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GEOCHEMICAL FEATURES OF FLUID INCLUSIONS FROM GEOTHERMAL SYSTEMS AND GRANITIC BODIES IN JAPAN

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ABSTRACT

We have studied fluid inclusions in minerals from geothermal systems and granitic bodies in Japan, especially their gas compositions. Five types of fluid inclusions occur in the studied samples: (1) monophase liquid, (2) aqueous two-phase liquid-rich, (3) aqueous two-phase vapor-rich, (4) liquid CO_2-bearing aqueous and (5) halite and/or other daughter mineral-bearing polyphase inclusions. In general, fluid inclusions of low salinity and gas content (types 1, 2 and 3) occur in active geothermal systems in Japan. Coexistence of fluid inclusions of types 2 and 3 (boiling of geothermal fluids) sometimes occur in very active geothermal systems. Highly saline fluid inclusions (type 5) occur in the magma-related geothermal system (Kakkonda), and liquid CO_2-bearing inclusions (type 4) occur in the geothermal reservoir is in limestone (Mori). As for granitic rocks in Japan, various granitic rocks mainly of Cretaceous-Paleogene to Neogene with small numbers of Paleozoic, Triassic-Jurassic and Quaternary are exposed. Probably, fluid inclusions of types 2, 3, 4 and 5 are trapped fluids under cooling processes of the crystallized granitic magmas. Aqueous two-phase liquid-rich inclusions (type 2) are the most ubiquitous among the types. Daughter mineral-bearing polyphase (type 5) and vapor-rich inclusions (type 3) are more frequently in Neogene and Quaternary granitic rocks than in the Cretaceous ones, or are also common in granitic rocks intruding contemporaneous volcanic rocks. The Neogene granitic rocks are thought to be eroded less than the Cretaceous ones, and their exposed levels are shallow. That is, the shallower plutons have the more saline fluids in inclusions. Experimental data on the NaCl-H_2O system and solidus of granitic rocks also shows that highly saline fluid in which halite is stable at room temperature are trapped in inclusions at shallower depths. Gas analyses by a quadrupole mass spectrometer show relatively high gas contents of the inclusions in granitic rocks, compared with the geothermal systems in Japan. X_{CO_2}/X_{CH_4} ratios of the fluid inclusions in the granitic rocks have a wider range than that of X_{CO_2}/X_{N_2} ratios, and the former ratios may reflect the difference of fO_2 around the granitic rocks where the fluids flew. CO_2-CH_4-N_2 ternary plots of the fluid inclusions show a different trend from those of the hydrothermal systems. X_{CO_2}/X_{CH_4} ratios of fluid inclusions from S-type granites intruded the Paleogene-Miocene sediments are quite low. These results suggest that gas compositions of inclusion fluids are probably controlled by fluid-rock interaction in the granitic rocks and/or in sedimentary rocks around the granitic rocks, or, possibly, by source of granitic magma.

Keywords: fluid inclusions, gas analysis, active geothermal system, granitic rock, magnetite-series, ilmenite-series, I-type, S-type, Japan.

1. INTRODUCTION

Portions of crustal fluids are trapped as fluid inclusions in minerals through precipitation at hydrothermal activities and crystallization of magmas. The chemical compositions of the fluids may be controlled by boiling, dilution and fluid-rock interactions before trapping, but it is difficult to observe the processes directly. However, studies of fluid inclusions may give us clues to reveal the processes. For the purpose, we have studied various fluid inclusions in hydrothermal and igneous minerals from geothermal systems and granitic bodies in Japan, especially gas compositions of the inclusions. In this presentation, we will summarize the geochemical features of crustal fluids at magmatic and hydrothermal stages base on the fluid inclusion studies.

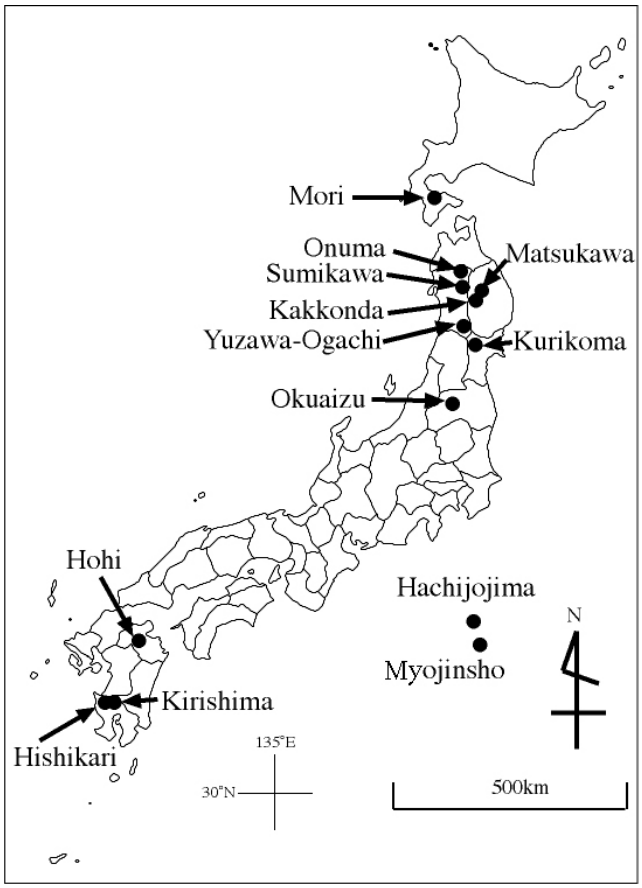

Figure 1. Studied geothermal areas where fluid inclusion samples were collected.

2. FLUID INCLUSIONS OF GEOTHERMAL SYSTEMS AND GRANITIC BODIES IN JAPAN

2.1 Geothermal Systems

There are many geothermal areas in Japan, and some of them with high energy potential are used for geothermal power generation. We have analyzed fluid inclusions in hydrothermal minerals collected from such geothermal areas, in order to reveal geochemical features of geothermal fluids and thermal structures and histories. Figure 1 shows the studied geothermal areas where analyzed fluid inclusions were collected, and analytical results are published and presented in various journals and meetings. The studied areas are Mori (Muramatsu *et al.*, 1997), Sengan including Onuma, Sumikawa and Matsukawa (Sasaki *et al.*, 1992; Muramatsu *et al.*, 2001), Kakkonda (Sawaki *et al.*, 1999; Muramatsu *et al.*, 2000), Yuzawa-Ogachi (Sawaki and Sasada, 2003), Kurikoma (Sawaki *et al.*, 1998), Okuaizu (Sasada *et al.*, 1992), Hachijojima (Sasaki *et al.*, 1995b), Hohi (Sasada *et al.*, 1992), Kirishima (Sawaki *et al.*, 1997), Hishikari (Ueno *et al.*, 2001), Myojinsho (Sasaki *et al.*, 1995a) and Broadland in New Zealand (Sasada *et al.*, 1992). Hishikari is famous as an epithermal type gold deposit, and is an ancient geothermal system. Myojinsho is a submarine caldera, and hydrothermal minerals are formed.

Five types of fluid inclusions occur in the studied samples from geothermal systems and granitic rocks: (1) monophase liquid, (2) aqueous two-phase liquid-rich, (3) aqueous two-phase vapor-rich, (4) liquid CO_2-bearing aqueous and (5) halite and/or other daughter mineral-bearing polyphase inclusions. In general, fluid inclusions of low salinity and gas content (types 1, 2 and 3) occur in active geothermal systems in Japan. Coexistence of fluid inclusions of types 2 and 3 sometimes occur in very active geothermal systems, and it suggests boiling of geothermal fluids. Highly saline fluid inclusions (type 5) that coexist with type 3 occur at the deep geothermal reservoir in the Kakkonda geothermal area, where a very young Quaternary granitic rock is intruded and plays a role of a heat source. Liquid CO_2-bearing inclusions (type 4) occur in the Mori geothermal area, where the geothermal reservoir is in limestone.

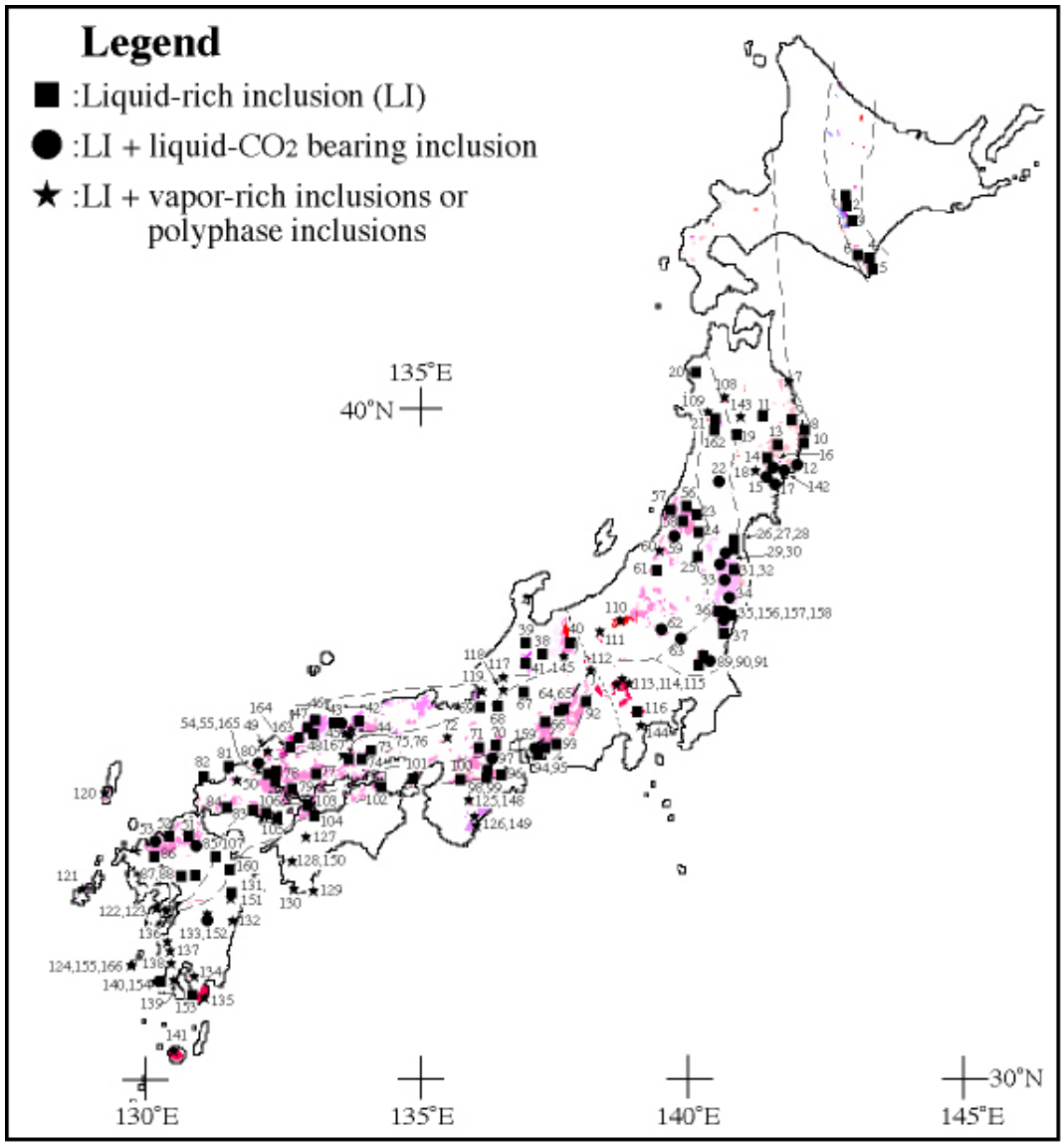

Figure 2. Granitic rocks in Japan and localities of the analyzed samples with occurrence of fluid inclusions. The numbers corresponds to those in Table 1. LI: liquid-rich two phase inclusions.

2.2 Granitic Rocks in Japan

Figure 2 shows distribution of granitic rocks in Japan and sampling points. A large amount of I-type granitic rocks of Cretaceous to Tertiary (Paleogene) ages are exposed in Japan. Magnetite-series granitic rocks occur mainly in the Kitakami (NE Japan) and Sanin (SW Japan) areas. In the Abukuma (NE Japan) and Ryoke (SW Japan) belts, ilmenite-series granitic rocks are accompanied by metamorphic rocks (mainly gneiss). In the Sanyo area (SW Japan), ilmenite-series granitic rocks occur as batholiths, as well as in Sanin. S- and I-types and ilmenite-series granitic rocks of Tertiary (Neogene) mainly occur in the Outer zone of southwestern Japan and the Hidaka belt in northeastern Japan. The S-type granitic rocks are thought to be originated in partial melt of the lower crustal materials (sediments). In the "Green Tuff" region in the western and northern sides of the Japanese Islands and in the Fossa Magna area, I-type and magnetite-series plutons are sporadically exposed. Pre-Silurian granite (Hikami) is in the Kitakami area without relation to the Cretaceous ones. Jurassic granite (Funatsu) is in the Hida belt. Quaternary ones (Kakkonda, Takidani and Yaguradake) also sporadically occur.

Table 1. List of the studied granites

No.	Province	Pluton	No.	Province	Pluton	No.	Province	Pluton	No.	Province	Pluton
1	Hidaka	Nisshotoge	43	Sanin	Neu	85	Sanyo	Masaki	127	Outer Zone	Ishizuchi (Omogo)
2	Hidaka	Pipairogawa	44	Sanin	Yubarako	86	Sanyo	Saga	128	Outer Zone	Takatsukiyama
3	Hidaka	Tottabetsu	45	Sanin	Yubara	87	Sanyo	Tsutsugatake	129	Outer Zone	Ashizuri
4	Hidaka	Oshirabetsu	46	Sanin	Rengeji or Daito	88	Sanyo	Kikuchi	130	Outer Zone	Kashiwajima
5	Hidaka	(Middle tonalite: Sarurugawa)	47	Sanin	Daito	89	Ryoke	Inada	131	Outer Zone	Okueyama
6	Hidaka	(Basal tonalite: Menashunbetsugawa)	48	Sanin	Hiyodori	90	Ryoke	Tsukuba	132	Outer Zone	Osuzuyama
7	Kitakami	Kuki	49	Sanin	Tamagawa	91	Ryoke	Yamanoo	133	Outer Zone	Ichifusayama
8	Kitakami	Taro	50	Sanin	Kumogi	92	Ryoke	Ikuta or Habakawa?	134	Outer Zone	Takakumayama
9	Kitakami	Tanohata	51	Sanin	Kaho	93	Ryoke	Shinshiro?	135	Outer Zone	Osumi
10	Kitakami	Miyako	52	Sanin	Sawara	94	Ryoke	Busetsu	136	Outer Zone	Shibisan
11	Kitakami	Himekami	53	Sanin	Itoshima	95	Ryoke	Kamihara or	137	Outer Zone	Mukaeyama
12	Kitakami	Goyosan	54	Sanyo (Mag)	Kake (or Nukui)	96	Ryoke	Kimigano	138	Outer Zone	Kinpozan
13	Kitakami	Tono	55	Sanyo (Mag)	Togochi	97	Ryoke	Ao	139	Outer Zone	Suzuyama
14	Kitakami	Hitokabe	56	Sanyo (Joetsu)	Wasada	98	Ryoke	Kabuto	140	Outer Zone	Nomamisaki
15	Kitakami	Senmaya	57	Sanyo (Joetsu)	Nihonkoku gneiss	99	Ryoke	Kabuto or Ao	141	Outer Zone	Yakushima
16	Kitakami	Akagane (Koganetsubo Mine)	58	Sanyo (Joetsu)	Ohibara	100	Ryoke	Kouoyama	142	Others・Unknown	Hikami
17	Kitakami	Matsuiwa (Oya Mine)	59	Sanyo (Joetsu)	Kusozu-Kogawa	101	Ryoke	Nojima	143	Others・Unknown	Kakkonda
18	Kitakami	Tabashine	60	Sanyo (Joetsu)	Gozu	102	Ryoke	Shido	144	Others・Unknown	Yaguradake
19	Kitakami	Obonai	61	Sanyo (Joetsu)	Hayadegawa	103	Ryoke	Namikata	145	Others・Unknown	Takidani
20	Abukuma	Shirakamidake	62	Sanyo (Kanto)	Sori	104	Ryoke	Hojo	146	Others・Unknown	Gilgit (Pakistan)
21	Abukuma	Taiheizan	63	Sanyo (Kanto)	Ashikaga	105	Ryoke	Gamano	147	Others・Unknown	Manshera (Pakistan)
22	Abukuma	Yuzawa	64	Sanyo (Naegi)	Agematsu	106	Ryoke	Kibe	148	Others・Unknown	[Xenolith/Omine]
23	Abukuma	Kodake	65	Sanyo (Naegi)	Naegi	107	Ryoke	Taio	149	Others・Unknown	[Xenolith/Kumano]
24	Abukuma	Wakamatsuyama	66	Sanyo (Naegi)	Toki	108	Green Tuff	Ryugamori	150	Others・Unknown	[Xenolith/Takatsukiyama]
25	Abukuma	Sekine	67	Sanyo	Kaoredani	109	Green Tuff	Nibetsu	151	Others・Unknown	[Xenolith/Okueyama]
26	Abukuma	(Abukuma: Kawabari)	68	Sanyo	Kaizukiyama	110	Green Tuff	Tanigawadake	152	Others・Unknown	[Xenolith/Ichifusa]
27	Abukuma	(Abukuma: Fusamata)	69	Sanyo	Kojaku	111	Green Tuff	Suzaka	153	Others・Unknown	[Xenolith/Osumi]
28	Abukuma	(Abukuma: Kaihosaku)	70	Sanyo	Suzuka	112	Green Tuff	Wada	154	Others・Unknown	[Xenolith/Nomamisaki]
29	Abukuma	(Abukuma: Miharu)	71	Sanyo	Tanakami	113	Green Tuff	Karasawa	155	Others・Unknown	[Xenolith/Koshikijima]
30	Abukuma	(Abukuma: Tanaka)	72	Sanyo	Gyojasan	114	Green Tuff	Kofu (Tokuwa)	156	Others・Unknown	[Metamorphic rock/Taki]
31	Abukuma	(Abukuma: Akanuma)	73	Sanyo	Nibori	115	Green Tuff	Kofu (Shosenkyo)	157	Others・Unknown	[Metamorphic rock/Saragai]
32	Abukuma	(Abukuma: Uzumine)	74	Sanyo	Mannari	116	Green Tuff	Tanzawa	158	Others・Unknown	[Metamorphic rock/Babasawa]
33	Abukuma	Ishikawa	75	Sanyo	Hiroshima (Minagi)	117	Green Tuff	Kadohara	159	Others・Unknown	[Metamorphic rock/Ishizuka pass]
34	Abukuma	Miyamoto	76	Sanyo	Hiroshima (Takahashi)	118	Green Tuff	Nogohakusan	160	Others・Unknown	[Metamorphic rock/Hohi]
35	Abukuma	Tabito?	77	Sanyo	Hiroshima (Onomichi-Mitsugi)	119	Green Tuff	Imajo	161	Others・Unknown	[Metamorphic rock/Kioo]
36	Abukuma	Tabito or Torizone	78	Sanyo	Hiroshima (Kake-Kabe)	120	Green Tuff	Uchiyama	162	Others・Unknown	[Quartz vein/Arakawa mine]
37	Abukuma	Irishiken? (dyke in gabbro)	79	Sanyo	Hiroshima (Chausuyama)	121	Green Tuff	Goto	163	Others・Unknown	[Quartz vein/Tonbara]
38	Hida	Funatsu	80	Sanyo	Masago	122	Green Tuff	Amakusa	164	Others・Unknown	[Quartz vein/Hanaguri]
39	Hida	Funatsu (Shogawa)	81	Sanyo	Hiroshima (Abu)	123	Green Tuff	Tomioka	165	Others・Unknown	[Quartz vein/Kurotaki]
40	Sanin	Ariake	82	Sanyo	Hiroshima (Omishima)	124	Green Tuff	Koshikijima	166	Others・Unknown	[Quartz vein/Koshikijima]
41	Sanin	Shirakawa	83	Sanyo	Nakayamagawa	125	Outer Zone	Omine	167	Others・Unknown	[Septa (Yubara)]
42	Sanin	Ogamo	84	Sanyo	Hiroshima (Aio)	126	Outer Zone	Kumano			

As for the granitic rocks, the same five types of fluid inclusions occur in them. Monophase liquid inclusions (type 1) may be formed by trapping low-temperature meteoric water after emplacement of granitic bodies, and the other four types (types 2, 3, 4 and 5) probably trapped fluids under cooling processes of the crystallized granitic magmas, unless different hydrothermal episodes were superimposed. Aqueous two-phase liquid-rich inclusions are the most ubiquitous among the types. Daughter mineral-bearing polyphase and vapor-rich inclusions are more frequently in Neogene (Green Tuff region) and Quaternary granitic rocks (e.g. Kakkonda, Takidani) than in the Cretaceous ones, or are also common in granitic rocks intruding contemporaneous volcanic rocks. The Neogene granitic rocks are thought to be eroded less than the Cretaceous ones, and their exposed levels are shallow. That is, the shallower plutons have the more saline fluids in inclusions. Experimental data on the NaCl-H_2O system and solidus of granitic rocks also shows that highly saline fluid in which halite is stable at room temperature are trapped in inclusions at shallower depths (< 5 km in depth).

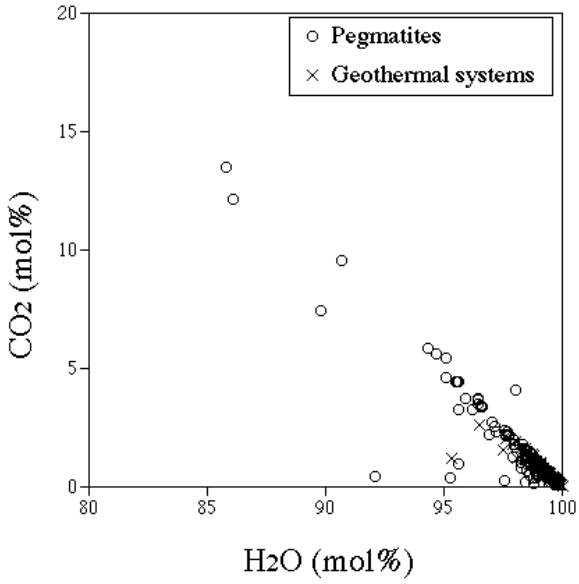

Figure 3. CO_2-H_2O plots for the fluid inclusions from geothermal systems and granitic rocks (pegmatites). "Pegmatites" includes samples of pegmatites, xenoliths in granitic rocks.

3. GAS ANALYSES

Details of the analytical systems and methods are shown in Sasada *et al.* (1992). The analytical system consists of the quadrupole mass spectrometer (QMS), capacitance manometer, cold trap, infrared furnace and vacuum system, connected with stainless steel tubing. Analyzed minerals are quartz and anhydrite from geothermal systems and quartz in pegmatites and xenoliths in the granitic bodies and their related rocks.

The bulk gas analyses show that H_2O is the main constituent of all the fluid inclusions 88-100 mol% (Figures. 3, 4), except for one sample from the Ryoke belt (CO_2 = 35 mol%). Results of analyses shows that the gas contents of fluid inclusions from geothermal systems are generally lower than those of granitic rocks (Figures. 3, 4). For example, CO_2 of inclusions from geothermal areas are less than 5 mol% (generally less than 1 mol%), whereas those of granitic rock exceed 10 mol% (Figure. 4).

As for the granitic rocks, CO_2 is the main gas for almost the samples (12 mol% >) except for the one sample. Subordinate amounts of N_2 (2 mol% >), CH_4 (4.3 mol% >) and Ar (0.003 mol% >) are detected, except for inclusions in pegmatitic quartz from the Hidaka belt where granitic magma was probably produced by anatexis of metamorphic rocks. Fluid inclusions from the Hidaka belt are characterized by relatively high CH_4 (0.16-9.6 mol%) and N_2 (0.03-1.8 mol%) contents, compared to CO_2 (0.09-0.94 mol%). Fluid inclusions from S-type granitic rocks in the Outer Zone of southwestern Japan also have slightly high contents of CH_4.

4. DISCUSSION

The fluid inclusions from pegmatites and their related rocks are richer in gas components than those from the geothermal systems (Figures. 3, 4). That is, the fluid inclusions from the geothermal systems generally consist of 99 mol% H_2O with less than 1 mol% gases. The CO_2-CH_4-N_2 ternary plot (Figure. 5A) shows different trends between the geothermal and granitic fluids. CO_2/CH_4 ratios of the fluid inclusions from the geothermal systems have a narrower range than that of CO_2/CH_4 ratios. On the other hand, data of granitic rocks (Figure. 5B) display that CO_2/CH_4 ratios have a wide range. These differences might be attributed to the formation environments of the fluid inclusions: the fluid inclusions in pegmatites probably trapped fluids prior to degassing and dilution, or were formed through remarkable fluid-rock interactions before trapping, whereas geothermal fluids may be mainly originated from meteoric water.

The variation of CO_2/CH_4 ratios for granitic rocks (Figures. 4B, 5B) may reflect the difference of fO_2 around the granitic rocks where the fluids flew. CO_2/CH_4 ratios of inclusions from magnetite-series granitic rocks are slightly higher than those from ilmenite-series ones (Figure 4B), and the former data are plotted near the CO_2-N_2 axis of the CO_2-CH_4-N_2 ternary plot (Figure. 5B). Magnetite-series granites are thought to be formed under higher fO_2 condition

than ilmenite-series (e.g. Ishihara, 1977). The gas ratios are consistent with it, and the analytical results suggest that gas compositions of fluid inclusions are probably controlled by source of granitic magma. The high CH_4 and N_2 contents for the Hidaka granitic rocks are possibly related to the partial melt of sediments. Gas compositions of inclusion fluids are probably controlled by fluid-rock interaction in the granitic rocks and/or in sedimentary rocks around the granitic rocks, or, possibly, by source of granitic magma.

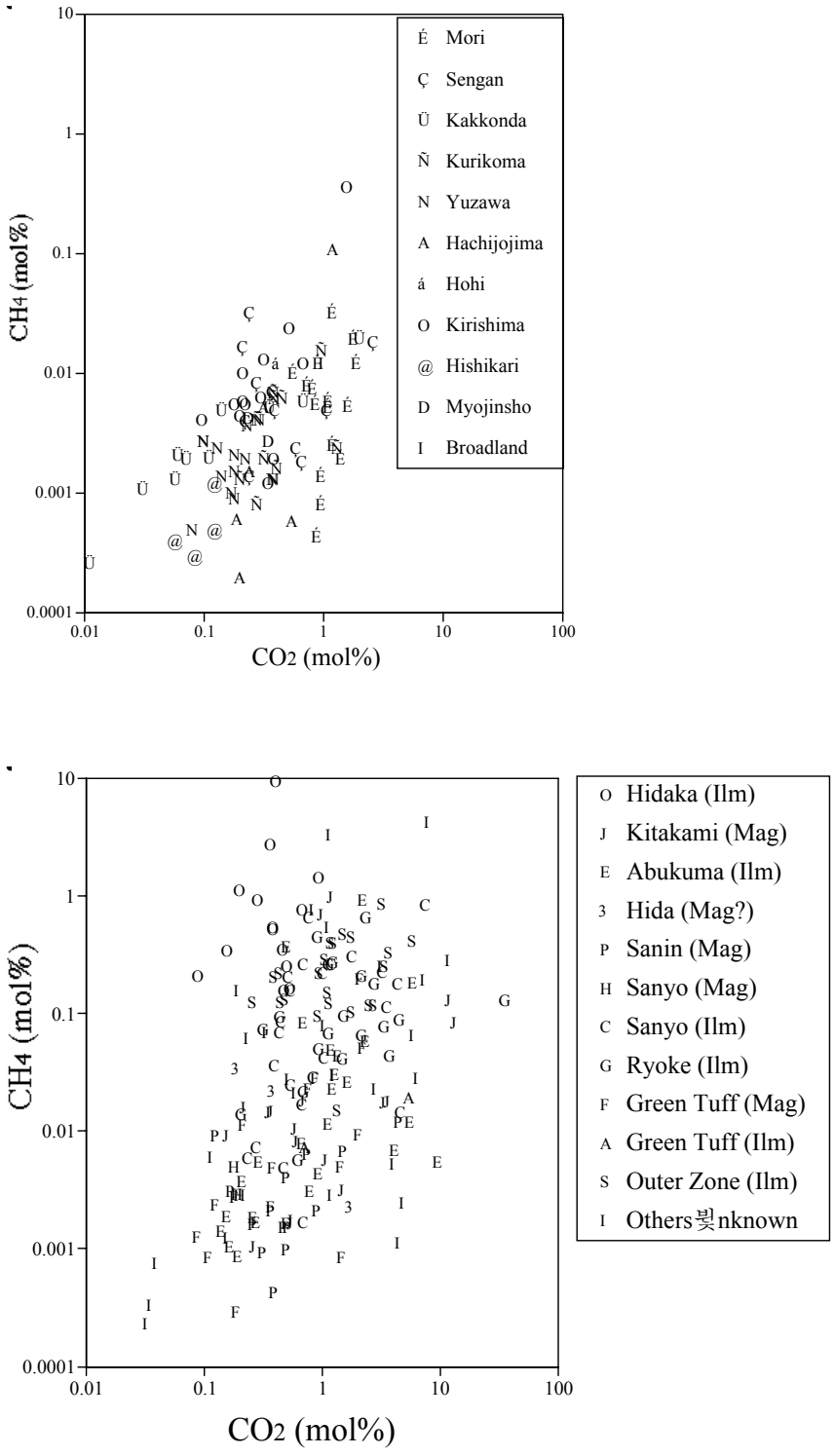

Figure 4. The variation of CO2/CH4 ratios for granitic rocks.

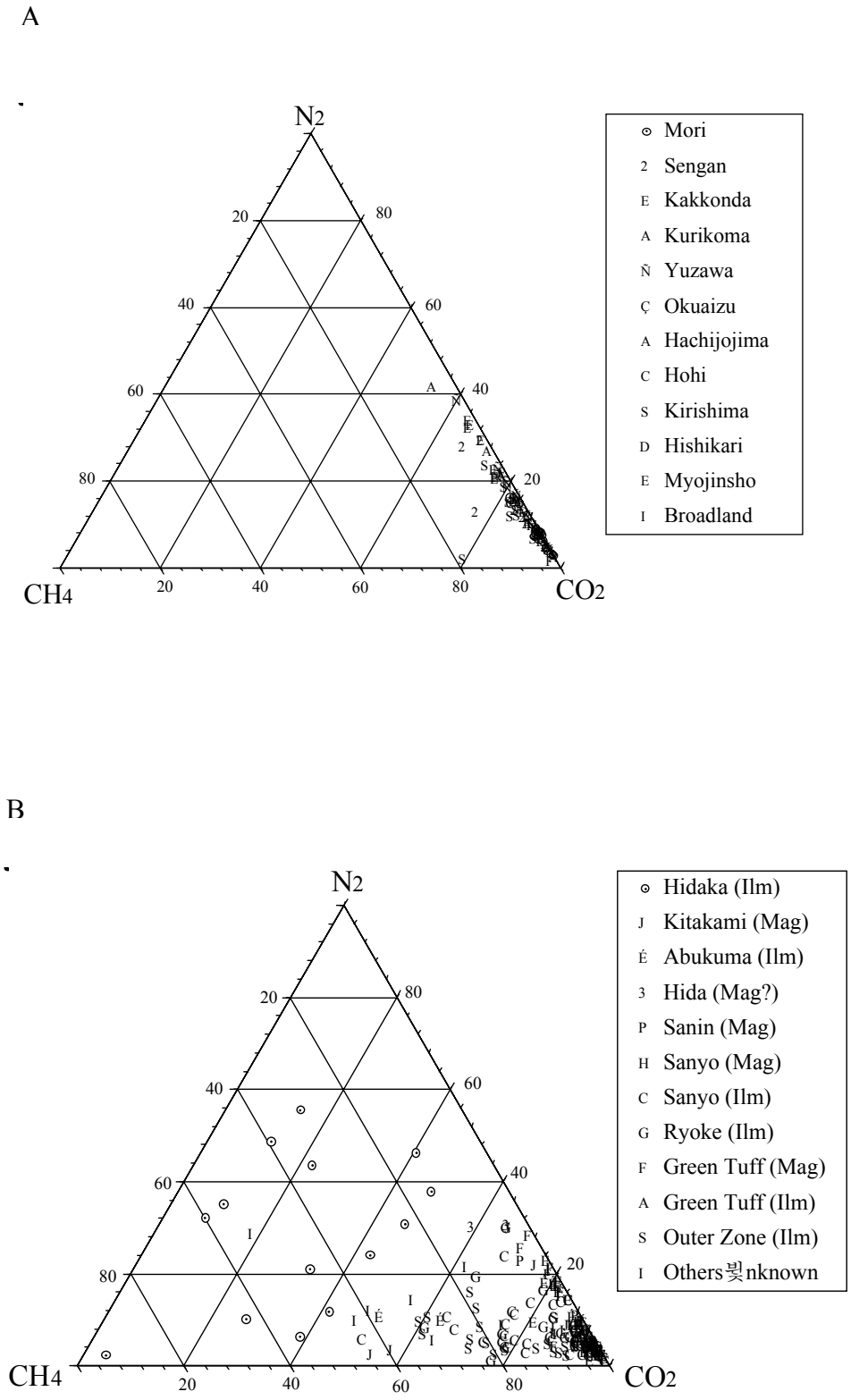

Figure 5. CO_2-CH_4-N_2 ternary plots for fluid inclusions from geothermal areas (A) and granitic rocks (B). Ilm: ilmenite-series; Mag: magnetite-series.

5. SUMMARY

In geothermal systems and granitic rocks in Japan five types of fluid inclusions occur: (1) aqueous two-phase liquid-rich, (2) aqueous two-phase vapor-rich, (3) monophase liquid, (4) liquid CO_2-bearing aqueous, and (5) daughter mineral-bearing multiphase inclusions occur. Occurrences of the fluid inclusions give us clues to reveal geological environments where and when the fluids were trapped. Gas analyses show that H_2O is the main component of all fluid inclusions, and CO_2 is the main gas for almost the samples with subordinate amounts of N_2, CH_4 and Ar, except for inclusions from Hidaka. The gas contents of fluid inclusions of granitic rocks are generally higher than those from geothermal systems. This suggests that fluid inclusions in pegmatites trapped fluids prior to degassing and dilution, or were formed through remarkable fluid-rock interactions before trapping. The fluid inclusions from Hidaka are characterized by relatively high CH_4 and N_2 contents, compared to CO_2. The fluid inclusions from S-type granitic rocks in the Outer Zone also have slightly high contents of CH_4. The high CH_4 and N_2 contents possibly related to the partial melt of sediments. The variation of CO_2/CH_4 ratios may reflect the difference of fO_2 around the granitic rocks where the fluids flew. CO_2/CH_4 ratios of inclusions from magnetite-series granitic rocks are slightly higher than those from ilmenite-series ones. Magnetite-series granites are thought to be formed under higher fO_2 condition than ilmenite-series. The gas ratios are consistent with it, and the analytical results suggest that gas compositions of fluid inclusions are probably controlled by source of granitic magma.

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