Indonesian Journal of Economic Geology (IJEG)
Volume I No. 2, September 2025: 80-90 e-ISSN 2809-1094

PARAGENETIC SEQUENCE OF ORE FORMATION IN INTERMEDIATE SULPHIDATION EPITHERMAL DEPOSIT: NEW INSIGHTS ON MINERALIZING PROCESSES AT KOTABUNAN, EAST BOLAANG MONGONDOW, NORTH SULAWESI

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Received Date: 30 January 2025, Accepted Date: 10 July 2025, Published: 30 September 2025

Abstract

The northern arm of Sulawesi is comparatively well mineralized with various types of deposits. One prospective area of mineralization is in Kotabunan, East Bolaang Mongondow Regency, North Sulawesi. This study aims to determine the geological conditions, hydrothermal alteration, mineralization style, and the sequence of ore mineral formation through paragenesis studies. Mineralization in the Kotabunan area is interesting to study because it provides quite representative evidence, with large amounts of ore mineralization exposed on the surface. The methods used are geological field mapping, thin sections, polished sections, XRF, and reflectance spectroscopy analysis. Based on geological mapping, the stratigraphy of the research area can be divided into 11 lithostratigraphic units and 11 faults, which are dominated by normal right-slip faults trending northwest-southeast and normal left-slip faults trending northeast-southwest. The alteration zones consist of quartz + chlorite + illite ± calcite alteration zones and quartz + illite + kaolinite ± sericite alteration zones. Mineralization in the research area occurs as veins and disseminations within rocks. The presence of mineralization in the form of veins is marked by the emergence of base metal veins controlled by northwest-southeast trending faults and indicates mineralization of the intermediate sulfidation epithermal (ISE) system. The results of polished section analysis show that the presence of disseminated mineralization is dominated by pyrite. In base metal vein mineralization, pyrite is present throughout the hypogene phase and is later replaced by galena. Then, galena is overprinted by intergrowth sphalerite. Tennantite occurs as an inclusion within sphalerite. Chalcopyrite is also present in veins as a replacement for sphalerite, galena, and pyrite in the form of disease texture. The presence of gold is also observed in veins that occur together with galena, as evidenced by the presence of gold in the same groundmass as galena and sphalerite. Then covellite replaces pyrite, indicating a supergene process, and hematite forms, ending the mineralization process. The results of this study have significant implications for understanding the ore-forming processes of ISE deposits in the Kotabunan District, East Bolaang Mongondow Regency, North Sulawesi.

Keywords: Alteration, Epithermal, Kotabunan, Mineralization, Paragen

Introduction

Gold is one of the ore minerals formed through the mineralization process. Mineralization occurs due to hydrothermal solutions interacting with host rocks. This interaction induces alteration processes that change the physical and chemical properties of the minerals comprising the host rocks (Pirajno, 2009). Alteration and mineralization processes typically occur over long periods and progress gradually, a phenomenon known as paragenesis. In Indonesia, one of the areas with gold mineralization prospects is Kotabunan, East Bolaang Mongondow, located within the Mining Business Permit (IUP) area of PT Arafura Surya Alam. This location is reachable with four wheel or two wheel vehicle, about 110 km south of Manado City.

The Kotabunan Prospect is one of several prospects in the northern arm of Sulawesi. Exploration and small-scale mining of gold deposits in the north arm of Sulawesi have been carried out since the Dutch era (1896 – 1931) (van Leuwen and Pieters, 2011). So far, studies conducted in this area have been limited to geological and alteration mapping related to mineralization. On the other hand, studies focusing on the paragenesis of mineralization, which characterize the sequence and processes of ore mineral formation, remain scarce. Therefore, a specific study on the sequence and formation processes of alteration and mineralization minerals in this area is needed. This research will provide a detailed understanding of the sequence of mineral formation that constitutes the mineral deposit system in the study area.

Research Methods

In this study, rock and vein samples were collected from outcrops in the research area. The samples were collected as hand samples using the chip sampling method at the outcrop. The secondary minerals in the research area were examined through thin section and polished section analyses (25 samples), spectrometer analysis (31 samples), and X-ray fluorescence (XRF) analysis to determine the valuable mineral content (19 samples). Thin section and polished section analyses were conducted at the Department of Geological Engineering, Bandung Institute of Technology. Spectrometer analysis was carried out at PT Arafura Surya Alam using Specterra 4.0 to detect the presence of secondary minerals, including clay and oxide minerals. This analysis is performed by irradiating rock samples with a specific wavelength. The XRF analysis was performed at PT Arafura Surya Alam using an Olympus XRF Analyzer to determine the geochemical content, particularly the valuable elements in the rocks, by providing X-rays on rock samples.

Geological Setting

Regional Geology

Northern Sulawesi is a Neogene volcanic arc overlain by Paleogene sedimentary-volcanic bedrock and possibly overlain by oceanic crust (Kavalieris et al., 1992). The tectonic development of Sulawesi is influenced by the complex convergent zone between the Eurasian, Pacific and Australian Plates. The main event of the three plates connection was the collision of the Banggai-Sula with the Sulawesi arc in the middle Miocene (Kavalieris et al., 1992), which resulted in a

clockwise rotation of the arc from a north-south orientation to a west - east orientation (Carlile et al., 1990; Kavalieris et al., 1992; Turner et al., 19

The northern arm of Sulawesi was also subducted in a westerly direction by the Molucca Sea Plate to the east of the northern arm of Sulawesi 10 million years ago (Hall, 2011). These tectonic interactions led to the development of a large structure in the North Arm of Sulawesi with a dominant direction of relatively northwest - southeast and partly northeast - southwest (Koesoemadinata, 2020). The development of regional structures on the North Sulawesi arm is influenced by regional compression caused by North Sulawesi subduction as well as East Sangihe subduction. The compression resulted in a northwest-southeast trending fault that formed a dilational jog that became the permeability pathway of the intrusions in the North Sulawesi Arc.

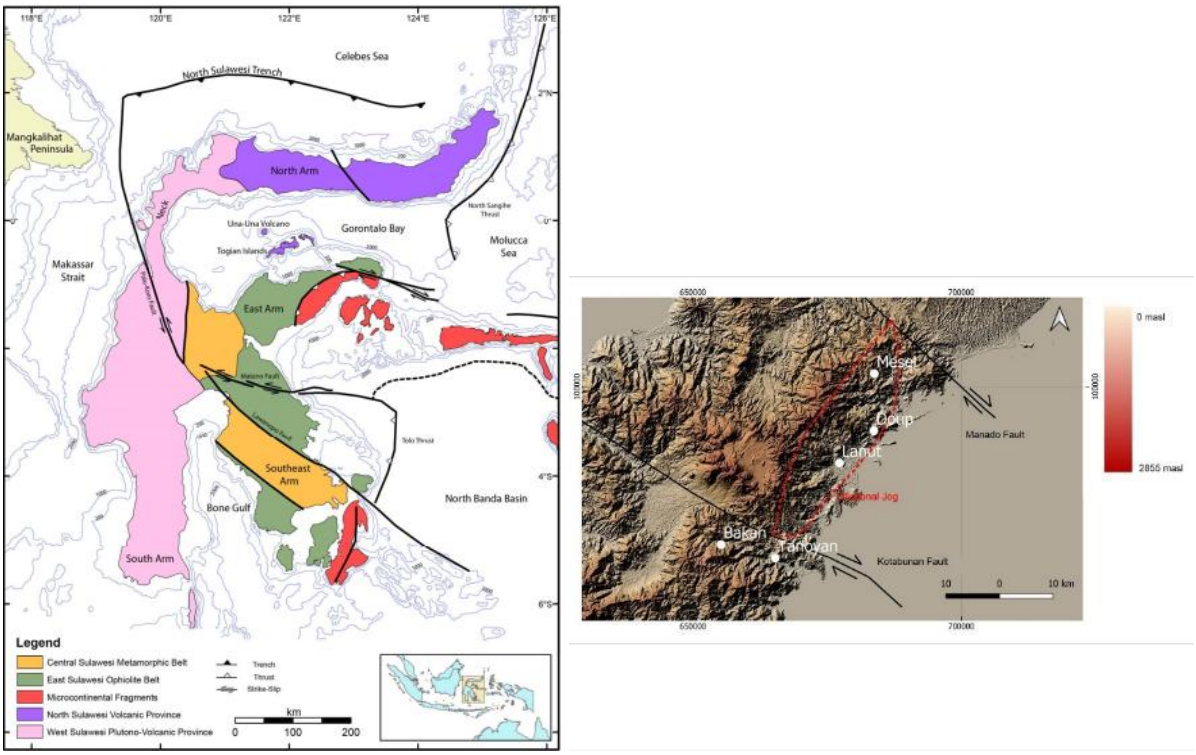


Figure 1. Principal tectonic subdivisions of Sulawesi (Pholbud et al., 2012)

The results of this tectonic activity are then grouped into five tectonic provinces, namely Western Sulawesi Plutono-volcanic Province, North Sulawesi Volcanic Province, Central Sulawesi Metamorphic Belt, East Sulawesi Ophiolite, and Microcontinental fragments of Australian origin (Pholbud et al., 2012). The study area belongs to the North Sulawesi Volcanic Province, which is composed of volcanic rocks with tholeiitic composition (Pholbud et al., 2012). Carlile et al. (1990) divided the northern Sulawesi arm into four geological sections: the Marisa Section, Gorontalo Section, Minahasa Section, and Sangihe Section. The westernmost part is called the Marisa Section, consisting of basalt and sedimentary rocks that are unconformably overlain by metamorphic bedrock. Furthermore, there is the Gorontalo section formed by Miocene-andesitic volcanic arcs with some diorite intrusions. This arm turns to the northeast in the form of the Minahasa section, which consists of Miocene-aged volcanic andesite that is mostly

covered by Pliocene-Recent volcanic deposits (Carlile et al., 1990). The volcanic arc continues northward through the Sangihe Islands, comprising the active volcanism of the Sangihe Section.

The study area is located at the western end of Section Minahasa. Sulawesi consists of highland areas with an average altitude below 2000 m

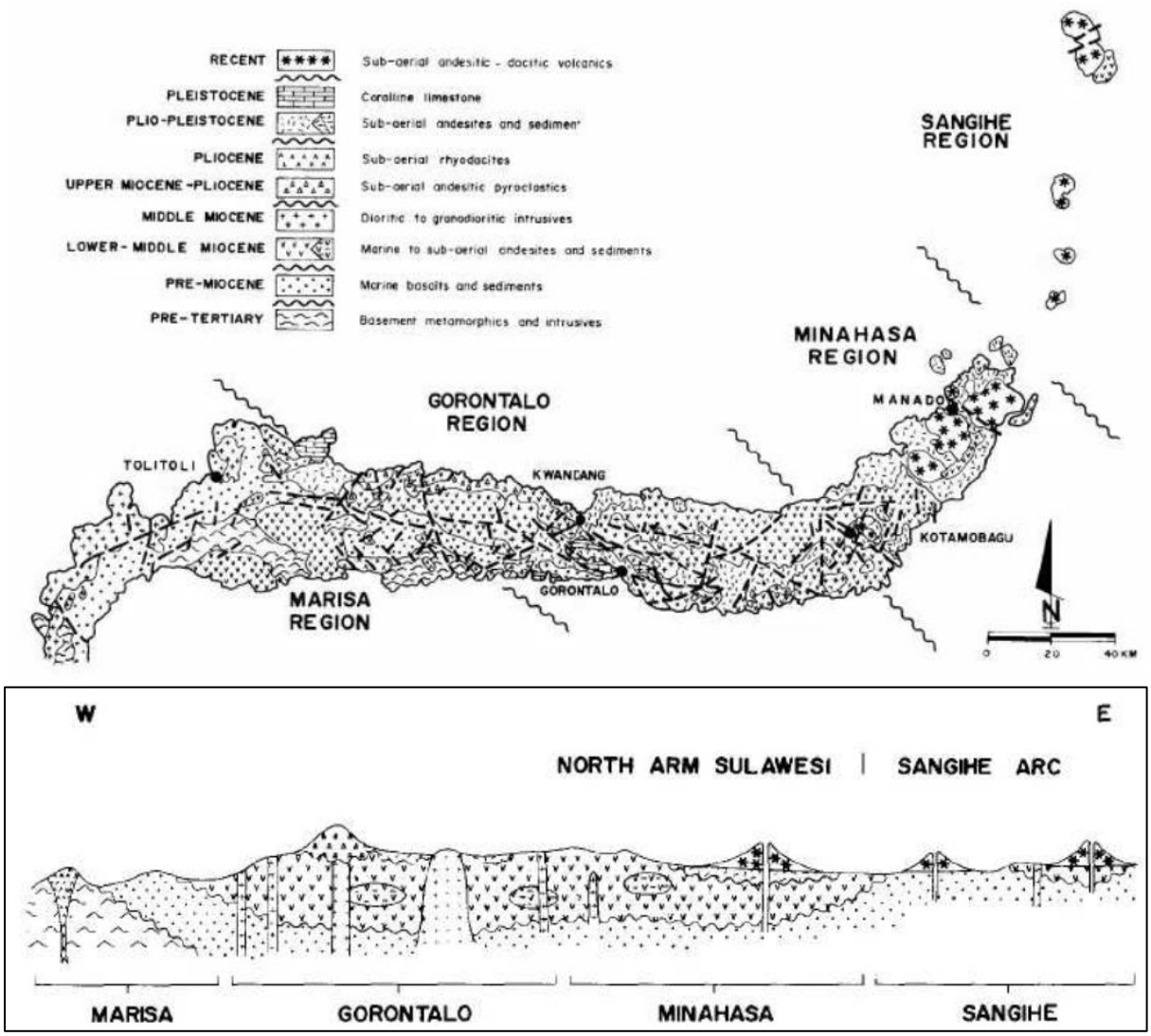


Figure 2. The geologic division of the North Arm of Sulawesi, as schematically shown by Carlile et al. (1990)

Geology of the Kotabunan Area

The study area is divided into several rock units based on an informal lithostratigraphic classification that groups units by similarities in descriptive characteristics and formation processes. Microscopic and macroscopic observations were conducted on rock samples from each

unit to obtain descriptive characteristics through mineral composition and rock texture. Based on this analysis, the stratigraphy of the study area consists of eleven lithological units.

This lithologic unit consists of igneous, sedimentary and volcanoclastic rocks. The igneous rocks in the study area consist of intrusions and lava units. These units, from oldest to youngest, are as follows: limestone unit, sandstone-limestone unit, andesite intrusion 1 unit, quartz

diorite unit, tonalite unit, claystone-sandstone unit, sandstone-conglomerate unit, tuff-epiclastic unit, dacite lava unit, andesite intrusion 2 unit, and alluvium-colluvium unit (Figure 3).

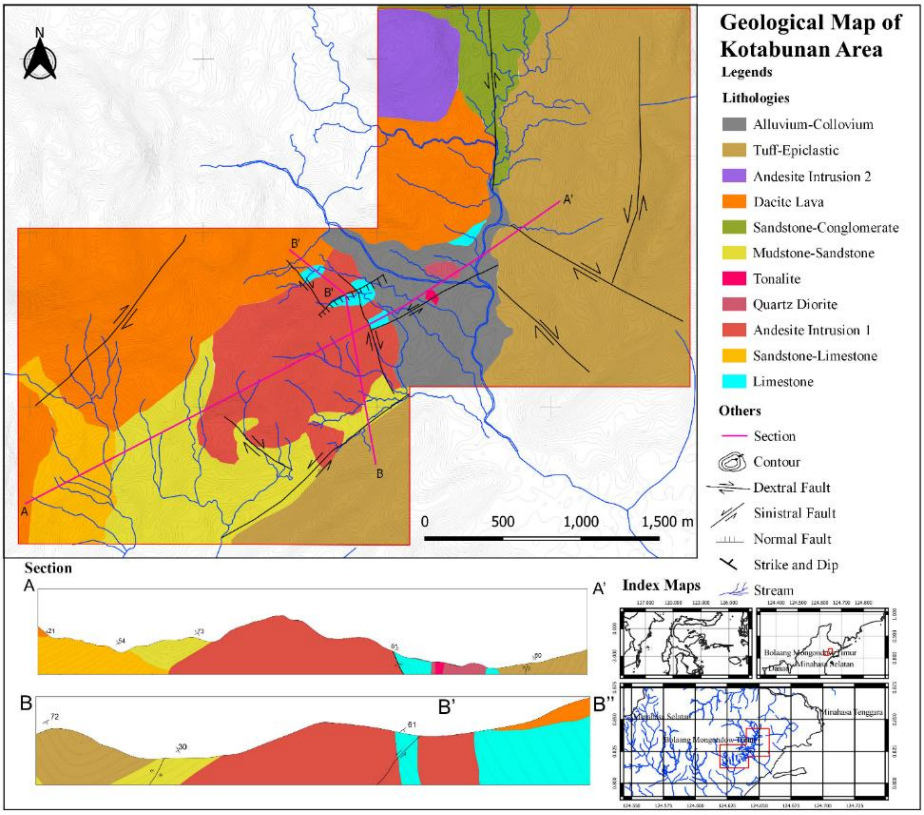


Figure 3. Geological map of the Kotabunan area

The stratigraphy proposed by Turner et al. (1994), which provides a general stratigraphy of the Mesel area (northeast of Kotabunan), will be used as a reference for formation correlation to estimate the age range of the rock units in the study area (Figure 4). The oldest rocks in the study area are Miocene age sedimentary rocks. The andesite intrusion 1 unit, which is not associated with the ore mineralization (pre-mineralization), occurred before the tonalite formation, which is responsible for ISE mineralization in the Kotabunan Area (syn-mineralization). Volcanic products, as well as hydrothermal activity, occurred from the Pliocene to Pleistocene.

The geological structures of the study area were identified using a combination of lineaments derived from DEM data and field observations. Eleven faults were identified in the study area, dominated by northwest-southeast trending dextral-normal faults, northeast-southwest

trending sinistral-normal faults, and north-south trending sinistral faults. Based on observations, the distribution of alteration and mineralization in the study area is controlled by northwest-southeast trending dextral-normal faults.

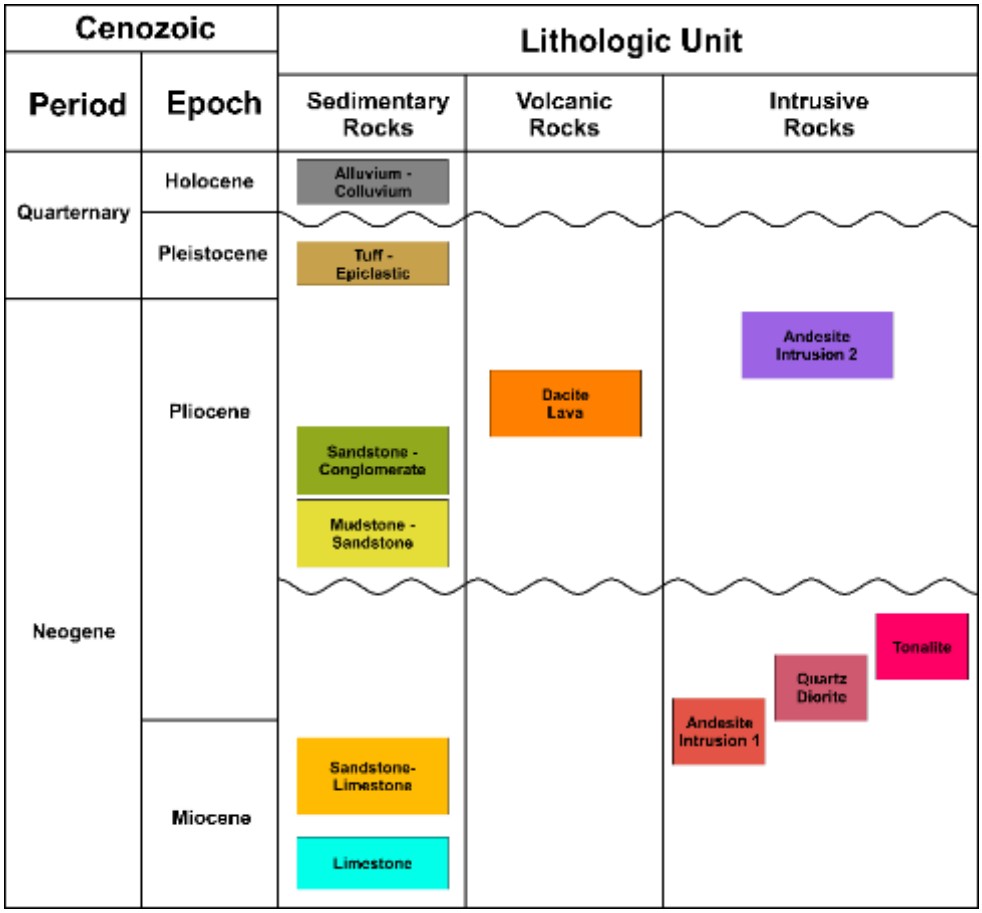


Figure 4. Lithostratigraphic Column of the Kotabunan area

Result and Discussion

Hydrothermal Alteration

Alteration observations in the research area were conducted using macroscopic observations, ASD spectral analysis of surface rock samples, and microscopic analysis of thin sections from these samples. The results indicate the presence of alteration zones and their associated alteration minerals in the research area, which include the quartz + illite + kaolinite ± sericite alteration zone and the quartz + chlorite + illite + calcite + montmorillonite alteration zone. The major structures in the research area are dominated by a NW-SE orientation, cutting across the altered zone within the area. The first structure to appear in the research area is a NW-SE-trending structure that represents a dextral fault and generally controls the distribution of the alteration zone. This structure is subsequently intersected by a NE-SW trending structure, consisting of normal faults

and sinistral strike-slip faults. The alteration in the research area occurs within quartz diorite, tonalite, and andesite 1 units.

The presence of quartz + illite + kaolinite ± sericite alteration, especially in tonalite rocks, is accompanied by the occurrence of veins filling fractures within the rocks. Quartz diorite is

characterized by white-colored rocks with an allotriomorphic granular texture, featuring phenocrysts such as sericite, opaque minerals, primary quartz, and hornblende. The groundmass consists of secondary quartz and clay minerals, with percentages ranging from 40-50%. On the

other hand, tonalite in the research area is characterized by phenocrysts dominated by secondary minerals, including secondary quartz, opaque minerals, sericite, and primary quartz embedded within a groundmass of clay minerals and secondary quartz.

In the andesite 1 intrusion unit, the groundmass is composed of clay minerals, while phenocrysts include plagioclase, hornblende, quartz, and chloritized minerals. Additionally, there is a limestone lithology characterized as packstone, dominated by skeletal fragments altered by clay minerals. These four lithologies are believed to host mineralization within an intermediate sulfidation epithermal system, with tonalite and quartz diorite lithologies undergoing the most extensive alteration. These rocks are characterized by their white colour, resulting from the intense alteration of primary minerals into clay minerals and sericite, which replace the original minerals in both lithologies.

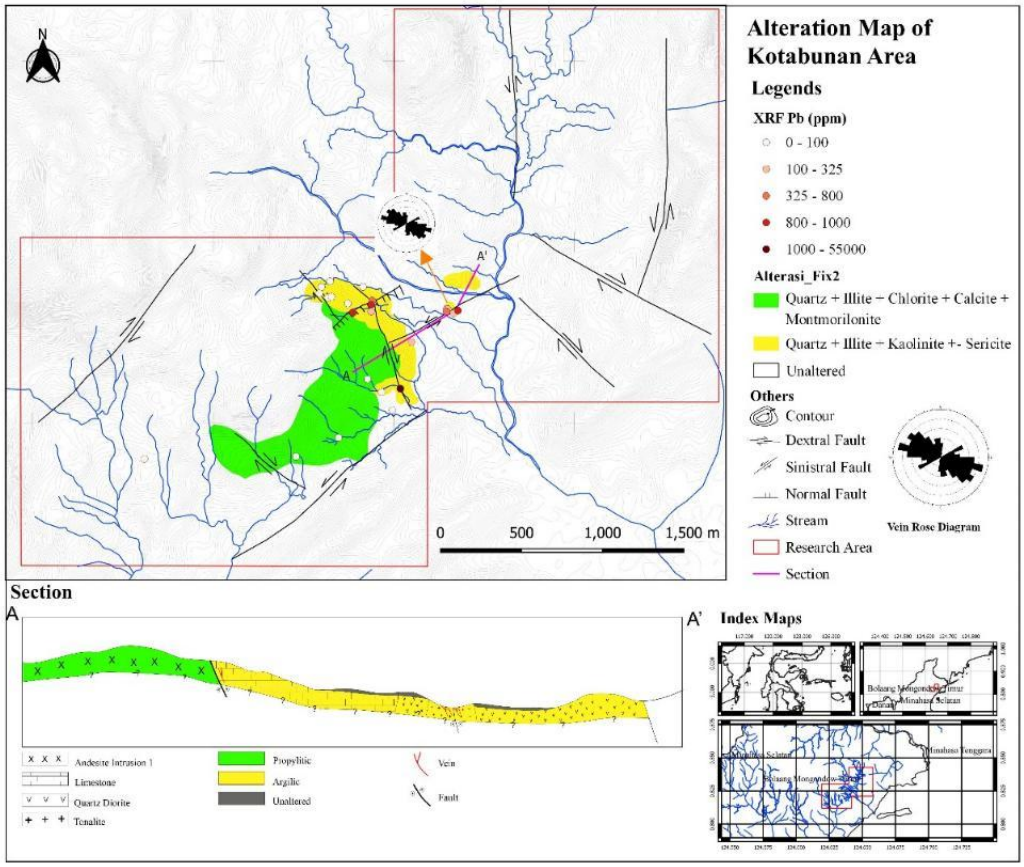


Figure 5. Alteration zone map of the Kotabunan area

The quartz + illite + kaolinite ± sericite alteration is distributed across tonalite, quartz diorite, and limestone lithologies. The alteration in tonalite and quartz diorite exhibits a highly intensive degree, replacing up to 80% of the primary minerals in the rocks. In contrast, limestone alteration is less extensive, with some samples showing only weak alteration. The quartz + chlorite + illite ± calcite alteration is found in andesite 1 lithology with weak to moderate intensity, replacing about 30%–40% of the primary minerals. Based on ASD analysis, the alteration in limestone is not highly intensive, showing moderate alteration levels that replace up to 50% of the rock.

From the ASD analysis of several samples, kaolinite, illite, and hematite are observable. Thin section analysis further confirms the presence of secondary quartz and sericite, which replace the groundmass and phenocrysts within the rocks (Figure 6A). Additionally, chlorite-illite-montmorillonite minerals are observed, as evidenced by thin section analysis showing phenocrysts replaced by these minerals (Figure 6B). Its formation is influenced by the circulation of hydrothermal fluids and their interaction with meteoric water. Permeability in structures near heat sources can cause fluids to intensively alter the wall rocks, leading to high-temperature clay alteration. This process also forms ore minerals in the structural zone in the form of vein/lets. Away from heat sources, chlorite mineral-rich alterations are formed. Interaction with meteoric water causes the formation of low-temperature clay minerals.

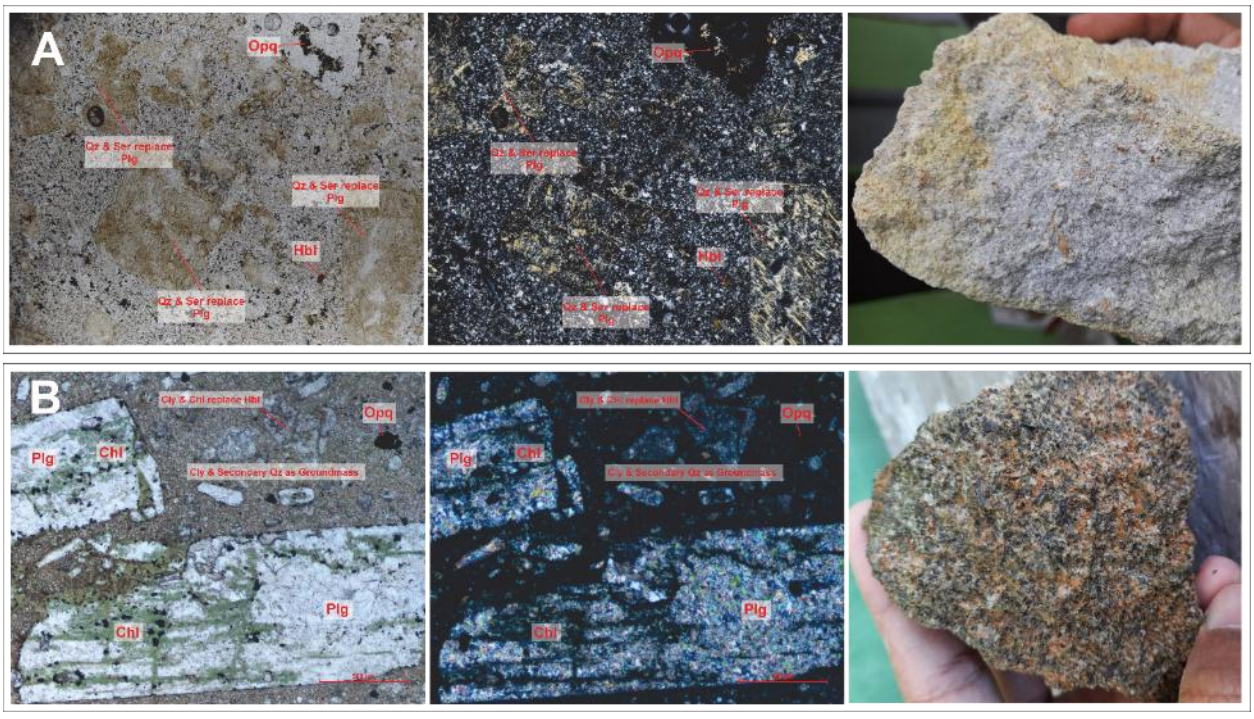


Figure 6. A) Parallel nicols thin section photomicrograph, cross nicols thin section photomicrograph, and hand sample specimen of quartz + illite + kaolinite ± sericite altered andesite rock; B) Parallel nicols thin section photomicrograph, cross nicols thin section photomicrograph, and hand sample specimen of quartz + chlorite + illite + calcite + montmorillonite altered andesite rock.

Mineralization Style and Ore Paragenesis

The mineralization present in the study area is mostly present in the form of basemetal veins. The presence of ore minerals in these veins tends to be dominated by minerals such as pyrite, galena, sphalerite, chalcopyrite, covellite, hematite, and goethite. The basemetal vein mineralization is present in tonalite and quartz diorite host rocks at quartz+illite+kaolinite+sericite alteration. In addition, disseminated pyrite mineralization is present in all alteration types. The presence of ore mineralization in this basemetal vein will then be subjected to paragenesis analysis to determine the sequence of mineralization formation. This type of mineralization strongly indicates intermediate sulfidation epithermal deposits (Wang et al., 2009). Most mineralization in the study area is associated with the presence of some sulphides such as chalcopyrite, pyrite, tenntite, galena, and sphalerite. Based on the results of x-ray fluorescence analysis of surface samples, mineralization in the Kotabunan area contains 2 – 471 ppm Ag, 0.008 – 5.49% Pb, and 0.003 – 31.65% Zn.

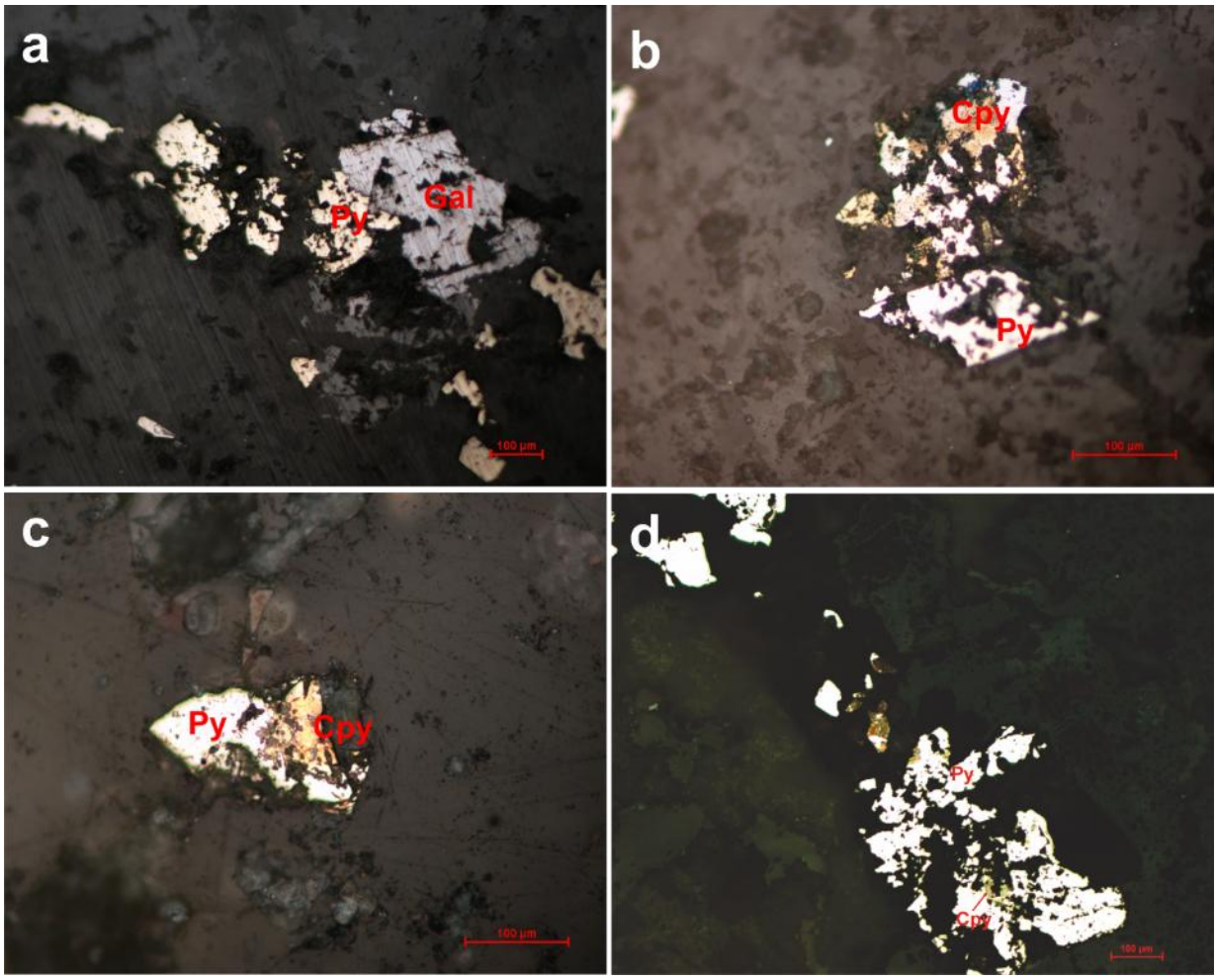


Figure 7. a) Polished section photomicrograph of sample ST-0001 at 10x magnification; b) Polished section photomicrograph of sample ST-0001 at 20x magnification; c) Polished section photomicrograph of sample ST-0002 at 20x magnification; d) Polished section photomicrograph

of sample ST-0001 at 10x magnification. Mineral abbreviation: Py= pyrite, Cpy= Chalcopyrite, Gal= Galena, Sph= Sphalerite, Cov= Covellite, Ten= Tennantite, Hem= Hematite

Paragenesis in mineralized ores is determined based on the relationship between minerals in the polished section of each sample. The relationship is then analyzed to obtain the order of formation of ore minerals based on overprints and inclusions in the minerals. On sample ST-0001, the polish section photomicrograph (Figure 7a) shows subhedral galena growing alongside pyrite marked with a firm boundary. A 20x magnification photomicrograph (Figure 7b) shows the chalcopyrite mineral replacing the pyrite mineral. This indicates that pyrite minerals are present before chalcopyrite minerals in these samples. Furthermore, the polished section photomicrograph at ST-0002 and ST-0002B (Figure 7c and 7d) shows that the pyrite mineral (Py) is starting to be replaced by the chalcopyrite (Cpy).

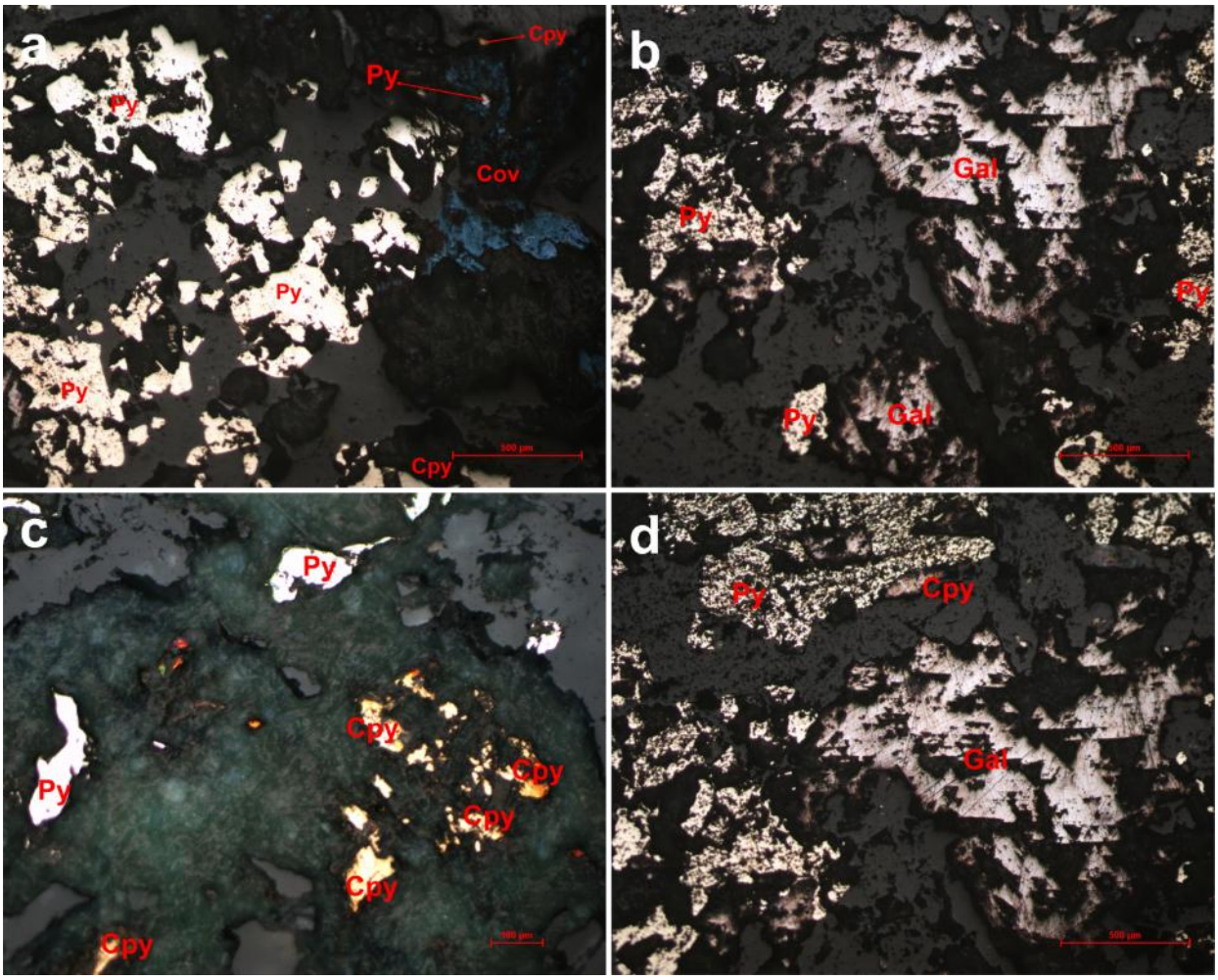


Figure 8. a,b) Polished section photomicrograph of sample ST-0003 at 5x magnification; c) Polished section photomicrograph of sample ST-0003 at 10x magnification; d) Polished section photomicrograph of sample ST-0003 at 5x magnification. Mineral abbreviation: Py= pyrite, Cpy= Chalcopyrite, Gal= Galena, Sph= Sphalerite, Cov= Covellite, Ten= Tennantite, Hem= Hematite.

On sample ST-0003, a 5x magnification photomicrograph (Figure 8a) shows that the pyrite mineral (Py) is abundant, with covellite mineral (Cov) present to replace the chalcopyrite (Cpy). Then, the photomicrograph (Figure 8b) shows the pyrite mineral (Py), which tends to be present along with galena (Gal). A 10x photomicrograph (Figure 8c) shows the pyrite (Py) present with chalcopyrite (Cpy). The photomicrograph (figure 8d) shows pyrite (Py), chalcopyrite (Cpy), and galena (Gal), with the chalcopyrite mineral present after the pyrite mineral.

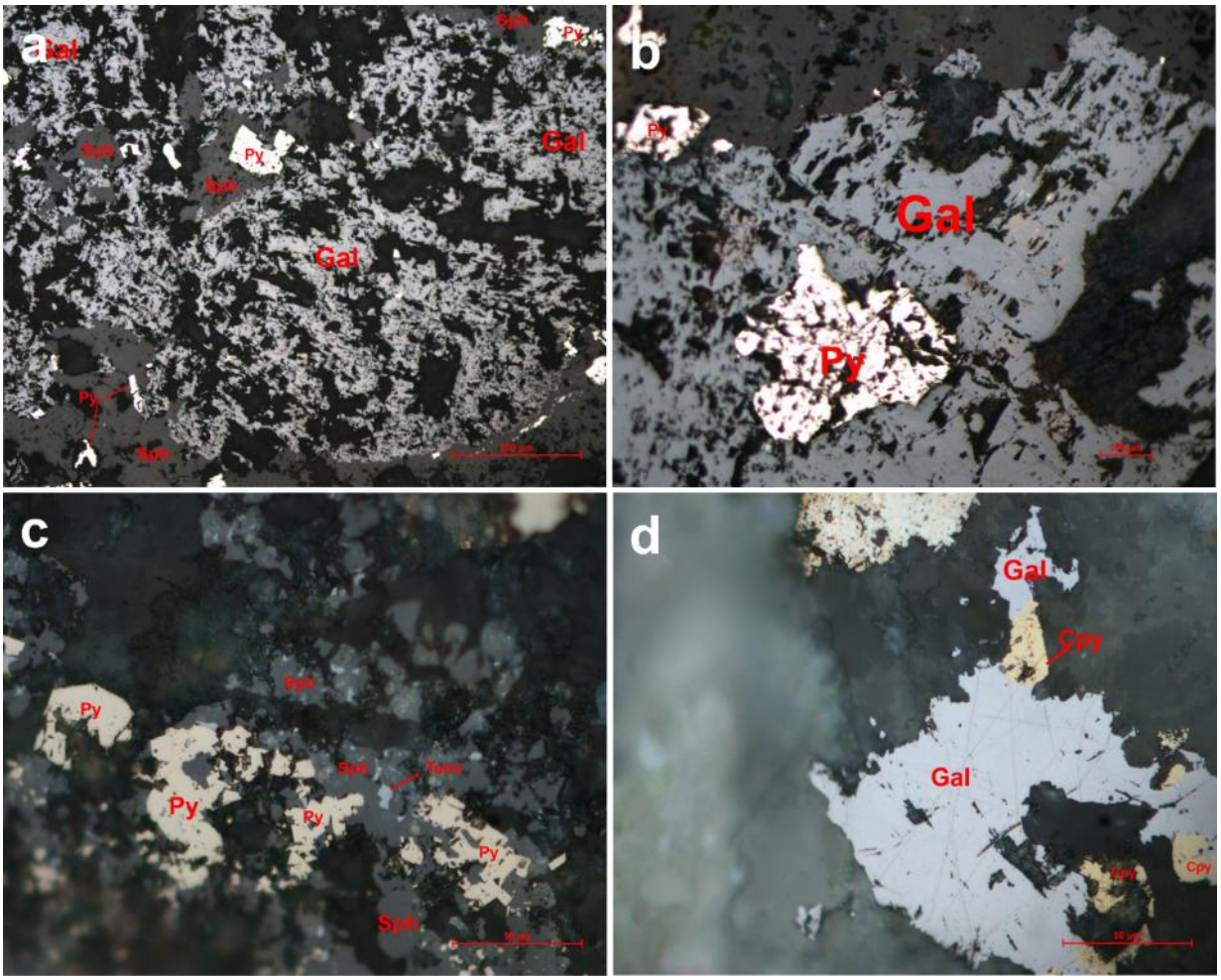


Figure 9. a) Polished section photomicrograph of sample ST-0004 at 5x magnification; b) Polished section photomicrograph of sample ST-0004 at 10x magnification; c, d) Polished section photomicrograph of sample ST-0005 at 50x magnification. Mineral abbreviation: Py= pyrite, Cpy= Chalcopyrite, Gal= Galena, Sph= Sphalerite, Cov= Covellite, Ten= Tennantite, Hem= Hematite.

On sample ST-0004, a 5x magnification photomicrograph (Figure 9a) shows the pyrite (Py) overprinting the sphalerite (Sph) mineral. In addition, it appears that the sphalerite mineral (Sph) present overprints the deformed galena (Gal). A 10x magnification photomicrograph (Figure 9b) shows the pyrite (Py) as an overprint in the galena (Gal). Furthermore, the ST-0005 sample, a 50x magnification photomicrograph (Figure 9c), shows chalcopyrite (Cpy) beginning to replace galena (Gal). Then, photomicrographs (Figure 9d) show the mineral sphalerite (Sph)

replacingpyrite (Py). It is seen that the existence of tenantite minerals (Tenn) that are present overprint in sphalerite minerals (Sph).

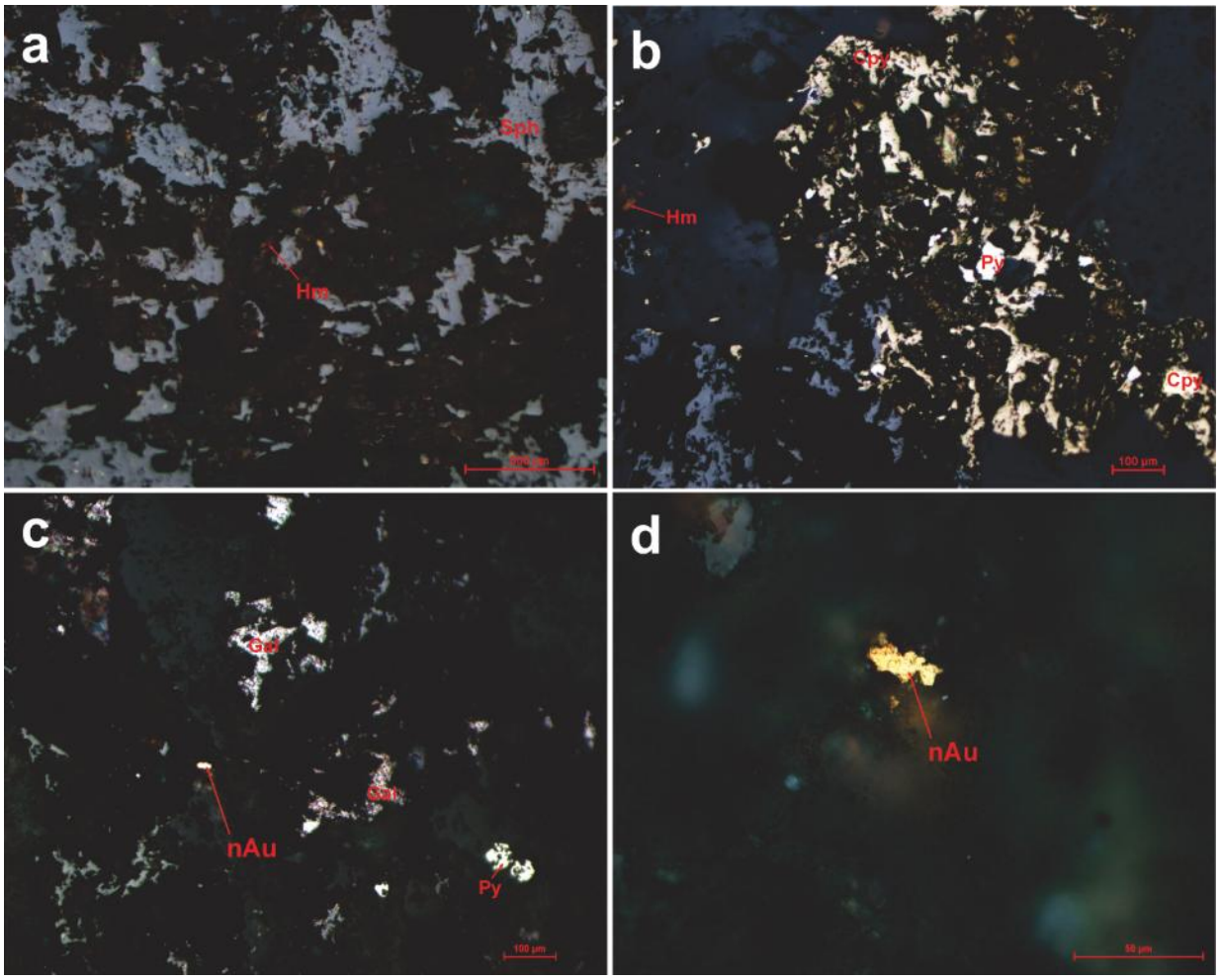


Figure 10. a) Polished section photomicrograph of sample ST-0006 at 5x magnification; b,c) Polished section photomicrograph of sample ST-0007 at 10x magnification; d) Polished section photomicrograph of sample ST-0007 at 50x magnification. Mineral abbreviation: Py= pyrite, Cpy= Chalcopyrite, Gal= Galena, Sph= Sphalerite, Cov= Covellite, Ten= Tennantite, Hem= Hematite.

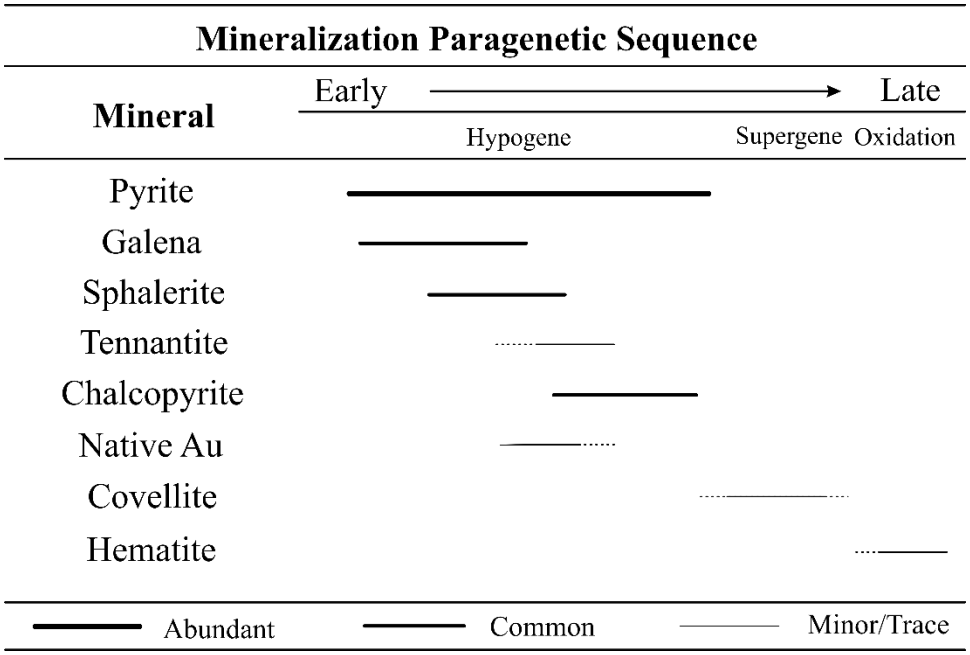
On sample ST-0006, a 5x magnification photomicrograph (Figure 10a) show the presence of hematite minerals (Hm) that replace sphalerite minerals (Sph). While in the ST-0007 sample, a 10x magnification photomicrographs (Figure 10b) show the presence of chalcopyrite (Cpy), which is replaced by pyrite (Py). Pyrite (Py) is present along with sphalerite, which is replaced by hematite (Hm). Sample ST-0007 also shows that there is gold present in the sphalerite (Sph) - galena (Gal) - pyrite (Py) vein. The gold is present as a free grain with a size of about 25

μm, which tends to be present almost at the same time as galena (Gal) and pyrite (Py) (Figures 7c and 7d).

Next, the paragenesis analysis of all samples was combined. This combination is intended to determine the sequence of ore mineral formation, arranged from early to late stages. (Table 1). Pyrite is present throughout the hypogene phase in base metal vein mineralization and is then replaced by galena. Then, galena is overprinted by intergrowth observed in veins that occur together with sphalerite, replacing the pyrite, which indicates the supergene process, and then hematite is formed, which ends the mineralization process of sphalerite. Tennantite occurs as an inclusion within sphalerite. Chalcopyrite is also present in veins as a replacement for sphalerite, galena, and pyrite in the form of disease texture. The presence of gold is also observed in veins that occur together with galena, sphalerite, and chalcopyrite. Covellite replaces the chalcopyrite, indicating the supergene process of this sulphide mineral (Idrus et al., 2015). Then hematite is formed, which terminates the mineralization process. In the Kotabunan area, valuable mineralization mostly associated with basemetal + quartz veins. Meanwhile, pyrite mineralization is present in all types of alterations characterized by a wide range of paragenesis.

The first sulfide phase that initiates mineralization is thought to be the presence of disseminated pyrite and quartz-pyrite veinlet associations that do not carry valuable mineralization or barren. Then, the formation of a basemetal mineralization sequence in the form of a vein, which plays an important role in the mineralization of precious metals, is evidenced by the presence of native Au and high levels of Pb-Zn-Ag based on XRF. In this basemetal vein sequence, sphalerite-galena mineral is believed to be the main carrier of Pb-Zn-Ag and leads to the formation of native Au. In this sequence, although the sulfide minerals in the base metal veins tend to be concurrent, the chalcopyrite minerals will close this sequence as a result of the decrease in sulfur fugacity and temperature in the sulfidation state (Einaudi et al., 2003).

Table 1. Paragenesis Sequence of Ore Formation Intermediate Sulphidation Epithermal at Kotabunan Prospect



Conclusions

Based on the geological mapping, the stratigraphy of the research area can be divided into 11 lithostratigraphy units, namely limestone unit, sandstone-limestone unit, andesite intrusion 1 unit, quartz diorite unit, tonalite unit, mudstone unit, sandstone-conglomerate unit, tuff-epiclastic unit, dacite lava unit, andesite intrusion 2 unit, and colluvium unit. The alteration zones consist of quartz + illite + kaolinite ± sericite alteration zones, and quartz + chlorite + illite + montmorillonite + calcite alteration zones. Both of them are formed in the Late Miocene-Early Pliocene and controlled by NE-SW sinistral and NW-SE dextral faults. Basemetal content is highly developed in the argillic alteration in the form of 1-3 cm vein.

The results of the combined paragenesis analysis of the samples show the presence of galena minerals in the early stages of formation, followed by sphalerite minerals. Subsequently, the tennantite mineral is formed, followed by chalcopyrite. Pyrite minerals were identified to be present during the hypogene mineralization stage from early to late stages, characterized by their overprint relationship with all ore minerals present. Gold is formed along with this hypogene process. Finally, covellite minerals are formed as a sign of supergene activity and hematite as a result of the oxidation process. Causative intrusions led to the formation of alteration and mineralization resulting from the mixing of magmatic hydrothermal fluids and meteoric hydrothermal fluids, forming veins that follow the structures in the study area.

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