamata, 1997) may support fissure eruption spreading spatially rather than central eruption. During the course of the eruption, the silicic magma layer became thinner. The deposits of the subsequent eruptions (4I-2 and 4I-3) are distributed only at the western area

outside the caldera. Their eruption occurred from only the western part of the ring fissure vent although the 4I-1P eruption occurred from the whole of the ring fissure. The evacuation of the silicic magma at the western vent lifted up the hybrid magma #1 (4I-2P and 4I-3P) and the upper part of the mafic magma layer (4I-2S and 4I-3S), which were drained into the fissure before all the silicic magma was evacuated (Fig. 14c). The eruption stopped before the magmas were exhausted.

During the first subcycle, the silicic magma mixed with the hybrid magma #1 and the uppermost part of the mafic magma layer, resulting in generation of the hybrid magma #2 erupting in the second subcycle (4II-1 and 4II-2) (Fig. 14d). The hybrid magma #2 was formed by magma interface disruption and mingling between the magma layers during the first subcycle eruption (Fig. 14c). After a short repose, the second subcycle began with tapping of the hybrid magma #2 at the top of

the magma chamber again. First, pyroclastic-flow eruption of 4II-1 the deposits of which are distributed at western area outside the caldera occurred from the western (fissure ?) vent occurred (Fig. 14e). The facts that 4II-1 contains large amount of lithic fragments and that glasses are much altered may suggest that this eruption was very explosive due to interaction of ground water and magma like phreatomagmatic eruption. Subsequently, the eastern vent opened and pyroclastic-flow eruption of 4II-2. Extracaldera deposits of this subcycle are deposited to the eastern area of the caldera. Finally, the eastern vent closed and the mafic magma (4II-3) the deposits of which are distributed at western area outside the caldera erupted from the western vent (Fig. 14f). The fact that the spatial distributions of deposits of the first and second subcycles overlap supports this single chamber model (see Fig. 1).

8. Concluding remarks

On the basis of petrological and geological data, we have reconstructed the pre-eruptive magma chamber and the eruption processes of the Aso-4 cycle. The complex petrological features and the occurrence of the pyroclastic deposits of the Aso-4 cycle are explained by a simple model of twice tappings from a single magma chamber. The example of the Aso-4 cycle provides us important information about phenomena accompanied by large scale volcanisms, such as repeated large-scale eruptions with short time interval and a reconfiguration of a magma chamber due to incomplete evacuation during one major eruption. We believe that the accumulation of the knowledge on such examples would improve our understandings of large scale volcanisms.

Acknowledgement

We acknowledge the years of contributions of Prof. K. Watanabe and late Dr. K. Ono's work to understanding of the geology of Aso volcano and also thank them for their kind instructions of everything about Aso to us. We thank Dr. Nakashima for his instruction of isotope analysis and staffs in Earthquake Research Institute, The University of Tokyo and Kobe University for allowing us to use EPMA. Some of the data in this study was obtained by Mr. H. Matsuura and Mr. H. Takaoka during their master and/or bachelor theses projects at Ehime University and Kumamoto University being supervised by T.K. We thank them for their cooperation and discussion in the field. We thank Dr. A. Schmitt and an anonymous referee for constructive comments. This work is partially supported by a grant-

in-aid for scientific research from Japan Society of the Promotion of Science.

References

- Andersen, D.J., Lindsley, D.H., 1988. Internally consistent solution models for Fe–Mg–Mn–Ti oxides: Fe–Ti oxides. *American Mineralogist* 73, 714–726.
- Andersen, D.J., Lindsley, D.H., Davidson, P.M., 1993. QUILF: a Pascal program to access equilibria among Fe–Mg–Mn–Ti oxides, pyroxenes, olivine, and quartz. *Computer Geoscience* 19, 1333–1350.
- Bacon, C.R., Hirschmann, M.M., 1988. Mg/Mn partitioning as a test for equilibrium between coexisting Fe–Ti oxides. *American Mineralogist* 73, 57–61.
- Blake, S., Campbell, I.H., 1986. The dynamics of magma mixing during flow in volcanic conduits. *Contributions to Mineralogy and Petrology* 94, 72–81.
- Blake, S., Ivey, G.N., 1986. Magma mixing and the dynamics of withdrawal from stratified reservoirs. *Journal of Volcanology and Geothermal Research* 27, 153–178.
- Bottinga, Y., Weill, D.F., 1972. The density of liquid silicate systems calculated from partialmolar volumes of oxide components. *American Journal of Science* 269, 169–182.
- Cas, R.A.F., Wright, J.V., 1987. *Volcanic successions*. Unwin Hyman Ltd, London.
- Committee of catalog of Quaternary volcano in Japan, 1999. *Catalog of Quaternary volcanoes in Japan*. Ver. 1.0. The volcanological society of Japan, CD-ROM.
- Fisher, R.V., Schmincke, H.U., 1984. *Pyroclastic Rocks*. Springer-Verlag, Berlin Heidelberg, p. 472.
- Freer, R., Hauptman, Z., 1978. An experimental study of magnetite–titanomagnetite interdiffusion. *Physics of the Earth and Planetary Interiors* 16, 223–231.
- Freundt, A., Tait, S., 1986. The entrainment of high-viscosity magma into low-viscosity magma in 31 eruption conduits. *Bulletin of Volcanology* 48, 325–340.
- Furukawa, K., 2006. *Geology and petrology of silicic volcanism in extensional plate convergent zone*. PhD Thesis, Kyoto University, Japan.
- Hildreth, W., 1981. Gradients in silicic magma chambers: Implications of lithospheric magma in eruption conduits. *Bulletin of Volcanology* 48, 325–340.
- Hildreth, W., Mahood, G.A., 1986. Ring-fracture eruption of the Bishop Tuff. *Geological Society of America Bulletin* 97, 396–403.
- Holland, J.D., Blundy, T.J.B., 1994. Calcic amphibole equilibria and a new amphibole–plagioclase geothermometer. *Contributions of Mineralogy and Petrology* 104, 208–224.
- Hoshizumi, H., Watanabe, K., Sakaguchi, K., Uto, K., Ono, K., Nakamura, T., 1997. The Aso-4 pyroclastic flow deposit confirmed from the deep drill holes inside the Aso caldera. *Programme and Abstract: Volcanological Society of Japan*, 1997, vol. 2, p. 5 (in Japanese).
- Housh, T.B., Luhr, J.F., 1991. Plagioclase-melt equilibria in hydrous systems. *American Mineralogist* 76, 477–492.
- Hunter, A.G., 1998. Intracrustal controls on the coexistence of tholeiitic and calc-alkaline magma series at Aso Volcano, SW Japan. *Journal of Petrology* 39, 1255–1284.
- Kamata, H., 1997. *Geology of the MiyanoHaru district*. With Geological Sheet Map at 1:50000, Geological Survey of Japan, 127p. (in Japanese with English abstract 5p.).

- Kaneko, K., Koyaguchi, T., 2000. Simultaneous crystallization and melting at both the roof and floor of crustal magma chambers: Experimental study using $\text{NH}_4\text{Cl-H}_2\text{O}$ binary eutectic system. *Journal of Volcanology and Geothermal Research* 96, 161–174.
- Koyaguchi, T., 1985. Magma mixing in a conduit. *Journal of Volcanology and Geothermal Research* 25, 365–369.
- Leake, B.E., Woolley, A.R., Arps, C.E.S., Birch, W.D., Gilbert, M.C., Grice, J.D., Hawthorne, F.C., Kato, A., Kisch, H.J., Krivovichev, V.G., Linthout, K., Laird, J., Mandarino, J.A., Maresch, W.V., Nickel, E.H., Rock, N.M.S., Schumacher, J.C., Smith, D.C., Stephenson, N.C.N., Ungaretti, L., Whittaker, E.J.W., Youzhi, G., 1997. Nomenclature of amphiboles: report of the subcommittee on amphiboles of the international mineralogical association, commission on new minerals and mineral names. *American Mineralogist* 82, 1019–1037.
- Lindsley, D.H., 1983. Pyroxene thermometry. *American Mineralogist* 68, 477–493.
- Lipman, P.W., 1967. Mineral and chemical variations within an ash-flow sheet from Aso caldera, south-western Japan. *Contributions to Mineralogy and Petrology* 16, 300–327.
- Machida, H., Arai, F., 2003. Atlas of tephra in and around Japan. University of Tokyo Press, p. 336 (in Japanese).
- Machida, H., Arai, F., Momose, M., 1985. Aso-4 ash: a widespread tephra and its implications to the events of Late Pleistocene in and around Japan. *Bulletin of Volcanological Society of Japan* 30, 49–70 (in Japanese with English abstract).
- Matsumoto, T., 1943. The four gigantic caldera volcanoes in Kyushu. *Japanese journal of geology and geography* 19, 1–57 (Special number).
- Matsumoto, A., Uto, K., Ono, K., Watanabe, K., 1991. K–Ar age determinations for Aso volcanic rocks-concordance with volcanostratigraphy and application to pyroclastic flows. *Programme and Abstract: Volcanological Society of Japan*, vol. 2, p. 73.
- Mathews, S.J., Sparks, R.S.J., Gardeweg, M.C., 1999. The Piedras Grandes-Soncor eruptions, Lascar Volcano, Chile; Evolution of a zoned magma chamber in the Central Andean upper crust. *Journal of Petrology* 40, 1891–1919.
- McBirney, A.R., 1984. *Igneous Petrology*. Freeman, Cooper and Company, San Francisco.
- Nakamura, Y., Kushiro, I., 1970. Equilibrium relations of hypersthene, pigeonite and augite in crystallizing magmas: microprobe study of a pigeonite andesite from Weiselberg, Germany. *American Mineralogist* 55, 1999–2015.
- Ono, K., Watanabe, K., 1983. The Aso caldera. *The Earth Monthly* 5, 73–82 (in Japanese).
- Ono, K., Watanabe, K., 1985. Geological map of Aso volcano, 1:50000. Tokyo: Geological Survey of Japan.
- Ono, K., Matsumoto, Y., Miyahisa, M., Teraoka, Y., Kambe, N., 1977. Geology of the Taketa district. With Geological Sheet Map at 1:50000, Geological Survey of Japan, p157. (in Japanese with English abstract 8p.).
- Ono, K., Watanabe, K., Komazawa, M., 1993. Structure of the Aso caldera implied by gravitational data. *The Earth Monthly* 15, 686–690 (in Japanese).
- Pownceby, M.I., Wall, V.J., O'Neill, H.St.C., 1987. Fe–Mn partitioning between garnet and ilmenite: Experimental calibration and applications. *Contributions to Mineralogy and Petrology* 97, 116–126.
- Ritchey, J.L., 1980. Divergent magmas at crater lake, Oregon: products fractional crystallization and vertical zoning in a shallow, water-undersaturated chamber. *Journal of Volcanology and Geothermal Research* 7, 373–386.
- Sack, R.C., Carmichael, I.S.E., Rivers, M., Ghiorsio, M.S., 1980. Ferric–Ferrous equilibria in natural silicate liquids at 1 bar. *Contributions to Mineralogy and Petrology* 75, 367–376.
- Sano, T., 2002. Determination of major and trace element contents in igneous rocks by X-ray fluorescence spectrometer analysis. *Bulletin of Fuji Tokoha University* 2, 43–59.
- Schmitt, A.K., de Silva, S.L., Trumbull, R.B., Emmermann, R., 2001. Magma evolution in the Purico ignimbrite complex, northern Chile: evidence for zoning of a dacitic magma by injection of rhyolitic melts following mafic recharge. *Contributions Mineralogy and Petrology* 140, 680–700.
- Shaw, H.R., 1972. Viscosities of magmatic silicate liquids: an empirical method of prediction. *American Journal of Science* 272, 870–893.
- Shibata, T., Yoshikawa, M., Tatsumi, Y., 2003. An analytical method for determining precise Sr and Nd isotopic compositions and results for thirteen rock standard materials. *IFREE Report for 2001–2002*, pp. 363–367.
- Suzuki-Kamata, K., Kamata, H., 1990. The proximal facies of the Tosu pyroclastic-flow deposit erupted from Aso caldera, Japan. *Bulletin of Volcanology* 50, 325–333.
- Suzuki-Kamata, K., Kamata, H., Bacon, C.R., 1993. Evolution of the caldera-forming eruption at Crater Lake, Oregon, indicated by component analysis of lithic fragments. *Journal of geophysical Research* 98, 14059–14074.
- Takahashi, T., Yoshikawa, M., 2005. Precise isotope determination of trace amounts of Sr aims at the insitu Sr isotope analysis by microdrilling technique. *Annual report FY2003, Institute for Geothermal Sciences*, pp. 46–48.
- Tamic, N., Behrens, H., Holtz, F., 2001. The solubility of H_2O and CO_2 in rhyolitic melts in equilibrium with a mixed $\text{CO}_2\text{–H}_2\text{O}$ fluid phase. *Chemical Geology* 174, 333–347.
- Watanabe, K., 1978. Studies on the Aso Pyroclastic Flow Deposits in the region to the west of Aso caldera, southwest Japan, I: Geology. *Memorial of the Faculty of Education, Kumamoto University. Natural Sciences*, vol. 27, pp. 97–120.
- Watanabe, K., 1979. Studies on the Aso Pyroclastic Flow Deposits in the region to the west of Aso caldera, southwest Japan, II: petrology of the Aso-4 pyroclastic flow deposits. *Memorial of the Faculty of Education, Kumamoto University. Natural Sciences*, vol. 27, pp. 97–120.
- Watanabe, K., 2001. History and activity of Aso volcano (Japanese title “Aso kazan no oitachi”). Ichinomiya-cho, Japan. (in Japanese).