Gas Geochemistry and Carbon-13 Systematics of Ungaran Geothermal Field, Central Java, Indonesia

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ABSTRACT

The Ungaran geothermal field has numerous thermal manifestations, e.g. fumaroles and hot springs with temperatures ranging from 31°C to 86°C. Fluid samples have been taken from fumaroles at Gedong Songo (Dasamuka) and gas bubbling from various hot springs to analyse its gas composition and carbon-13 ratio. The result of the gas analysis shows that the Ungaran's fluids are of meteoric origin with some degree of magmatic contribution, with CO_2 as the dominant gas. CO_2-H_2-Ar gas ratio grid shows that only samples from Gedong Songo fumaroles that plotted inside the two-phase grid with temperature about 280°C, while other samples plotted outside the grid indicates re-equilibration at lower temperatures. The range of carbon-13 ratio is about -8-32 $^o/_{oo}$ to -5.89 $^o/_{oo}$ (vs PDB). These values indicate that CO_2 in Ungaran geothermal fluids are magmatic origin, not from soil or carbonate source.

1. INTRODUCTION

Ungaran geothermal field is located about 30 km southwest of Semarang, the capital of Central Java province, Indonesia. Ungaran lies in quaternary volcanic belt namely Solo zone, which is in between north Serayu mountain range in the west and Kendang ridge in the east (van Bemmelen, 1949). The mountain is also a member of the volcanic arc with mount Sindoro, Sumbing, Telomoyo, Merbabu to the north. The Ungaran volcanic complex consists of older and younger volcanic body based on its volcanic activity history. The old Ungaran age is about 0.5 Ma, while young Ungaran is 0.3 Ma and situated inside the caldera formed by old Ungaran activity (Kohno et al, 2006).

Several thermal manifestations are spread around Ungaran in form of fumaroles, hot springs and altered grounds. The temperatures of the fumaroles are about 86°C and are located in Gedongsongo area, named Kawah Dasamuka as famous tourist attraction. While the hot springs are found in some locations including Gedongsongo, Nglimut, Diwak, Kaliulo, Kalianget and Klotok with temperature vary between 33°C to 46°C. Most of the hot springs is bicarbonate type, while hot springs located in fumarolic fields are acid sulfate type (Phuong et al, 2012).

Some geochemical studies have been done in attempt to characterize the properties of Ungaran geothermal field, since the early 1980's. The most recent geochemical study was done by Phuong et al (2012), which applied water geochemistry and soil gas survey to delineate the fracture system and hydrogeochemical model of Ungaran. In this paper, a gas geochemistry and carbon isotopic study is applied to characterize fluids equilibration temperature and its origin.

2. METHOD

2.1. Sampling

The gas samples were collected from fumaroles and bubbling gases associated with the hot springs. These samples were taken using pre-evacuated glass bottle filled with 50 mL of 6N NaOH (Giggenbach and Goguel, 1989; Arnorsson et al, 2006). The steam was condensed inside the bottle while CO_2 and H_2S are absorbed by NaOH solution. The rest of the gases were occupied the headspace in the sampling bottle due to its low solubility in water. Sampling was done in various locations around Mt. Ungaran as shown in figure 1.
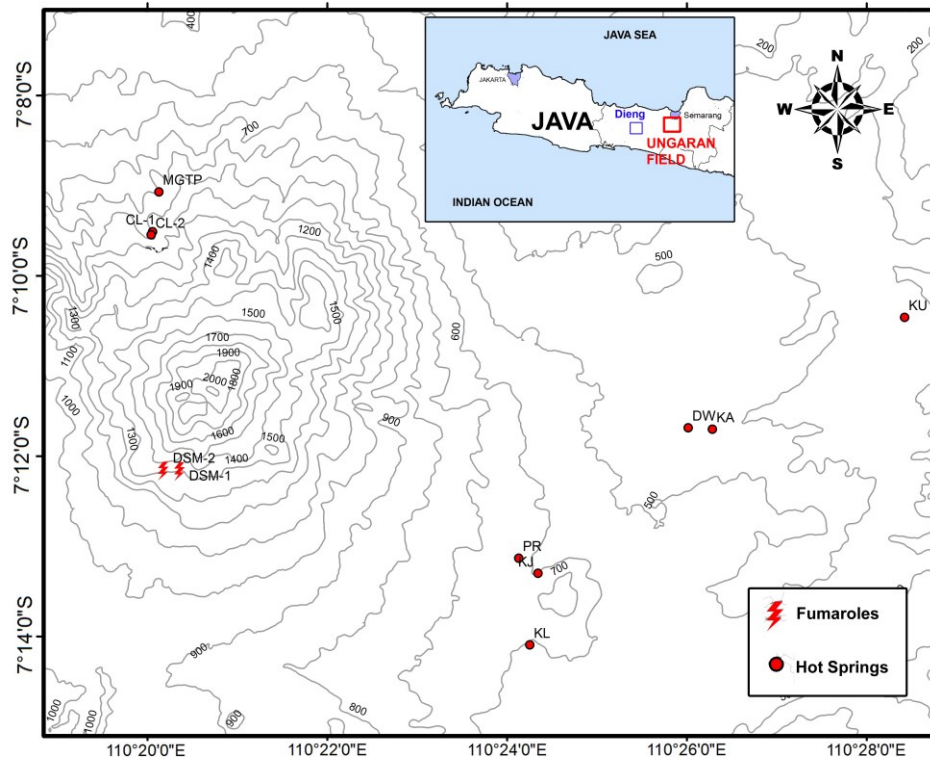
Figure 1. Sampling location of Ungaran thermal manifestation.

2.2. Analysis

Gas analysis was done by gas chromatography (Agilent 7890A) for He, H_2, N_2, O_2, Ar and CH_4 using Porapak column, He and Ar gas carrier and thermal conductivity detector. CO_2 and H_2S were analyzed using titration method (Giggenbach and Goguel, 1989). Carbon isotope of CO_2 analysis was performed using mass spectrometer VG SIRA-9 and measured against PDB standard.

3. RESULTS

Table 1 shows the analysis result of fumaroles and bubbling (seeping) gases. All samples have relatively small steam content, i.e. 16-37% mol, compare to steam derived from drilled wells. The low steam content inferred the effect of condensation during the rising of the fluids to the surface. This condensation might occur either due to deep location of reservoir or steam absorption into shallow aquifer, leads into bicarbonate water. The most dominant gas species is CO_2 that constitutes up to 99% of the NCG's, while H_2S composition is less than 1.2% mol, lower than some other geothermal fluids i.e. up to 32% (e.g. Kiyosu and Yoshida, 1988; D'Amore et al, 1992; Prasetio et al, 2010).

Table 2. Gas and $\delta^{13}C_{CO2}$ analysis results of Ungaran fumaroles and seeping gases. ND = not detected, NA = not analyzed. Others = He, H_2, N_2, Ar, CH_4 (measured, but not shown).

Code	Location	Gas (% mol)			NCG (%mol)	Steam (% mol)	$\delta^{13}C_{CO2}$ ($^{o}/_{oo}$ PDB)
		CO_2	H_2S	Others*			
DSM-1	Dasamuka	96.65	1.16	2.19	80.84	19.16	N.A.
DSM-2	Dasamuka	96.99	1.03	1.97	62.89	37.11	-7.44
KA	Kalianget	99.62	0.02	0.36	74.56	25.44	-5.80
KJ	Karangjoho	99.78	0.02	0.21	75.77	24.23	-5.92
KL	Klotok	95.01	0.04	4.95	76.93	23.07	-6.56
KU	Kaliulo	98.46	0.03	1.51	71.84	28.16	-6.34
MTP	Margotopo	99.68	0.22	0.10	72.86	27.14	-8.32
CL-1	Curuglawa	98.27	0.49	1.24	83.58	16.42	-7.92
CL-2	Curuglawa	99.65	0.29	0.06	65.23	34.77	-8.31
DW	Diwak	99.40	0.03	0.57	76.91	23.09	-7.13

4. DISCUSSION

4.1. Origin of gas and geothermometry

The origin of the gases can be inferred from a He-N_2-Ar trilinear diagram (Giggenbach, 1997) as shown in Figure 2. Gas composition of Ungaran geothermal fluids have typical characteristic of volcano-associated with largely meteoric contribution as seem from its N_2/Ar ratio. Fumarole samples at Dasamuka (DSM-1 and DSM-2) have higher N_2/Ar ratio, i.e. 268 and 387, indicate higher contribution of magmatic source than bubbling/seeping gas samples with N_2/Ar ratio below 100. This may suggest that Dasamuka (Gedongsongo) area is the up flow zone of the Ungaran geothermal field.
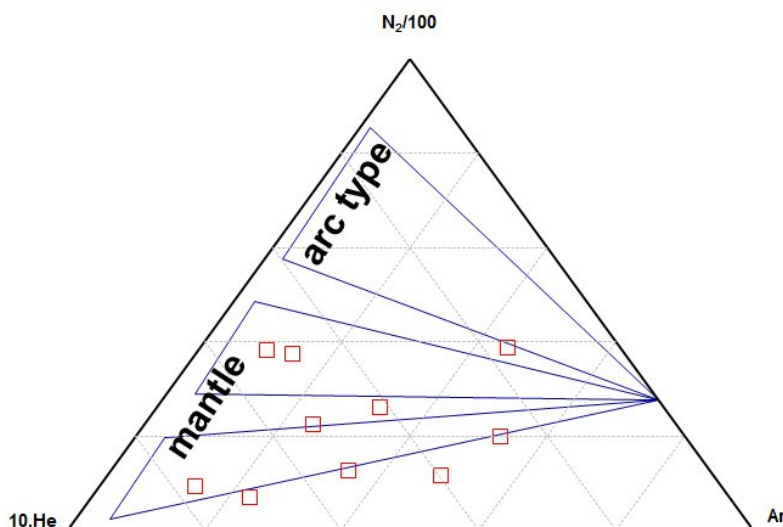

Figure 2. He-N_2-Ar trilinear diagram (Giggenbach, 1997) of Ungaran fluids.

Estimation of reservoir temperature was also done using various gas geothermometer calculations. Due to condensation process, as inferred from NCG concentration, single species geothermometer can not be applied (e.g. T_{CO2}, T_{H2S}). Application of these geothermometers would lead to over-estimation of reservoir temperature. Thus, gas-gas ratio geothermometers, i.e. H_2S/H_2 and CO_2/H_2 (Arnorsson and Gunlaugsson, 1985), H_2/Ar and CH_4/CO_2 (Giggenbach, 1991), were applied to minimize the effect of steam condensation (table 2). Calculation results show that geothermometer involving CH_4 for samples collected from bubbling/seeping gases at Margotopo and Curuglawa tends to have higher temperature estimation, up to 442°C. This phenomenon is probably caused by slow equilibration rate of CH_4, thus this geothermometer may reflect deeper equilibration temperature (Giggenbach, 1991). On contrary, under-estimated temperature were obtain for also samples collected from bubbling/seeping gases, i.e. at Kali Ulo, Margotopo and Curuglawa when using geothermometer calculations involving H_2 species (i.e. H_2/Ar, CO_2/H_2 and H_2S/H_2). These two contradictions are probably caused by some factors such as condensation and removal of gas species from the steam. Thus, it may suggest that Kaliulo, Margotopo and Curuglawa are out flow area.

Table 2. Gas-gas geothermometer of Ungaran geothermal fluids.

Code	Location		Geothermometer (°C)			
			H_2/Ar	CO_2/H_2	H_2S/H_2	CH_4/CO_2
DSM-1	Dasamuka	Fumarole	326	229	289	282
DSM-2	Dasamuka	Fumarole	339	227	290	284
KU	Kali Ulo	Bubbling gas	116	< 100	220	268
MTP	Margotopo	Bubbling gas	< 100	< 100	178	394
CL A	Curuglawa	Bubbling gas	< 100	< 100	186	442
CL B	Curuglawa	Bubbling gas	143	< 100	187	392

Beside simple geothermometer calculation, pair of gas-gas geothermometer, i.e. CO_2/Ar and H_2/Ar was also utilized to estimate not only reservoir temperature, but also to liquid-vapor equilibrium in reservoir (Giggenbach, 1991). Using a single fixed redox state (R_{H} = -2.8), the graphical representation of this geothermometer can be seen in figure 3. The graph shows that there are only two samples plotted inside the grid, i.e. fumarole samples (DSM-1 and DSM-2), and indicates two-phase equilibration with temperature

between 275-290 °C. Meanwhile, other samples are plotted below liquid equilibration line which may indicate H_2 re-equilibration at lower temperature, atmospheric Ar introduction or more negative redox state (Powell, 2000). Based on these two geothermometer methods (table 2 and fig. 3) the temperature of reservoir is estimated c.a. 280°C.
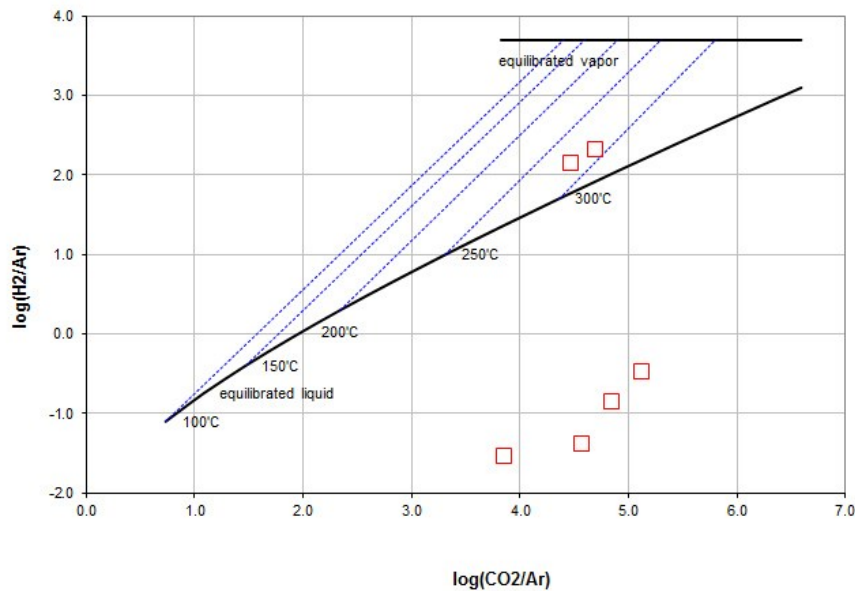

Figure 3. CO_2/Ar and H_2/Ar geothermometer grid of Ungaran fluids. Only fumarole samples plotted inside the grid.

4.2. Isotope ^{13}C of CO_2

As seen from Table 1, $\delta^{13}C_{CO_2}$ composition of thermal manifestation vary between -8.32‰ and -5.80‰. Compared to other $\delta^{13}C_{CO_2}$ values as summarized by Birkle et al (2001), this value overlaps values of $\delta^{13}C$ from magmatic CO_2 and atmospheric CO_2 sources, not apparent contribution from soil CO_2 (fig. 4), thus indicates the contribution of magmatic and atmospheric sources. This finding is also in agreement with the gas analysis results (fig. 2) which inferred magmatic and meteoric contribution to the fluids. Although the main reservoir rocks are inferred to be consisted of Tertiary marine sedimentary rocks (Budiardjo et al, 1997) the $\delta^{13}C$ value only slightly overlaps with $\delta^{13}C$ of marine bicarbonates value. The $\delta^{13}C$ is also distributed into more enriched values towards east and southeast of the field (Fig. 5).
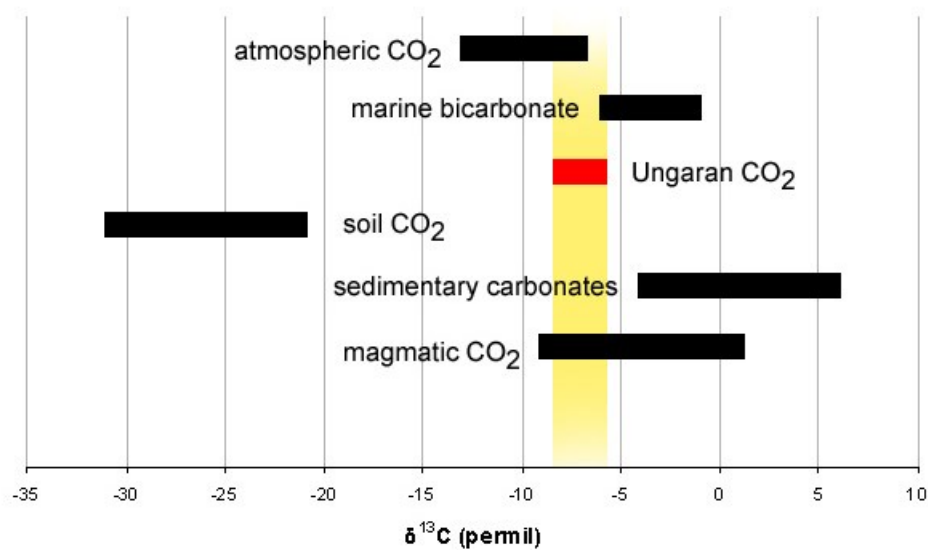

Figure 4. $\delta^{13}C_{CO_2}$ composition of Ungaran thermal fluids (red ribbon), compare to other $\delta^{13}C$ sources (Birkle et al, 2001).
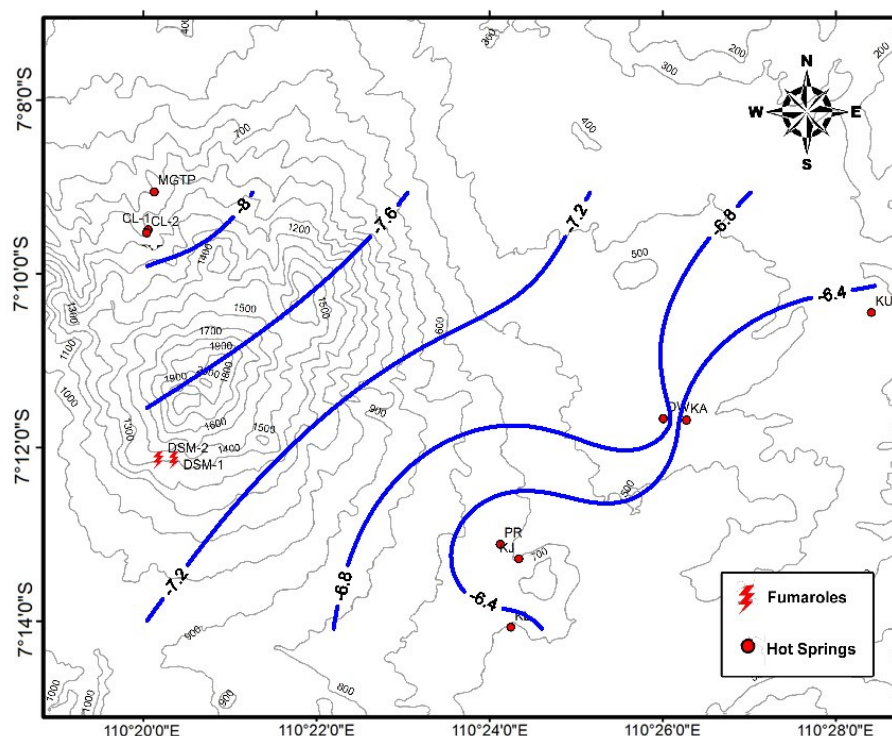
Figure 5. Distribution of δ¹³C in Ungaran geothermal field. The value is more enriched towards east-southeast of the field.

5. CONCLUSION

Ungaran geothermal fluids are meteoric origin, mixed with magmatic source as can bee seen from its gas compositions. The fluids are mainly ascending at Dasamuka (Gedong Songo) area as an up flow zone. The gases in reservoir i.e. CO_2-H_2-Ar shows two-phase equilibrium at temperature about 280°C. The $\delta^{13}C_{CO2}$ value inferred its origin, i.e. magmatic and atmospheric, which is in agreement with gas analysis results. Moreover, $\delta^{13}C_{CO2}$ value is more enriched towards east and southeast of the field. Further study is needed to asses the factors influencing the variation of $\delta^{13}C_{CO2}$ value.

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