

Geochemical Modeling of Acid Fluid from Volcanic Geothermal Areas, Case Study of Indonesian Geothermal Prospects

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ABSTRACT

The Indonesian geothermal fields are mostly located at the high elevation of volcanic areas, which are situated at more than 1500 m high. They are indicated by a small release of SO₂, HCl, high H₂S and other minor H₂, O₂+Ar, CH₄, and NH₃ gases. High subsurface temperatures (220°C-300°C) of exploration and exploitation drillings, combined with results of geochemical, geological and geophysical studies, have been used to construct a geochemical model of the geothermal system of the area. The release of gas (SO₂) on the low part of the volcanic crater rim is a function of in-depth magma process, e.g., vapour melt separation during coming up generating magma. The existence of SO₂ indicates a high-temperature magmatic component, rising from underlying magma. On the other hand, the flank of this volcanic fumaroles has no SO₂ gas, high H₂S and CO₂. The existence of H₂S assumed a low-temperature gas presenting at the reservoir. The H₂, O₂+Ar, CH₄, and NH₃ gases probably indicate a secondary hydrothermal component slowly rising from a two-phase, saline brine vapour, and covering the magmatic system. The H₂ to CH₄ and NH₃ shows a decreasing in equilibration for individual species, interpreting slow rate varieties (CH₄ and NH₃) are formed at depth within the hydrothermal zones. The SO₂-H₂S controls the rapidly rising magmatic component. The H₂S and CH₄ seem to be stable in low-temperature rock conditions, particularly at the geothermal reservoir, outside of volcanic vents. Modelling of acidic conditions within the hydrothermal zones from geothermal prospects may be indicated by presenting of active volcanic gases, eq. The Sorik Marapi, Lahendong and Namora Ilangit volcanic vents. Volcanic vents indicate a high-temperature fluid interaction toward the surface, where SO₂ and H₂S react with rocks. However, deep drillings to the geothermal reservoir, which is located northern part of outside the Sorik Marapi volcano, represent neutral chloride fluids.

1.0 INTRODUCTION

Indonesia's geothermal exploration and exploitation prospects are mostly located at the high elevation of volcanic areas, which are situated at more than 1500 m above sea level (Fig.1). The areas have been carried out in the '70s to early '80s, which started with 30 MWe at Kamojang (West Java). Since then, developments have intensively carried out, producing approximately 807 MW in 1998 (Nasution and Sukhyar, 1998). However, in 1998 to the early 2000s, a crisis occurred, affecting production capacities stuck none. Started 2004, the intensive exploration and exploitation increase significantly, produce 1300 MWe (Nasution and Supriyanto, 2011). Right now, the production capacity produces approximately 2,058 MW, which are the last supported by the Ulubelu Unit 4 (55 MW), Karaha Unit 1 (30 MW), Lumut Balai 40 MW, and the Sorik Marapi Modular (45 MW).

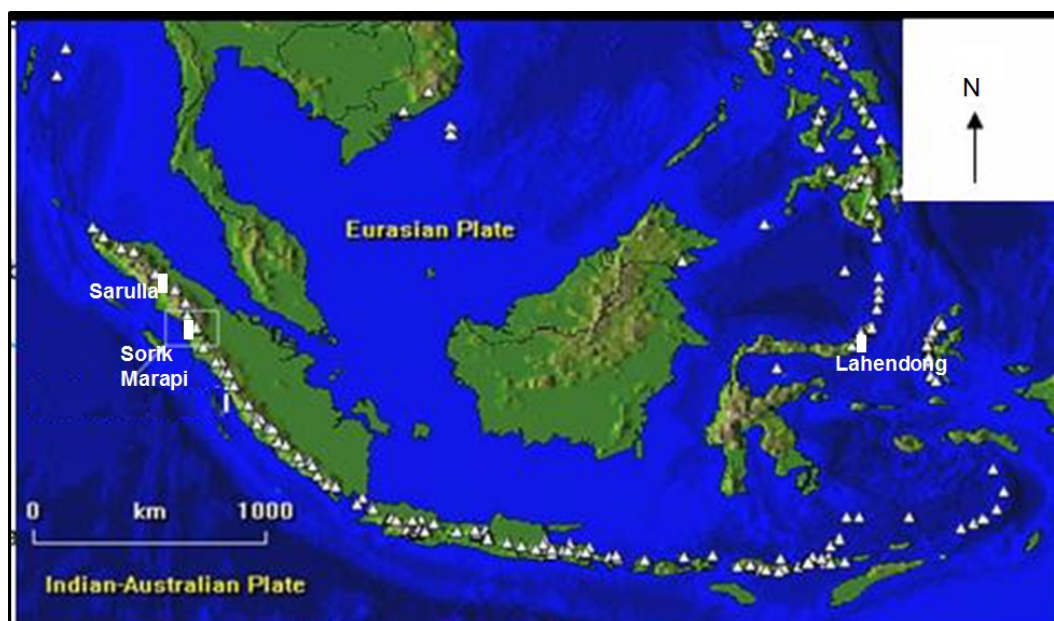


Fig. 1 Distribution of Geothermal Study areas of Indonesia

Some geothermal drillings have produced a low-pH Na-Cl-SO₄ fluid, such as the Namora-I-Langit, North Sumatra (Gunderson et al. 2000), Lahendong, particularly well no. 23 (Nasution and Assat, 2007) and the Sorik Marapi hydrothermal system (Nasution and Pramulyana, 2007). The effects of acid fluid on geothermal materials are corrosive installation materials. In contrast, to further from

volcanic vents, they appear more neutral pH Na-Cl fluid, which is represented by Silangkitang, Lahendong and Sibangor Tonga drilling fluids. These Na-Cl water distributed directly controlled by the Faults, rather than close to fractures of volcanic vent.

The last 25 years of studies in fluid geochemistry, particularly gas equilibria in hydrothermal environments, have been extensively investigated (e.g., Giggenbach, 1991; D'Amore and Panichi, 1980; Arnorsson et al., 1990; Chiodini 2017). The rising high-temperature gases under the Earth's surface could trigger the release of fluids and gases at a 10-fold increased rate. This would cause the injection of high-temperature steam into surrounding rocks (Chiodini et al. 2017).

This paper tries to describe chemical fluid characteristics, to discuss fluid flow and made acid fluid models, in order to hinder low-pH Na-Cl-SO₄ water during drilling.

2.0 METHODS

The chemical analysis of water composed of Na, K, Li, Ca, Mg, Fe, NH₄, As, HCO₃, Cl, SO₄, B, F and SiO₂. The analyses utilize methods of gravimetry-volumetry, electro-analysis (pH meter), and Spectro -analysis (spectrophotometry UV-visible, and photometry).

While the gases consist of 1. Non-condensable gas (H₂, O₂+Ar, N₂, CH₄, dan CO) and 2. The condensable gas in NaOH, such as SO₂, H₂S and S total. The first analytical methods utilize gas chromatography; the second analytical methods use gravimetry – volumetry. The other gases (HCl, HF, NH₃ and SO₂) use the spectrophotometry method.

3.0 WATER AND GAS CHEMISTRY

Water chemistry of acid fluids from study areas, are related to acid brines, which represented in three areas:

3.1 Lahendong field and its water chemistry

The Lahendong geothermal field is located in the northern arm of Sulawesi, in North Sulawesi Province (Fig.2). The arm consists of Lembeyan Ridge on the east and sedimentary environment on the west. The volcanoes of the NE Sulawesi area occur above the southern extension of the Minadanao-Sangihe-Minahasa intra-oceanic volcanic arc system with the subduction zone dipping to the NW under the Minahasa area. The oldest volcanic rocks in the area of Lahendong geothermal system are the upper Tertiary island arc tholeiites of the "Lambeyan Complex" (Fig.2).

The acid fluid of the Lahendong Geothermal field is shown by the surface manifestation of fumaroles, lake water and deep geothermal water of wells (Well no.23 and 28), as shown in Table 1. On the surface, the lake Linau acid water shows a pH of 2-3, located near acid hot springs. The acid fluids of production wells have been represented by LHD-23 and LHD-28, showing a pH of 2.7 to pH 3.2 (Table 1). They produce high SO₄, Cl and SiO₂ concentration, appearing in Table 1.

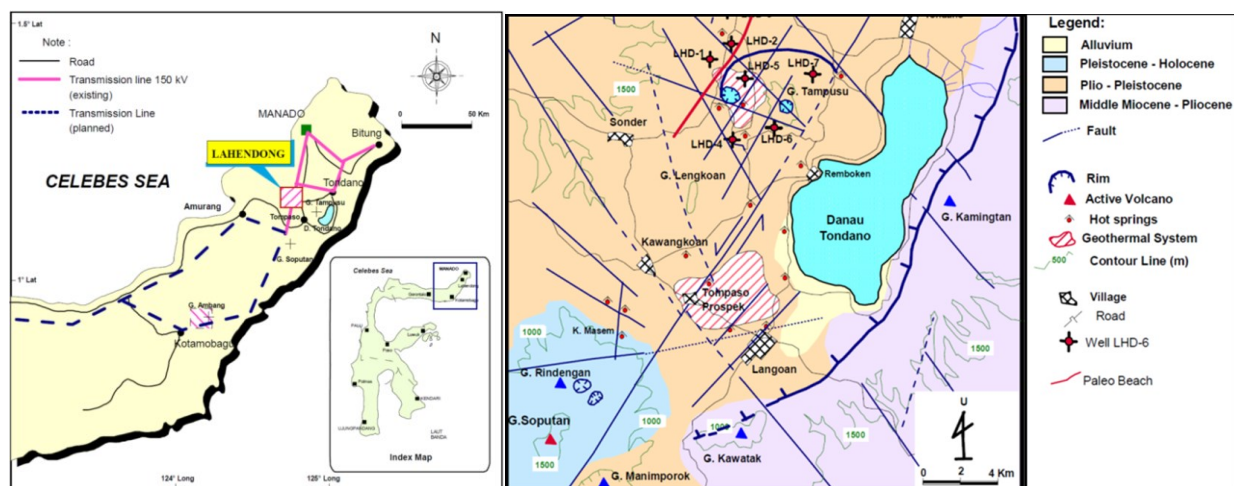


Fig. 2 The Lahendong Geothermal Field, showing the location and its development area (from Siahaan et al., 2005)

Table 1. Geochemical water of Condensate Lahendong Geothermal Field (Brehme et al. 2014)

No	Well number	Wellhead Elev.[m] asl	EC [!S/cm]	Ph	SO ₄ [mg/L]	Cl [mg/L]	SiO ₂ [mg/L]	RES-T [°C]	Water table [m] asl	TVD [m] bgl	Permeable zone from to [m] bgl
1.	23	879.2	9700	2.7	1609	1559	461	274	605	1693	1481 1693
2.	28	820.0	4620	3.2	523	991	364	200	620	1897	1750 1918

Lahendong, currently has developed a geothermal field with four units of 4×20 MW power plants, producing approximately 80 MWe. However, few wells produce acid fluid, with high production capacity. Consequently, recover the acid fluid to neutral fluid takes a high cost. Therefore, the future production drilling targets have to be considered not to go to acid fluid (low pH), particularly under the central volcanic vent or under a crater-lake.

3.1.2 The Sarulla Field and Namora I langit water chemistry

The Sarulla Geothermal Field is located in the Tapanuli Utara District, North Sumatra province (Fig. 1). The fields are evaluated as commercial geothermal systems within the Sarulla Block. They composed of Silangkitang, Namora I Langit, and Si Bual-Buali Prospects.

Namor I langit thermal features compose of fumaroles, boiling acid pools, boiling mud, acid sulfate springs, and acid sulfate alteration rocks. They show high temperatures, from boiling point to 119°C, a low pH and high SO₄ content (Table 2). One of the fumaroles represented repeatedly temperature 200°C (Gunderson et al., 1995). The wells drilled in this field represented highly productive wells, with sub-surface well temperatures between 260°C - 276°C. However, one of the productive wells (well no 24) produced a low-pH Na-Cl-SO₄ fluid (Ganefianto et al., 2015).

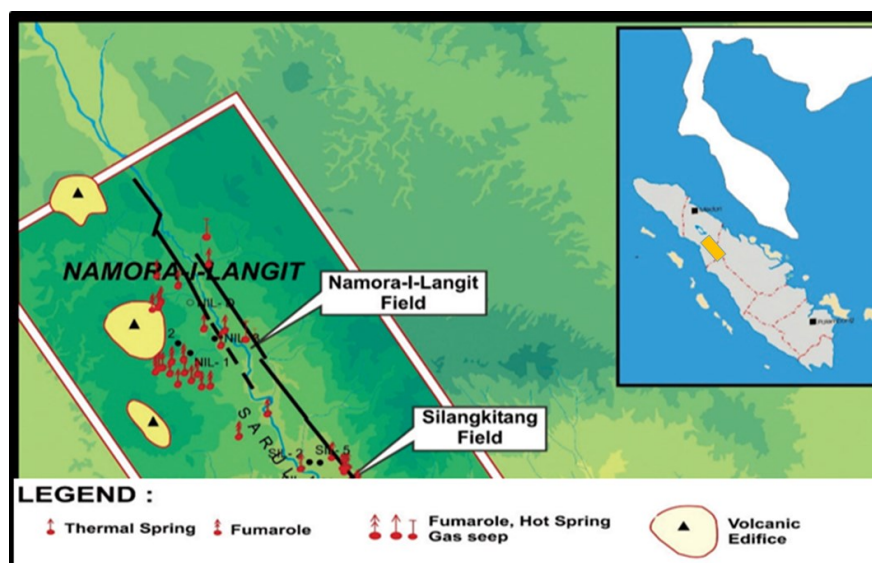


Fig. 3 The Namora I Langit Geothermal Field, which one acid well occur (from Ganefianto et.al.2015)

Table 2. Geochemical water of Namora I Langit (Gunderson et al. 2000)

W.Nil Acid			Chemical analyses of hot and warm water from fumaroles and crater lake in ppm										
	T°	pH	Li	Na	K	Ca	Mg	SiO ₂	B	Cl	SO ₄	HCO ₃	NH ₄
	97	3.09	0.05	27.96	34.72	20.14	5.30	344.7	0.24	7.10	256	0.00	0.3

3.1.3 Sorik Marapi and Water chemistry

The Sorik Marapi Geothermal field is located in Mandailing Natal Distrik, North Sumatra Province, geographical position 0°41'11,72" S and 99°32'13,09" E (Fig. 4).



Fig. 4 The Sorik Marapi Geothermal Field and its development area

Hot springs, fumaroles and solfataras of Mt. Sorik Marapi show temperatures between 34 – 276°C, distributing along with Sumatra fault structures and passing through Kotanopan-Panyabungan-Natal areas. Most thermal discharges are located between 500 - 2000 m a.s.l., having low pH (2–3.59), high SO₄ (353.85-1648.08ppm) and NH₃ (2-3.24 ppm) contents which represent SO₄ type water (Table.3). They indicate a high steam temperature containing volcanic gases (H₂S, SO₂, HCl and HF) condensing at the vadoze zone. RD-2 hot spring has a neutral pH and high HCO₃ concentration, representing a Bicarbonate type of water. It indicates that CO₂ reacts with shallow groundwater and silicate rocks below the surface, forming HCO₃ water. Ratios of Na/Ca (0.9) and Cl/HCO₃ are small, assuming a low shallow reservoir temperature.

High Boron concentrations (18.33-24.61ppm) of Crater Lake thermal discharges are interpreted as high reservoir temperature, dissolving sedimentary materials.

Table 3 Geochemical water of Sorik Marapi (Note: 1. Roburan Dolok, 2. Sibangor Tonga, 3. Sorik Marapi Crater Lake)

Location of geothermal areas	Chemical analyses of hot and warm water from fumaroles and crater lake in ppm												
	T°	pH	Li	Na	K	Ca	Mg	SiO ₂	B	Cl	SO ₄	HCO ₃	NH ₃
1. Hot spring (593m asl.)	98.9	2.52	0.05	8.64	0.5	8.26	2.48	33.15	0.00	355.18	353.85	0.00	2.26
2. Hot spring (840m asl.)	97.8	2.15	0.53	19.09	5.6	5.16	1.86	84.07	0.00	195.96	470.38	0.00	3.24
3. Crater water (2088m asl.)	34.1	1.10	1.10	14.55	10.0	11.35	5.82	72.59	18.33	244.95	1221.15	0.00	1.13

3.2 Gas chemistry

The released of gases from volcanic areas is a function of deep processes. They may act as vapor-melt separation during the generation and rise of the magmas. They also occur at the shallow processes and activities within the volcanic structures. The gases may be divided as follow:

3.2.1 The gases of Sorik Marapi volcanic crater rim

The Sorik marapi volcanic crater (2088 m asl.) shows a fumarolic field (1-B), representing a high-temperature vent on the surface, reaching 249°C. The fumaroles contain H₂O ± 82.54 %, and gases, which composing CO₂ 12,48 % mol, SO₂ 1,78% mol., H₂S 0,39 % and HCl 0,21% mol (Table 4).

Table 4. Geochemical Gases of Sorik Marapi

		Roburan	Sibanggor	Sorik Marapi
GASES	Unit	Fum-RD	Fum-ST	Sol 1-B
pH		< 3	< 3	< 3
Temp. °C		94.5	90	249
H ₂	% mol	0.020	0.001	0.001
O ₂ + Ar	% mol	0.010	0.001	0.010
N ₂	% mol	0.120	0.070	0.340
CH ₄	% mol	0.001	0.000	0.000
CO ₂	% mol	0.550	2.270	12.48
SO ₂	% mol	0.000	0.620	1.780
H ₂ S	% mol	0.160	0.080	0.390
HCl	% mol	0.000	0.090	0.210
NH ₃	% mol	0.140	0.230	2.260
HF	% mol	0.002	0.002	0.003
H ₂ O	% mol	99.01	96.62	82.54

3.2.2 The Gases of fumarole areas

The surface manifestations on the outer flank of the volcano to the north, show fumarole and a mud pool of Roburan Dolok (RD) and a fumarole field of Sibangor Tonga (ST), which located at high elevation (590-840 m asl.). They still yield high-temperature fumaroles between 90 – 94.5°C. The fumaroles contain high water content as steam, approximately 96 – 99 % mol. The gasses represent concentrations of SO₂ between 0.00 and 0.62 % mol, respectively. The volcanic gasses are HCl, HF, H₂S and other minor gases, such as (Table 4).

4.0 DISCUSSION

Geothermal fluids of three areas (Lahendong, Namora I langit and Sorik Marapi, geothermal fields) are meteoric and oceanic water origin. Although fluids in andesitic geothermal systems, near subduction areas, contain significant proportions of evolved connate and magmatic waters (Ármannsson and Fridriksson, 2009). The origin of geothermal waters is vital in geothermal studies. It helps in discriminating the chemical properties of the thermal waters and also their sources of recharge. Stable isotopes studies (mainly ²H and ¹⁸O), Helium (³He/⁴He), ³⁴S (SO₄) play an essential role in hydrogeological investigations of both thermal and non-thermal waters because the isotopes carry imprints of the origin of the waters.

The three development areas have been recognised as productive geothermal fields. However, closed to the main up-flow zones of volcanic areas, they show acid fluids geothermal system. Therefore, carefully drilling has to be done to hinder getting acid fluid from the geothermal reservoir.

4.1 Acid fluid of Lahendong (LHD) geothermal reservoir

1990's data showed the geophysical MT surveys of Lahendong. They were shown by low resistivity and high resistivity layering. The combination of resistivity values, geology and fluid geochemistry, represented an integrated model (Fig.5), which show a high-temperature fluid flow to the surface through production wells LHD-23 and LH-28, as represented by Table 1.

The number of wells represents the acid fluid of surface and deep wells from the Lahendong geothermal field. They are represented by the surface manifestation of fumaroles, lake water, and deep geothermal water of wells (Well no.23 and 28), as shown in table 1. The LHD-23 is a directional well towards the depth reservoir, which is nearly 2000 m below the Lahendong's crater (Fig.5). The depth water temperatures were shown by geo-thermometry and Well Head Temperature (WHT), ranging from 220°C to 340°C. The highest production capacity shown by LHD-23, showing 35 MW. However, it shows a low acid fluid pH 2-3.

Chemically, the LHD-23 fluid represents high SO_4 , Cl_2 , and SiO_2 concentrations, which are 1609 mg/L, 1559 mg/L and 461 mg/L respectively (Table 1). The low pH, high SO_4 , Cl and SiO_2 are probably caused by an introduction of volcanic fluids from a deeper level, such as hydrolysis of SO_2 ($4\text{SO}_2 + 4\text{H}_2\text{O} \rightarrow 3\text{H}_2\text{SO}_4 + \text{H}_2\text{S}$) (Truesdell, 1991). The high Cl content was probably caused by volatilization and transport of HCl in superheated steam ($\text{HCl}(\text{g}) \rightarrow \text{H}^+ + \text{Cl}$). While the highest SiO_2 concentration is probably caused by high solubility of SiO_2 in the high-temperature fluid of deep reservoir. An acid fluid model on Fig.5 represents them.

The results of geological and geophysical and geochemical studies of Lahendong (2007), the construction of a geochemical model of acid fluid flow from a geothermal system may be developed (Fig.5).

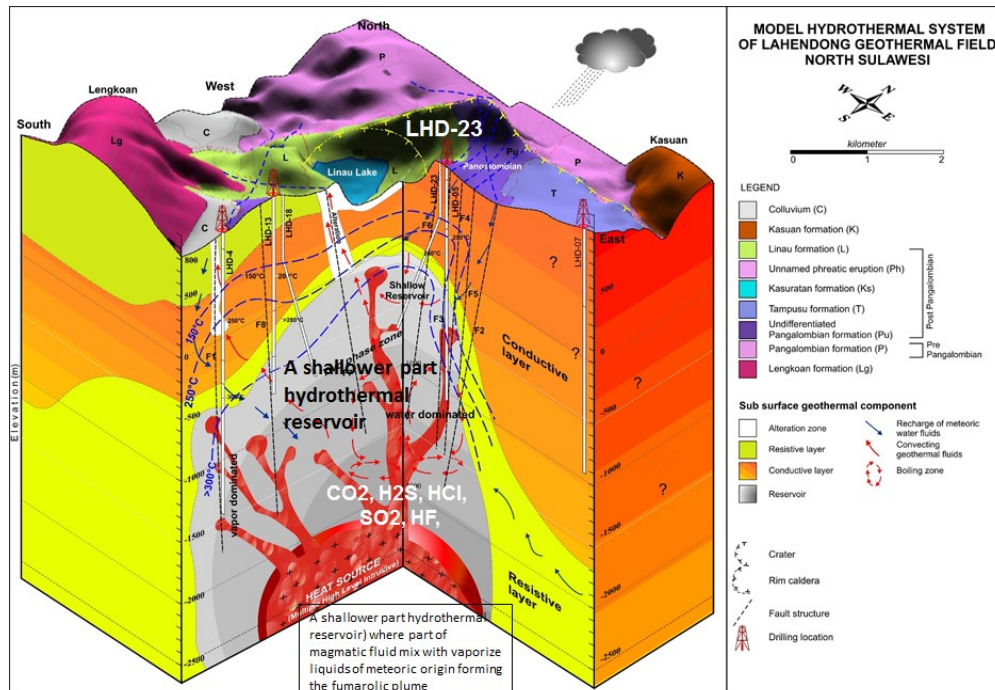


Fig. 5 the geochemical modelling of acid fluid of hydrothermal reservoir from Lahendong Field

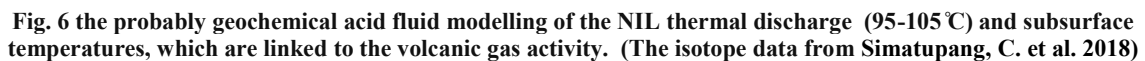
4.2 Acid fluid of Namora I Langit (NIL) geothermal reservoir

There are four wells drilled in the Namora-I-Langit (NIL) field. They were highly productive wells, encountering an extensive, high permeability geothermal system (Ganefianto et al. .2015). The wells all produced fluids with temperatures in excess of 260°C, with a maximum measured temperature of 276°C. Three of the wells produced neutral Na-Cl brine, but the fourth produced a low-pH, Na-Cl- SO_4 fluid. The permeability of the Namora I Langit system appears to be widely distributed and is not directly controlled by the Great Sumatra Fault. They were controlled by an inactive volcanic system of NIL.

The wellhead of NIL produced high Non-Condensable Gases (NCG) ranging from 6.0 – 9.0 wt.% with the highest one is shown by wells, close to the main crater of NIL. Meanwhile, relatively lower gas is shown by the further wells (Ganefianto et al. .2015).

The deep magmatic liquid probably supplies SO_2 below the crater of the NIL volcanic system, recognised from $\delta^{34}\text{S}$ isotope results. The less intensity of low resistivity allows a mixture of shallow and deep liquids. It results in the formation of SO_4 enriched liquid from SO_2 hydrolysis. The “heavy” (>15 ‰) dissolved sulfates are thought to be directly derived from SO_2 disproportionation at greater depth. The sulfates are typically found in thermal areas, which are sulfate derived from H_2S oxidation at or near the surface. Based on Bayon and Ferrer (2005) that sulfates with ratios > 15‰ are most likely produced by mixing of the deep, $\delta^{34}\text{S}$ (SO_4)-enriched fluid and the shallow, $\delta^{34}\text{S}$ (SO_4)-depleted water, such as in thermal features and steam condensates. The similar $\delta^{34}\text{S}$ values observed in all the sampled wells indicate that NIL SO_4 comes from a similar source. The modelling of acid fluid from NIL is shown in figure 6.

The other one that the SO_4/Cl ratios of NIL over 3, much closer to Sorik Marapi acid crater lake, indicating magmatic gases might contribute to the geothermal system. The values of $\delta^{34}\text{S}$ (SO_4) are large > 10‰, and they increase due to SO_4 concentration, probably a contribution magmatic SO_2 to the high-temperature brain water reservoir (Fig.6).



The Sorik Marapi Geothermal Field is located west and northwest of Sorik Marapi volcano, which is an active andesitic stratovolcano, on the western part of the Sumatra Fault Zone (SFZ). The volcano is located within the Panyabungan Graben.

Gaseous profiles of Sorik Marapi H₂O, H₂, CO₂, H₂S, SO₂, CH₄, He, N₂, and Ar molar concentrations are as a function of depth (Table 4). These gases represent the acidic conditions within the hydrothermal zones of fumarole, indicate an intensive fluid-rock interaction toward the surface, where H₂S intensively reacts with surrounding rocks. Based characteristic of gases, they indicate that there were high-temperature gases release through volcanic structures from a volcano with different surface temperatures. Gas ratios and geothermometry of steam discharge from the Mt. Sorik Marapi fumarole indicate steams are derived from a deep fluid source, with a temperature of 370 to over 400°C.

2. A shallower part hydrothermal Reservoir, where magmatic fluid mix and vaporize liquids of meteoric origin forming the solfatara vapour plume

1. A deep gas zone (volcanic gas), which is located at 4-5 km depth (Chiodini, 2017) and which supplies hot gases to the system

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The solfataric gases of Sorik Marapi are indicated by the release of SO₂, HCl, high H₂S and other minor H₂, O₂+Ar, CH₄, and NH₃ gases (Table 4). The existence of SO₂ indicates a high-temperature magmatic component, rising from underlying magma. The release of the gas (SO₂) on the low part of the volcanic crater rim is a function of deep magma process, e.g., vapour melt separation during coming up generating magma (Fig.7).

On the other hand, the flank of this volcanic fumaroles has no SO₂ gas, high H₂S and CO₂. The existence of H₂S assumed a low-temperature gas presenting at the reservoir. The H₂, O₂+Ar, CH₄, and NH₃ gases probably indicate a secondary hydrothermal component slowly rising from a two-phase, saline brine vapour, and covering the magmatic system (Fig.7). The exploration and exploitation drillings, to the northern flank of Sorik Marapi, at Sibangor Tonga area (Fig.4), represent high subsurface temperatures (220°C-300°C).

The Sibangor Tonga geothermal fluid has neural pH water at the Well Head (Fig.4). The H₂ gas becomes CH₄ and NH₃, which show a decreasing in equilibration for individual species, interpreting slow rate varieties (CH₄ and NH₃). They are formed at depth within the hydrothermal zones. The rapidly rising magmatic component is controlled by the SO₂-H₂S (Giggenbach, 1991). The H₂S and CH₄ seem to be stable in low-temperature rock conditions, particularly at the geothermal reservoir, outside of volcanic vents. Therefore, the boundary of acid fluid of volcanic vents to the neutral chloride brain is approximately 0.75 to 1 km in diameter.

Modelling of acidic conditions within the hydrothermal zones from geothermal prospects may be indicated by presenting of active volcanic gases, eq. Lahendong, Namora I langit, and the Sorik Marapi volcanic vents. The volcanic vents indicate a high-temperature fluid interaction toward the surface, where SO₂ and H₂S react with the rocks.

5.0 CONCLUSION

Modelling of acidic conditions within the hydrothermal zones from geothermal prospects may be indicated by presenting of active volcanic gases, eq. The Sorik Marapi, Lahendong and Namora Ilangit volcanic vents. Volcanic vents indicate a high-temperature fluid interaction toward the surface, where SO₂ and H₂S react with rocks. The existence of SO₂ indicates a high-temperature magmatic component, rising from underlying magma.

On the other hand, the flank of these volcanic fumaroles, particularly north flank of Sorik Marapi have no SO₂ gas, high H₂S, and CO₂. The existence of H₂S assumed a low-temperature gas presenting at the reservoir. The H₂, O₂+Ar, CH₄, and NH₃ gases probably indicate a secondary hydrothermal component slowly rising from a two-phase, saline brine vapor, and covering the magmatic system.

The acid geothermal waters (pH = 3-5) associated with high enthalpy fluids are discharged from the high-temperature reservoirs (over 350°C) in the Mt. Sorik Marapi hydrothermal systems. High Boron concentrations (18.33-24.61ppm) of Crater Lake thermal discharges are interpreted as a high reservoir fluid temperature dissolving sedimentary materials.

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