GEOCHEMISTRY OF THE UNGARAN GEOTHERMAL SYSTEM, CENTRAL JAVA, INDONESIA

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ABSTRACT

Due to water chemistry analysis, most of thermal waters in Gedongsongo are characterized as diluted bicarbonate waters. This water is presented a mixing process between geothermal water and shallow groundwater. Only water in Kaliulo has a very high Na and Cl concentration (more than 4000 ppm) as well as HCO₃. Thus, it can be explained that these water flow through marine sediment at depth. Isotopic results confirmed that a similar origin of the geothermal fluids and the meteoric water as well as small oxygen shift due to interaction with country rock. Moreover, it is elucidated that water in Kaliulo represents not only mixing between meteoric water and connate water that trapped into Tertiary marine sediment at depth but also rock/water interaction. Emanometry of Rn, CO₂, and Hg are also conclusive in the presence of fracture zone for the migration of geothermal fluid and center zone in the southern flank of the Ungaran geothermal system. The CO₂ and Hg data show consistency with suggestion that maybe the centre zone of geothermal system around Gedongsongo area. CO₂ data is presented of fault along valley nearby the fumarole.

Key Words: Hydrogeochemistry, Stable isotope, Soil air gas.

INTRODUCTION

The Gedongsongo area, main manifestation of the Ungaran geothermal prospect, is located in the southern part of Ungaran volcano. It is approximately 30 km southwest of Semarang, the capital city of Central Java province, and lies between 7°13’30” – 7°11’00” S latitude, and 110°19’30” – 110°22’00” E longitude (Fig.1B). Several thermal manifestations occur at Gedongsongo area as fumaroles, hot/warm springs, and acid surface hydrothermal alteration rocks.

Recently, Budiardjo et al. (1997) reported the result of resource characteristics of the Ungaran geothermal field, suggesting the hot water at Gedongsongo originate as a steam heated meteoric water while hot spring waters located in other area fall into neutral bicarbonate-sodium chloride water originated as diluted outflowing waters from the Ungaran volcano geothermal reservoir. The aim of this paper is to understand hydrogeochemical conditions of thermal water, the center of the Ungaran geothermal system, and also fracture zone due to soil air gas survey.

GEOLOGICAL SETTING

Structural analysis in this area revealed that the Ungaran volcanic system is primarily controlled by the occurrence of the Ungaran collapse structure that runs from the West to Southeast of the Ungaran. The old volcanic rocks of the Pre-caldera formation are controlled by the Northwest-Southwest and Southeast- Southwest faulting system. The volcanic rocks of the post caldera appear to be less structurally controlled by regional faulting system. The pre-caldera volcanic rocks and the Ternary marine sedimentary rocks are inferred to be the main reservoir rocks.
The Gedongsongo is the main geothermal resources area of the Ungaran prospect associated with the Quaternary andesitic volcanic complex of G. Ungaran. The stratigraphy of the Ungaran volcanic area composed of andesitic and perlitic lava and pyroclastic rocks of volcanic breccia of post Ungaran caldera formation. This formation, overlying the old volcanic rocks was formed before the Ungaran caldera formation which in turn overly Tertiary volcanic rocks (Budiardjo et al., 1997).

**SURVEY AREA AND SAMPLE COLLECTION**

The samples collected were as follow: 46 for soil air gas survey (Radon, CO₂, Hg-air and Hg in soil), 25 for chemical analysis (HCO₃, Cl, NO₃, SO₄, Na, K, Mg, Ca) and stable isotope analysis (¹⁸O and ²H) of water. Water samples were taken from thermal springs: five in the Gedongsongo around fumarole, two in Kendalisodo and one in Diwak, Kaliulo. Eleven local water samples are collected in Gedongsongo, Bumen, and Banaran, including well waters in Candi village, Gelaran and river water in Kendalisodo, Diwak, Derekan and Kaliulo (Fig.1A). Temperature, electrical conductivity, pH of the water samples were measured in the field.

Soil air gas surveys were conducted in the area approximately 1.3 km NS by 1.5 km NE in the east of the fumarole (Fig.1A). Measurements were made at intervals of 50 and 100 m for soil air gas. A hole of 0.6 m depth was made by hammering steel pipe. Soil samples at 0.6 m depth were collected in the field and sealed in plastic bags to analyze mercury in soil in laboratory. To capture Hg in soil from the ground, gold needles were hanged into the hole of 0.6 m in depth and 5 cm in diameter for a week. An available gold needle should be pure with 10 cm length, 1 mm diameter, 1.5 g weight and 3.16 cm² in the effective surface (Koga, 1984) (Fig. 2).

**FIELD MEASUREMENT AND ANALYSIS METHODS**

**Soil Air Gas**
Radon and Thoron gas measurement is carried out on-site by using a Radon detector (portable RD-200, EDA Instruments Co. Ltd.) (Fig. 3), theirs concentration were calculated from three counts in each minute obtained from three sequential minutes. A gas detector tube (Komyo-Kitagawa Instruments Co. Ltd.), the SA-type with precision of 0.1-2.6% is usually used. However, in the Ungaran geothermal area, it presents high CO₂ concentration over 2.6% so it was rechecked by higher precision of 1 – 20%. The gold wire is analyzed in laboratory by Rigaku Mercury SP-3 (Nippon Instruments Co.).

**Stable Isotope**
On-site sampling procedure for isotope analysis as described follow. First, filter the water using 0.45 mm HA and Sterifil Aseptic System (Fig 4). A bottle ought to rinse inside and out with the filtered water to be sampled 3 times. Then cap the bottle tightly as possible. Twenty-milliliter samples were collected for stable isotope analysis.

Mass spectrometric methods are by far most effective means for measuring isotopic abundances. A mass spectrometer separates charge atoms and molecules on the basic of their masses based on their motions in magnetic and/or electrical fields (Hoefs, 1980).

**RESULT AND DISCUSSION**

**Water Chemistry**
Chemical composition as well as type of the hot/warm springs and local cold water (such as well and river water) collected in Gedongsongo area are presented in Table 1. The chemical and physical processes affecting the thermal water composition and their spatial relationship can be utilized in a series of diagrams such as Cl-HCO₃-SO₄ and Na/1000-K/100- ÓMg diagram (Giggenbach, 1988) showing the relative concentrations of aqueous components. The conductivity and temperature ranges from 0.036 mS/cm to 19.83 mS/cm and up to 60°C, respectively. In general, these waters are neutral pH from 5.36 to 7.87 but UGW-1 is acidic pH (3.45).

**Cold water**
There were twelve samples of cold water collected during field works. All the local cold waters of Gedongsongo area emerge from the volcanic rocks. The emergence altitude of these waters varies from sea level to a maximum of
1100 m a.s.l. From Fig. 5, these waters are HCO$_3$-waters and plot within the bicarbonate sector of the triangular diagram excluding UGW-1, UGW-2 and UGW-15A, 15B are near acid sulphate water and chloride-bicarbonate water, respectively.

These waters can be defined into three groups such as Ca-HCO$_3$ (UGW-5, 13, 18), Ca-Mg-HCO$_3$ (UGW 6, 7, 8A, 8B, 14, 16, 17B) and Na-Ca-HCO$_3$-Cl (UGW- 10). Moreover, most of the parameters of cold water are neutral pH from 6.02 to 7.87, low Cl, K and Na less than 10ppm, and 15ppm respectively; and Mg up to 20ppm except, HCO$_3$- concentration is higher from 100 to 300ppm because shallow groundwater is rich of bicarbonate. Especially, UGW-10 and UGW-16 have high Ca content represented effect of limestone formation in the eastern part of this area. Generally, they are more dilute than are hot water with conductivity usually small (less than 1 mS/cm). This is likely to be the result of the addition of CO$_2$-rich gas to the shallow groundwater, resulting in the conversion of dissolved CO$_2$ to HCO$_3$.

**Thermal water**

Thermal waters in the area have Ca-Mg-(Na)-(Ca)- HCO$_3$, Ca-Na-Mg-SO$_4$-HCO$_3$, and Na-Ca-Cl-HCO$_3$ group. The Ca-Mg-HCO$_3$ waters characterized by low salinity (Cl) and relatively high temperature (from 22 – 56°C). HCO$_3$ is the principal anionic component. Its concentration varies in a wide range, from 39ppm to 465ppm. These waters show very low concentration of Cl and SO$_4$ below 3ppm and 13ppm, respectively but some samples show high SO$_4$ content of 245ppm (UGW-1), 137ppm (UGW-2). It is representative of mixing with local recharge water (rich bicarbonate water). On the other hand, high content of HCO$_3$ in water samples is due to dissolution of CO$_2$ gas at depth or effected by the limestone formation in the east of Ungaran (UGW-9, 15A, 15B, 17A, 21) thermally degradated at depth and release CO$_2$. This released CO$_2$ will dissolve into the water and form HCO$_3$.

The UGW-1 and UGW-2 present acid sulphate water belong to Ca-Na-Mg-SO$_4$-HCO$_3$ group. It has high concentration of NH$_4$ as well as SO$_4$ (components associated with steam heating), and variable but low concentrations of F less than 0.25ppm and Cl below 1.5ppm (components associated with magmatic volatiles or with high-temperature interactions between rock and water) (Janik et al, 1992). However, relatively far toward the south of fumarole, SO$_4$ concentration of UGW-11 (50.33ppm) is lower than that of UGW-1, 2 as well as Ca and Mg component while increase HCO$_3$ content to 97.6ppm. Chloride is trace amount in UGW-11. Most of steam-heated springs occur in the andesitic highland of the south flank of the Ungaran volcano.

Two separate areas of Cl-HCO$_3$ warm springs were investigated. The first area occurs in Kaliulo (UGW-9) in the eastern part of Ungaran. The second area is located in Kendalisodo-southeast of the Ungaran region (location UGW-15A, 15B, Fig. 1). The neutralchloride warm springs in Kaliulo (about 39°C) that issue from altered andesitic rocks surrounding by rice field with surface discharge less than 1 liter/min. High Na and Cl concentration (more than 5000ppm) may be the water flows through marine sediments at depth. Because CO$_2$ bubbling was observed in the discharging water at Kaliulo is evidence for this possibility. This means CO$_2$ has partial pressure in the water.

Figure 5 and 6 present Cl-SO$_4$-HCO$_3$ and Na/1000- K/100-ÖMg diagram Giggenbach (1989). From Figure 6, all waters in Gedongsongo area are plotted within ‘immature water’ field excluding UGW-9 in ‘partially equilibrium’ field. As Giggenbach (1989) pointed, KNa temperature reflects conditions at considerable depth. Plot of UGW-9 suggest the temperature at depth in this area, about 1600°C. However, most waters at Gedongsongo are dilution or other effects as they are plotted on the corner of Mg. On the other hand, this diagram also expresses waters that are suitable for using geothermometry to predict reservoir temperature (Nicholson, 1993). However, minimum value of reservoir temperature that can be estimated by using geothermometer for water in Kaliulo is approximately 140°C.

**Stable Isotope**

The isotopic data of the springs, well and river waters collected in Gedongsongo and its surrounding area are reported in Table 1. All these waters are plotted in the dD versus d$^{18}$O diagram and they lie on or close to the meteoric water line.
(Fig. 7) with the exception of the three samples (9, 15A, 15B) that show an oxygen-shift as well as enrichment of deuterium. This may be an evidence for meteoric origin of geothermal waters in the region. The lower in deuterium of thermal water than that of cold water, representing the highland recharge, where waters originated (altitude more than 950m up to 1500m) and no oxygen-18 shift is observed. It means that there is no isotopic exchange between hot waters and host rock. The oxygen shift that is due to water-rock interaction is from 0.2 to 4‰. The oxygen-18 content around Gedongsongo are around -8‰, it means that the oxygen shift is around 0 (about 0.2). While the samples in Kendaliisodo (15A, 15B) their oxygen shift is around 2.7. The longest oxygen shift is found in Kaliulo (UGG-9). The correspondence of deuterium contents of thermal and local meteoric waters is generally taken to indicate a predominantly meteoric origin of geothermal discharges, while the enrichment in oxygen-18 is taken to reflect isotopic exchange of the waters with reservoir rocks at elevated temperatures.

Sample UGW-9, 10, 17A, 17B, 18, 19, 20 and 21 in Kaliulo, Diwak, Tangkil and Derekan with more enrichment of dD than in Gedongsongo. The isotopic composition of the well (UGW-10, 17B, 18) tends to be more enriched. This shows that there is a process of evaporation in those depths. If the UGW-9 just has oxygen shift without enrichment of deuterium, it can state that this water was formed by water-rock interactions as explanation of UGW-15A, B samples in Kendaliisodo. Because the water interacted completely with rock, the oxygen will be shifted to the right. However, as shown in Table 4.5, meteoric water near the same elevation of UGW-9 is close to UGW-10 and UGW-18. Then there is a possibility to consider that mixing between meteoric water with connate water.

From Fig 7, we can state that the trend of water in Kaliulo is as same as in other sedimentary basin, i.e. enrichment of deuterium and strong positive shift \(^{18}O\). The characteristics deviation from the meteoric water line for sedimentary basin brines maybe is attributed due to isotopic exchange with \(^{18}O\) –rich sedimentary minerals particularly carbonate at elevated temperatures. Nevertheless, the isotope shift to the left side of water in Kaliulo is interpreted that it originated as mixing between connate water and meteoric water along with precipitated fracture minerals (carbonaceous formation) and subsequently been diluted by meteoric waters.

The solution of thermal water at Kaliulo that originates from mixing between meteoric water and connate water that has been altered by reaction with rock is also more convinced by correlation between deuterium and chloride (Fig 8). Constate water has high D and \(^{18}O\) content so the thermal waters that mix with this water are more enrichment of dD (-31.03 to - 20.78‰) and \(^{18}O\). On the other hand, the more \(^{18}O\) content of the water may result from equilibrium with rocks at high temperature, but the higher deuterium content cannot similarly explain and it is possible that some of these waters results from surface evaporation (foregoing premise). The thermal water probably originates at high-altitude precipitation with an isotopic composition similar to point UGW-10, and then it gains chloride and undergoes an oxygen isotope shift to a composition near UGW-9. This implies that the water of UGW-9 (Cl > 4000ppm) was formed by a mixing of the overlying connate water trapped in marine sediment.

From Fig 8, mixing possibility of water in Kaliulo is more plausible due to the similar content of Cl vs. dD to other mixing between connate water and meteoric water of Green Tuff formation, Ibusuki thermal water, Japan and Campi Flegrei, Italy. The chemistry of Green Tuff thermal waters is strongly controlled by interaction between hot meteoric waters and the submarine formations forming Na-(Ca)-Cl-SO\(_4\)-type waters (Sakai and Matsubaya, 1977). Like, Shimogamo of Izu Peninsula, Ibusuki is one of the typical coastal thermal water system in Japan (Sakai and Matsubaya, 1977). It is formed by mixing process of meteoric and oceanic waters in chemical interactions between the thermal waters and volcanic rocks. The coastal thermal waters of Campi Flegeri, Italy are quite distinct isotopically and appear to require a generative process that would enrich seawater in \(^{18}O\) and deuterium while depleting it in chloride (Truesdell et al., 1980).

**Soil Air Gas**

*Abnormality and subdivision of area based on Radon content*

Total concentrations of Radon and Thoron gases
are shown in Fig 9. High abnormality was detected around the fumarole (UGG-1, 2, 3, 7, 24, and 25), valley in the east of the fumarole (UGG-6, 19, 20, 21) and around manifestation in the west of the fumarole (UGG-17, 32).

From cumulatively frequency plot of Radon and Thoron gases, there are primarily within high class excluding some points have high abnormality concentration of Radon (more than 350 cpm) (Fig 10). This represents fluctuation in a highly fractured area, and reflects its high change range (Fig 9).

Figure 11 presents a preliminary map of the Rn/Tn ratios. Zones where the Rn/Tn ratio is high indicate fault zones extending to deep levels, trending NS along valley next to the fumarole. There are two abnormality of Rn/Tn in the southeast and west of the fumarole (UGG-27, 32) but around there show low Rn/Tn ratio so that values are not evidence for determining location of fault. Besides, some sampling sites such as UGG-31 to UGG-36 and UGG-37 to UGG-46 in the western and eastern part, respectively, have very low Rn/Tn ratio (up to 0.1). This likely presents margin of geothermal system, limited geothermal activities occurred around main manifestation in Gedongsongo area.

**CO2 gas concentration**

CO2 gas is relatively inert and exsolved early from a degassing magma body (Wu Y, 2003). At Gedongsongo the CO2 gas content is relatively high (1 – 10%), excluding some samples are more than 20% (over scale of measurement equipment). Gas concentration around the fumarole (UGG-1, 2, 3, 24, 25) and in the west (UGG-32), southeast (UGG-6, 19, 20, 21) is higher than those present in the eastern part. CO2 is also same tendency to Radon, for example, around the fumarole has the highest value in CO2 and low value in the easternmost part. High concentration of CO2 soil gas may be attributed to degassing from geothermal fluid that ascending through faults. Figure 12 displays CO2 value of points from the south to the north of the fumarole. This implies that from the south to the north the CO2 concentration increases.

Map of CO2 gas concentration interpret center of geothermal system that is around fumarole in Gedongsongo including valley in the east (high concentration of CO2) (Fig. 13). CO2 results also show a consistency with Radon map, that is, geothermal activities, fault zone trending North-South and it has boundary in the eastern part.

**Mercury gas**

In general, high Hg-air concentrations imply the presence of heat source at depth now. From Hg-soil air map (Fig 14), anomalies of mercury concentration are scattered distribution around Gedongsongo area. This presents center of geothermal system in this area. On the other hand, high concentrations imply that there is a heat source below this area (Unoki et al, 2003). Low Hg-air and high soil-Hg in UGG-6, 19, 20, 21 (Fig 15) interpret that source was once existed below the survey area and still present at depth. Besides, the low mercury values in the eastern part of this area suggest that heat source may only present in a relatively small area.

**CONCLUSIONS**

Preliminary interpretations of results are summarized as follow:

1. The type of thermal water in Gedongsongo area is divided into 2 distinct types: sulphate and bicarbonate water. Steam heated water or sulphate water are Ca- (Na)-Mg-SO4-HCO3 type in Gedongsongo area. Diluted bicarbonate water such as Ca-Mg-HCO3 and Na-(Ca)-HCO3-Cl type water form the second group in surrounding area e.g. Bumen, Banaran, Kendalisodo, Diwak, Derakan and Kaliulo.

2. Because of mixing thermal waters and most of water samples are plotted within ‘immature water’ field in Giggenbach (1988), it can not use geothermometry to estimate reservoir temperature of this geothermal system. However, silica geothermometers maybe give the minimum range of reservoir temperatures from of 140°C.

3. Based on isotope geochemistry, it is evident that the waters at Gedongsongo are quite dilute in comparison to most geothermal fluids. The hydrothermal systems within this area are predominantly meteoric in water origin. Moreover, the isotopic and chemical compositions indicate that the local meteoric water, the oceanic water contribute in varying proportions to the waters and chemicals in the thermal waters which are actively discharged...
from Kaliulo area.

4. Schematic hydrogeochemical model is proposed as shown in Fig 16 for the Ungaran geothermal system. It also displays location of upflow zone and the geothermal fluid in Gedongsongo is fed by the lateral flow from that main uprising zone. Then flowing down approximately 15 km, chloride cooler water is discharged.

5. The center of geothermal system and/or a subsurface heat source is around fumarole in Gedongsongo and the valley east to the fumarole. Besides, there is possible a fault zone trending North- South direction. This data also show a boundary that limited area of geothermal system not extending toward the eastern and western part.

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REFERENCES


### TABLE 1: Chemical analyses of water collected in the Gedongsongo area and its surrounding.

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<tr>
<th>Location</th>
<th>Code</th>
<th>Temp (°C)</th>
<th>pH</th>
<th>EC (μS/cm)</th>
<th>HCO₃⁻</th>
<th>F</th>
<th>Cl</th>
<th>NO₃⁻</th>
<th>SO₄²⁻</th>
<th>Na⁺</th>
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Note: Concentrations are in ppm.
* Well water.
** River water.
FIGURE 1a: Map of sampling location. 1b: Index of research location

FIGURE 2: Gold wire for Hg measurement in soil

FIGURE 3: Radon measurement in soil air (Shigeno, 1992).
FIGURE 4: Sterifil Aseptic System equipment.

FIGURE 5: Relative Cl, SO4 and HCO3 concentration (ppm) of waters in Gedongsongo area.

FIGURE 6: Na/1000-K/100-ÖMg ternary diagram modified for Giggenbach (1989).
**FIGURE 7:** dD vs d$^{18}$O composition of waters in Gedongsongo and its surrounding area. (American Gulf Coast, Southern Israel, Michigan and Alberta basin – Hoefs, 2004)

**FIGURE 8:** Deuterium and chloride composition of thermal waters derived from oceanic and connate water. Open and closed symbols present thermal and meteoric waters in Campi Flegrei, Ibusuki, and Green Tuff (Truesdell et al, 1980).
FIGURE 9: Concentration of Radon and Thoron

FIGURE 10: Cumulatively plot of Radon concentration

FIGURE 11: Preliminary map of Rn/Tn ratios
FIGURE 12: Concentration of CO2 gas

FIGURE 13: Preliminary map of CO2 gas concentration
FIGURE 14: Preliminary map of mercury content in soil air

FIGURE 15: Preliminary map of mercury content in soil
FIGURE 16: The schematic hydrogeochemical model of the Ungaran geothermal system