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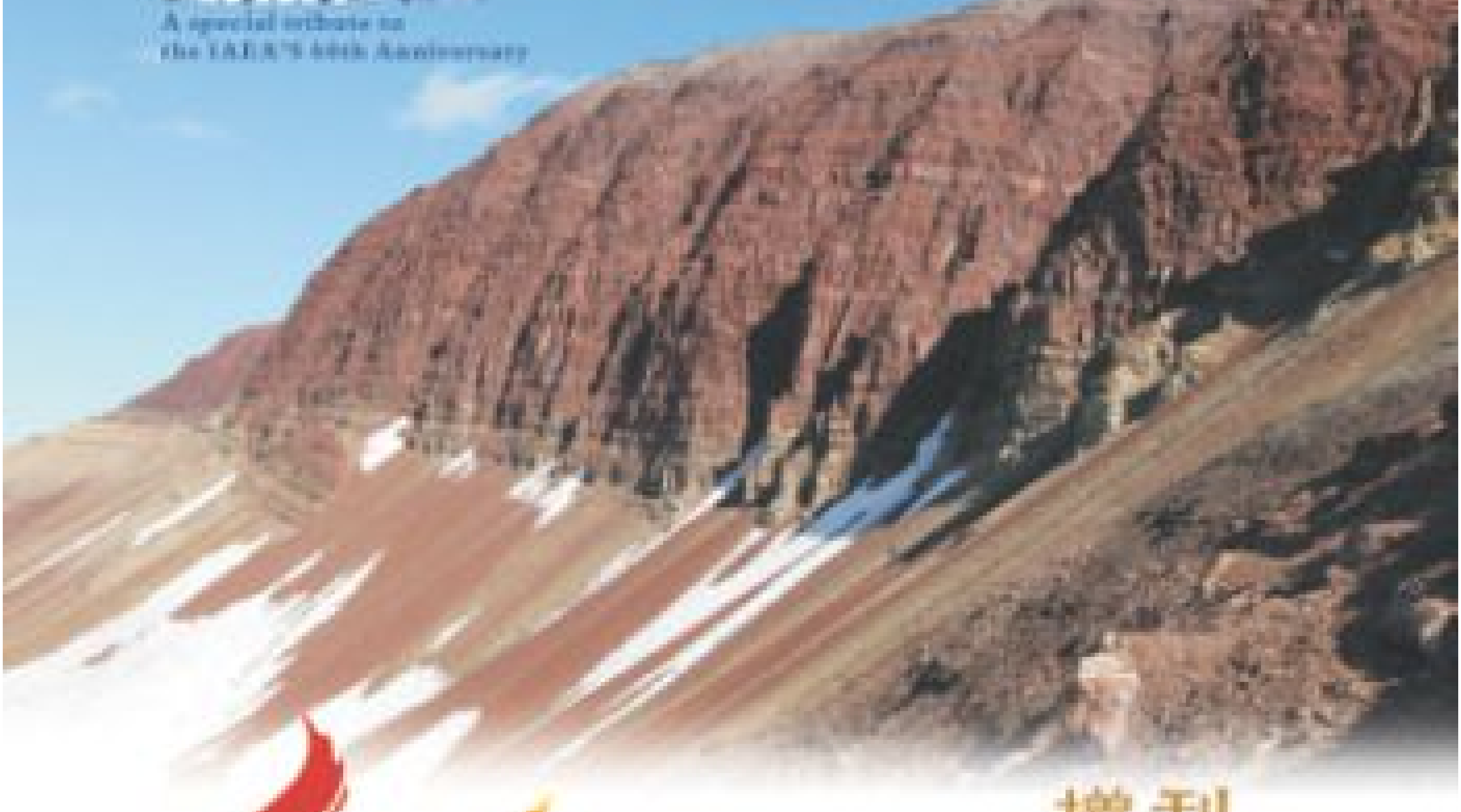
A special tribute to
the IAEA'S 60th Anniversary



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Foreword

Uranium exploration and mining in the Asia region is predicted to significantly increase in the coming years, with new nuclear power plants coming online as the demand for more low-carbon energy to power their economic development grows. The current future requirements for uranium in the region will be largely met by imported uranium. Increased power production requires more uranium for which countries need to develop effective, responsible and sustainable production plans. Even though mining in general is important to the socio-economics of most countries and increasing investments are expected, uranium sources so far identified are of low-grade, and current production negligible. The region has less than 7% of the share in the world identified uranium resources and only about 3% share in annual global production. The potential for finding new deposits of uranium are considerable high, but the current uranium exploration and mine development expenditures are low at about 11% of the total world expenditure. The potential for increasing domestic uranium production also exists, however several factors prevent this growth from materializing. The major challenges are absence of clear policies, lack of adequate expertise in exploration, mining and processing, poor stakeholder engagement and mounting mining waste issues, which often lead to failures or delays in commercial project execution. Many value-addition opportunities that could guarantee higher sectorial growth and economic returns are often missed, which often also lead to rising environmental issues. The four-year IAEA Regional TC Project RAS2019 "Conducting the Comprehensive management and Recovery of radioactive and Associated Mineral Resources" was launched in 2016 for supporting sustainable mining and production of uranium and other minerals. The overall objective of the project is to to enhance capacity in sustainable extraction of radioactive and associated materials such as uranium, thorium and rare-earth elements that can strengthen the socioeconomic resilience of the mining sector in Asia and the Pacific region. This special edition of East China University of Technology journal is one of the significant outcomes of the RAS2019 Project Coordination Meeting and Workshop on "Socio-economic outlook of radioactive and associated minerals production in Asia and the Pacific" held in Yogyakarta, Indonesia on 22 – 25 March 2016. All the contributed papers not only highlight the current progress of uranium explorations and comprehensive management and recovery of radioactive and associated mineral resources in participating states in the region, but also contribute to the celebrations of the 60th Anniversary of both the IAEA and ECUT.

This publication has been possible due to the excellent teamed-work of all involved in the project RAS2019. Thanks and appreciation are due to all authors and editors for their excellent work and cooperation to produce high-quality papers. Special thanks to Prof. Liu Xiadong of ECUT for putting his time, technical guidance and tireless effort to ensure that this publication can be materialized; to IAEA technical officer, Dr. Harikrishnan Tulsidas, for his technical guidance and continuing support; and to the IAEA Programme Management Officer, Dr. Syahril, for his initiative, guidance and support on this work.

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Identification of Mineralization Type and Specific Radioactive Minerals in Mamuju, West Sulawesi

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Abstract: Mamuju is the capital city of West Sulawesi province that has high value of natural radiation dose rate (radioactivity). The specific minerals that expose the radioactivity are not determined clearly. The purpose of this study was to determine of the specific radioactive minerals and the type of mineralization in Mamuju area. The study was conducted by geological mapping and radiometric measurements in order to identify the form and distribution of radioactive mineralization. Geochemical analysis was done by field sampling, mineralogy and SEM-EDS analysis to determine the specific radioactive mineral. The distribution of the value of radiation dose rates and occurrences of mineralizations are found only on the Tapalang, Ampalas, Adang and Malunda alkaline volcanic rocks complex. Radioactive minerals formed as a primary mineral are davidite ($(U,Ce,Fe)_2(Ti,Fe,V,Cr)_5O_{12}$) and thorianite ($ThO_2$). Beside these secondary minerals such as gummite ($UO_3 \cdot nH_2O$) and autonite ($CaO \cdot 2UO_3 \cdot P_2O_5 \cdot 8H_2O$) as well as the primary minerals were also identify along the sulphide minerals. Radioactive mineralization type are corresponds to volcanic related uranium deposit structural bound and supergene enrichment type which are controlled by lithology, geological structure and hydrothermal process. Existance of davidite, thorianite, gummite and autonite in alkaline volcanic rock will produce high natural radioactivity. Therefore those areas present a good potential for uranium exploration.

Key Words: radioactive; mineralization; alkaline volcanic rocks; Mamuju

Introuduction

Mamuju, the capital city of West Sulawesi Province, has the highest radiation dose rate in Indonesia. The radiation dose in this area reached 2 800 nSv/h (Iskandar et al., 2014). Radioactivity measurement was conducted using radiometric-carborne survey, using gamma surveymeter Exploranium GR-130 type on the main road network in Sulawesi island and other area in Indonesia (Figure 1). High radioactivity in Mamuju area was detected from Naturally Occurring Radioactive Material (NORM) in the rocks belonging to the Adang volcanic formation. The NORM identified in the distribution of Adang volcanic formation (Syaeful et al., 2014).

Mamuju area has a different geological setting comparing to other radioactive mineral exploration area in Indonesia. Radioactive mineral occurrences on volcanic rocks in Indonesia was found in other location such as Kawat Area East Kalimantan with different kind of volcanic rocks. The association lithology of uranium occurrences in Kawat Area is felsic volcanic rocks such as ryolite rhyolite (Ngadenin et al., 2011). Result of rocks affinity analysis for Adang volcanic complexes are ultrapotassic affinity (shosonitic or ultra potassic), while the other volcanic rocks have lower K_2O contents. These complexes were emplaced in specific geological setting, the active continental margin (ACM), which is located distant from subduction zone, and had long magmatic forming delay time (Sukadana et al., 2015). These conditions generate higher K_2O composition in the rocks. As would be ex-

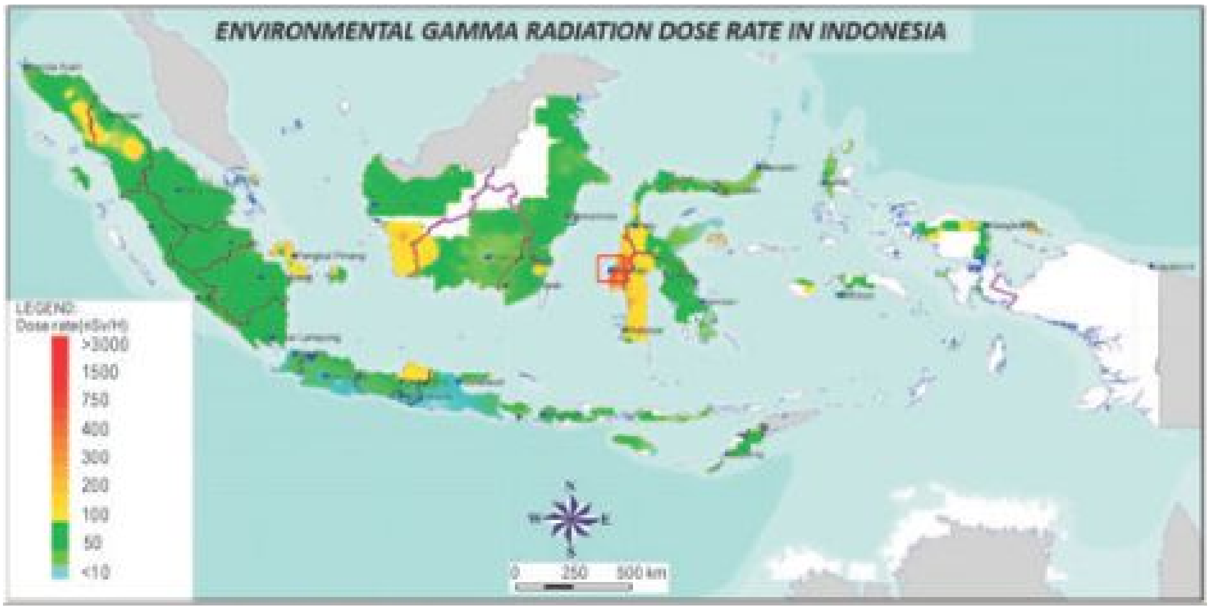


Figure 1 Map of gamma radiation dose rate in Indonesia (Iskandar et al., 2014).

pected from their potassic-alkaline nature, the volcanic rocks contain high amounts of Large-Ion Lithophile Element/LILE (Ba, Sr, Rb and K), light rare earth element (LREE), radioactive elements (U, Th) and zircon (Zr). Magma differentiation stages for Adang volcanic rocks were vary and resulting variation of ultrapotassic rocks. These conditions were created due to many different factors, which influence magma composition from fluid source, mantle-melting process to interaction with continental crust (Pearson et al., 2003).

Uranium mineralization type classification generally follows two alternative approaches. They are focusing on descriptive features of the mineralization and genetic aspects. Descriptive features include the identification on rocks type and orebody morphology (geological classification) or on their genetic aspects (genetic classification). Uranium deposit classification partly described in tabel 1 is used to classify uranium deposit in Mamuju Area. This classification is completed with number of uranium type deposits in the world (Bruneton et al., 2016).

In the past, geological data collection related to radioactive mineral in Mamuju area is very limited. Specific radioactive minerals and mineralization deposit types have not been studied yet. Since the high radioactivity map in Mamuju published to now, geological and detailed radiometric measurement are conducted in the area. The objectives are to determine the radioactive minerals and their deposit types by geological study, radiometric mapping and geochemistry laboratory analysis.

1 Method

The research location focused on radioactive mineralization in Mamuju Region, West Sulawesi Province. Research was conducted by field and laboratory works. The field work include geological and radiometric mapping using RS-125 gamma spectrometer to delineate the distribution of lithology and radiometric anomaly. Trenching and sampling were conducted in the location with high radiometric value. Identification of specific mineral performed by hand specimen observation using small handy microscope with 120x magnification. The samples also analyzed in laboratory using mineragraphy section. This section is completed with CN-85 film for autoradiography to detect the alpha tracks. The Scanning Electron Microscope-Energy Dispersive Spectroscopy (SEM-EDS) analysis conducted to identify the specific minerals, mainly radioactive minerals (Gu, 2003). Analysis of SEM-EDS conducted on three samples, Botteng 1, MJU 01 and MJU 21.

Table 1. Part of uranium deposit type classification (Bruneton and Cuney, 2016)

No	Type of Deposit	Subtype	Number of Deposits
1	Intrusive	1.1. Anatectic	55
		1.2. Plutonic	31
2	Granite-Related	2.1. Endogranitic	80
		2.2. Perigranitic	52
3	Polymetallic iron oxide breccia complex		16
4	Volcanic related	4.1. Structurebound	98
		4.2. Strata-bound	18
		4.3. Volcano-sedimentary	4

2 Result and Discussion

2.1 Mineralization Distribution

Volcanic rocks in Mamuju area are influenced by repetition of magmatism process. Distribution of volcanic complexes is identified from the form of specific crater location. Adang volcanic rocks complex in Mamuju Area was classified into seven volcanic complexes. They are Tapalang, Ampalas, Adang, Malunda, Sumare, Labuhan Rano and Karampuang volcanic complexes (Figure 2). Each volcanic complex is composed by different kind of volcanic rocks successions, such as lava, volcanic breccia and pyroclastic deposits. Distribution of lava as primary eruption product can only be found surrounding the center of volcano. Lavas are identified as lava flow and lava dome.

Radiometric value as result of gamma ray measurement indicated the correlation between volcanic activity and radioactivity value. High radioactivity value reflects the stage of volcanism and is generally found in center of volcanoes. High value of radioactivity are located in several places, which are Adang, Ampalas, Tapalang and Malunda volcanic complexes.

Three other volcanic centers reflect low radioactivity value. They are Labuanrano, Sumare and Karampuang complexes. According to the field observation, the three volcanic complexes are fresher than other complexes. Qualitatively, it is an indication that they have a different age with other volcanic complexes. Labuanrano, Sumare and Karampuang complexes are younger than other volcanic complexes.

High value of radioactivity is reflecting the distribution of radioactive minerals. Four complexes that have high value of radioactivity have different type of rock and stage of mineralization. Mineralization in Adang volcanic complex was found in vein filling structures and within pores of volcanic rocks.

The disseminated radioactive mineral occurrence in Adang, found in the area composed by leucitic lava, mainly in volcanic centers. Radioactive minerals formed mainly in weaker zones like volcanic center aperture and extensive geological structures oriented NE-SW, N-S, and E-W. The other volcanoes like Malunda, Tapalang, and Ampalas volcanic complex generally found as alteration, supergene enrichment, lateritization, and increasing of ferromagnesian mineral in the surface (Indrastomo et al., 2016). The ratio of Th/K shows the evidence of the process where higher the Th/K ratio distributed on the older volcanic rock (Malunda, Tapalang, and Ampalas) (Syaeful et al., 2014).

From the radioactive mineralization pattern where uranium and thorium have higher grade on ultrapotassic rocks in Adang complex, there are differences between mineralization process on uranium and thorium in other area. Uranium mineralization in Tapalang, Ampalas and Malunda volcanic complex occurred in the rocks fractures, mainly at the volcanic centers and intensive structural pattern, and influenced by hydrothermal process. These are the indication that uranium deposit is a structure bound deposit, not the strata bound deposit. Meanwhile, the thorium deposit are found in the complex structural area and influenced by

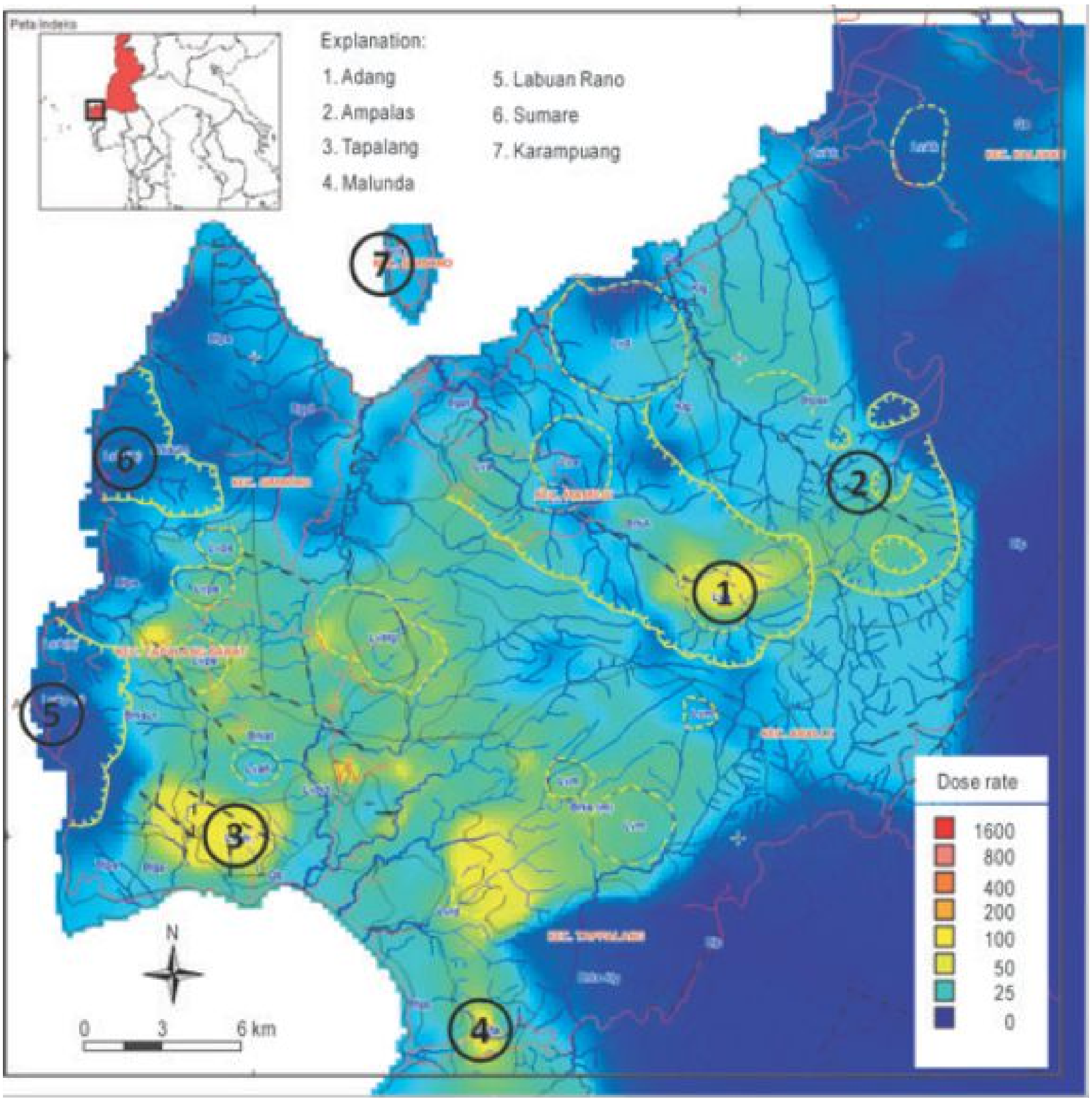


Figure 2 Distribution of radiation dose rate anomalies mainly in the volcanic center and structural zones.

hydrothermal and lateritization process. These are the indication that thorium deposit classified as alkaline igneous complex and supergene enrichment (Bruneton et al., 2016; Gaafar et al., 2014).

2.2 Mineral Identification

Radioactive mineral occurrence was detected in the field from their radiometric values using gamma spectrometer. At Botteng and Takandeang (Tapalang volcanic complex), radiation dose rate are above 1 500 nSv/h. Radioactive minerals identified from these locations are the phosphate hydrate group with fluorescent and apple green colors on the sample. They are gummite ($UO_3 \cdot nH_2O$) (Pagel, 1982) and autunite ($CaO \cdot 2UO_3 \cdot P_2O_5 \cdot 8H_2O$) (Shvareva et al., 2009) as seen on Figure 3. These minerals are relatively easy to observe because of their abundance in the rocks. This mineral has a specific fragment and forms an ideal cleavage. Gummite is uranium secondary mineral with hydrate oxide group, lemonade yellow color, and massif. The crystal form is orthorhombic, but commonly it is difficult to identify it. These two minerals commonly occurred within rocks pores, calcitic gas cavities and minor fractures on rocks. Occurrences of these minerals are reflected the advance process in the rocks formation. The mineral formation also influenced by ground water leaching (Shvareva et al., 2009).

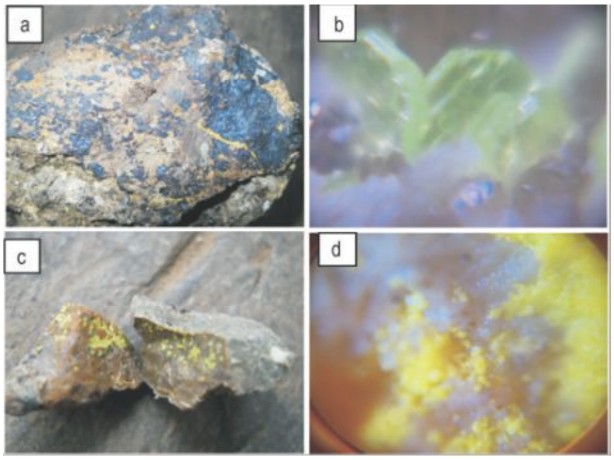


Figure 3 (a) Autunite apperance on rock sample; (b) Autunite observation using 120x magnifier; (c) Gummite on rock; (d) Gummite with 120x magnification.

Primary uranium minerals are very difficult to identify in the field. Laboratory analysis then conducted to identify them. The first analysis is mineragraphy section completed by autoradiography analysis using CN-85 film along three days. The result of the autoradiography analysis is high intensity of alpha tracks in CN-85 film (Figure 4). It is not easy to determine the radioactive minerals from the section. Another analysis method is used to solve the problem. The SEM-EDS analysis then conducted using mineral mapping and mineral pointing techniques.

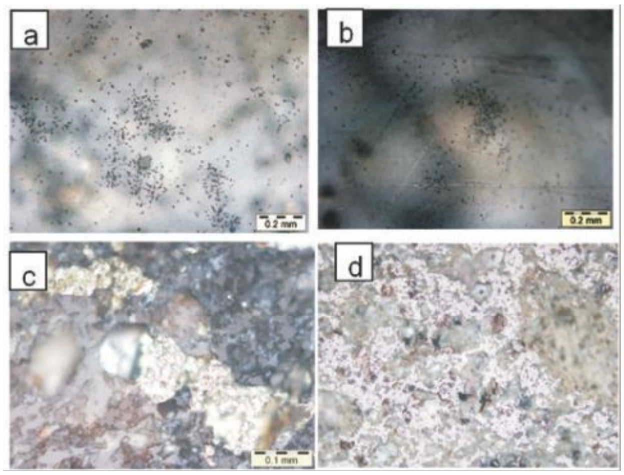


Figure 4 (a, b) Alfa (α) particle tracks penetrate CN 85 film; (c, d) Radioactive minerals occurrences and their associates.

2. 3 Scanning Electron Microscope-Energy Disperse Spectroscopy (SEM-EDS) Analysis

Analysis on Botteng 1 conducted both in mineral mapping (Figure 5) and mineral point analysis (Figure 6). The point analysis results chemical composition of sample. By using the chart of composition and crystal form (Pagel, 1982), the mineral identified as davidite ($(U,Ce,Fe)_2(Ti,Fe,V,Cr)_5O_{12}$)). Analysis on MJU 01 sample using point mineral analysis (Figure 7) indicate a composition of davidite ($(U,Ce,Fe)_2(Ti,Fe,V,Cr)_5O_{12}$)) as radioactive mineral on the sample. Analysis on MJU 21 sample characterizes an amorphous the amorf crystal with high percentage content of thorium (Figure 8). Radioactive mineral identified from the sample is thorianite (ThO_2).

2.4 Mineralization Type

Radioactive minerals in volcanic rocks are very rare, especially in Indonesia. Moreover, the occurrences of radioactive mineral (uraninite, pitchblende) are generally associated with acid/ryolitic volcanic rocks. The radioactive minerals in Mamuju are identified as thorianite (ThO_2) and davidite ($(U,Ce,Fe)_2(Ti,Fe,V,Cr)_5O_{12}$)) as well as secondary minerals in the form of gummite and autunite. These minerals are metamict mineral which are very difficult to identify in the fresh rock. The result of SEM-EDS analysis shows that davidite ($(U, Ce, Fe)_2(Ti, Fe, V, Cr)_5O_{12}$)) contains a radioactive substance which is disseminated on K-feldspar. The formation of radioactive minerals and deposits in the Mamuju area greatly influenced by lithology, hydrothermal process and geological structure.

2.5 Lithology

The lithology distribution is one controlling factor of radioactive minerals occurrences. The composition of volcanic complex in Mamuju has high affinity to the K-calk alkaline shosonitic with basaltic to andesitic rock types. These rocks can be formed by the influence of several things such as the source of magma derived from the melting of the mantle wedge which caused by high content of Ca and Al_2O_3 on the rock. High levels of K_2O occured when magma as a result of remelting mantle moved into continental crust and en-

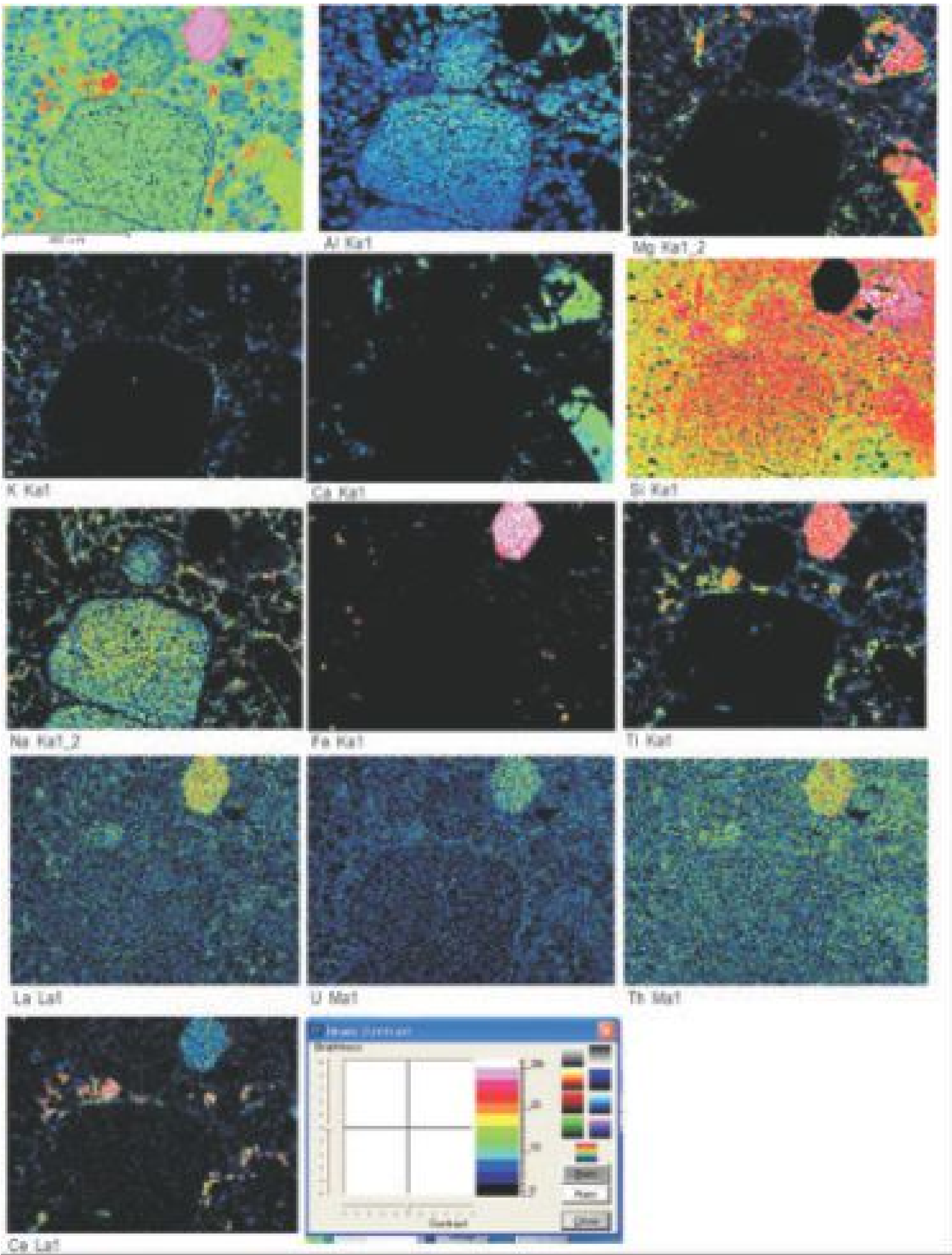


Figure 5 Mineral mapping using SEM-EDS for sample Botteng 1.

riched by the continental crust contamination which was very mature. This process did not have any linear correlation with subducting oceanic crust during subduction time. The enrichment of K_2O is associated with incompatible elements, including radioactive elements. The longer magma interact with continental

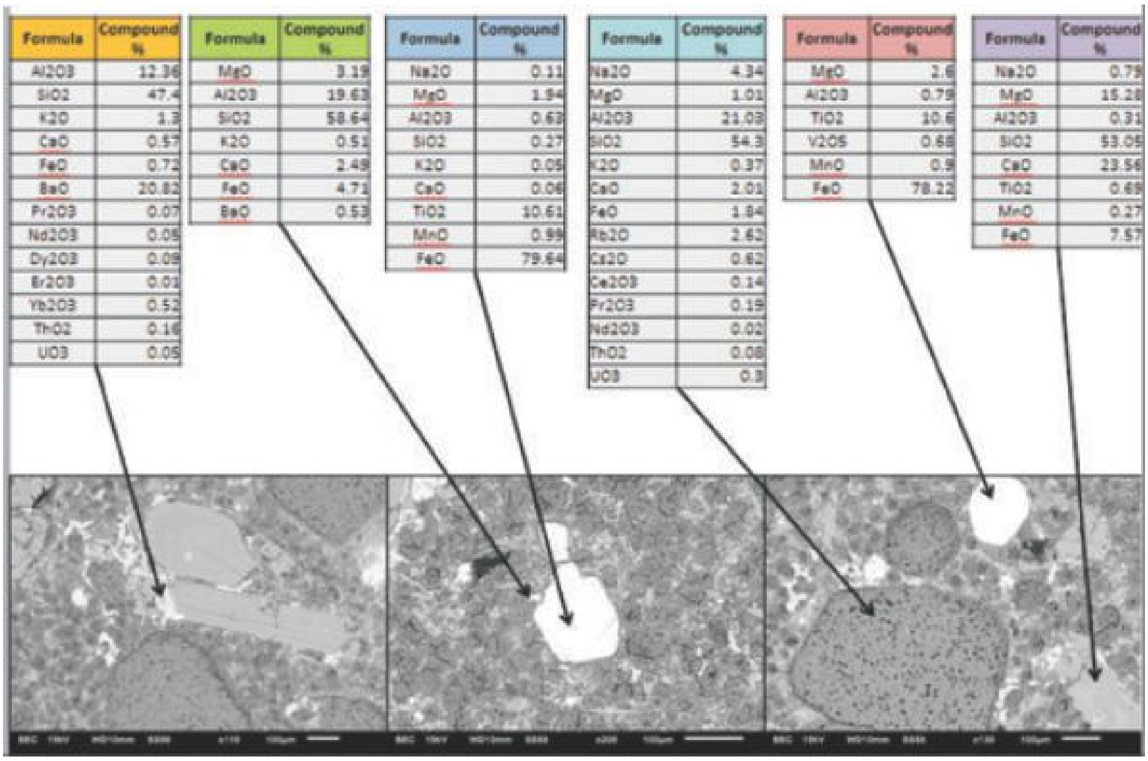


Figure 6 Result of point analysis SEM-EDS for sample Botteng 1.

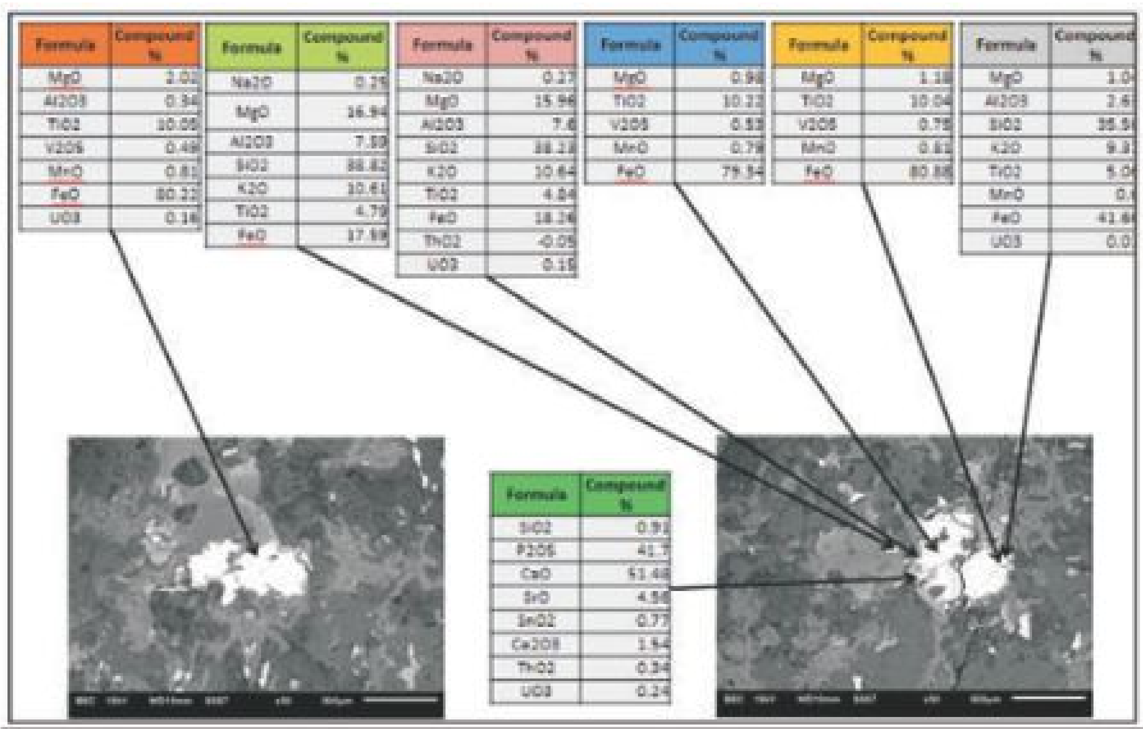


Figure 7 Result of point analysis SEM-EDS for sample MJU 01 Salunangka.

crust will influence the high content K_2O in volcanic rocks. This process will be linier with the increase of the radioactive elements content in the rocks (Burwash et al., 1978).

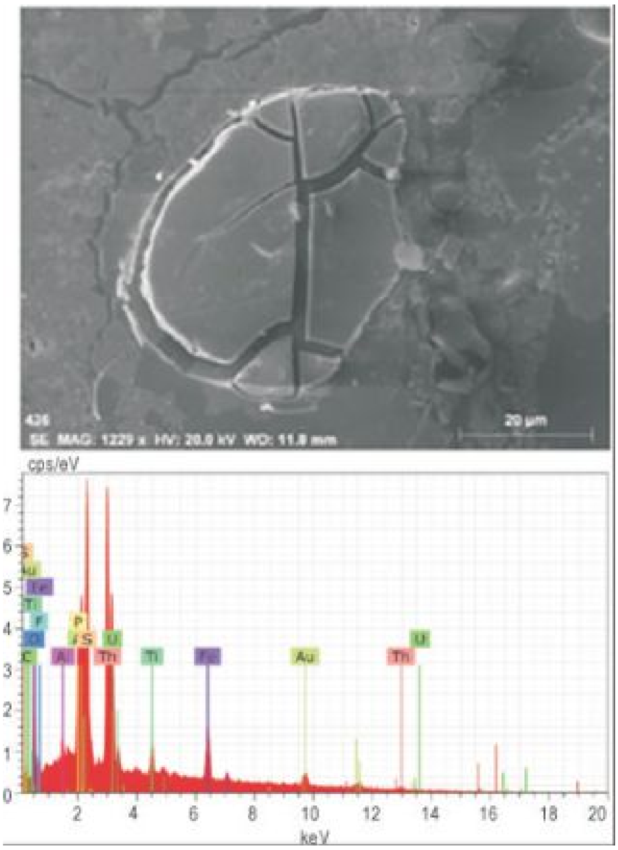


Figure 8 Result SEM-EDS analysis for sample MJU 21.

According to the natural enrichment process, it is concluded that the rock with shosonitic affinity or ultrapotassic became the source of a radioactive elements. In Mamuju area, it is concluded that the volcanic rocks of Adang complex is the source of radioactive minerals. Mineralization in Adang volcanic complex formed as disseminated in the volcanic rocks pores. In other way, the high content of uranium also occurred on geological structure trap that have been affected by hydrothermal process. Radioactive minerals in Malunda, Ampalas and Tapalang are generally found in secondary mineral form and in red soil as product of supergene enrichment. They are older than Adang complex.

Distribution of radioactive minerals occurrences constrain by the distribution of high-K affinity of volcanic rocks. This mineralization did not occurred in other rock types (non-volcannics) or in the volcanic rocks that have low-K affinity, such as Sumare, Labuan Rano and Karampuang complexes (Maithani et al., 2011).

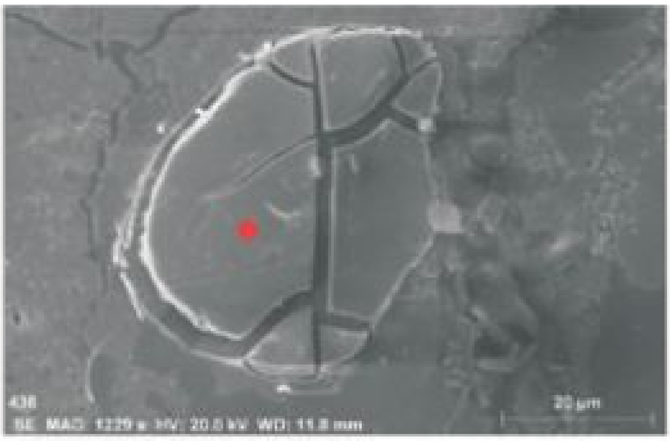
2.6 Hydrothermal Process

Hydrothermal process will mobilize radioactive elements contained in metamic minerals in rocks and will lead to the precipitation of new minerals due to the heat source and the resulting fluid (Ohnuki et al., 2004). This will allow the formation of mineralized/enrichment of radioactive elements in the alteration and structure zones. The concentration of radioactive element will be increased with the increasing of the lithdogy age. The radioactive minerals in rocks will be greater in older rock, especially in alteration zones.

Primary mineralization can occurred in the porous rock and structural vein (Horie et al., 2010). These process also produces the formation of secondary radioactive minerals with oxides and phosphates bonding (gummite and autunite). Repetition of volcanism stages can be characterized from hydrothermal manifestations that still exist in the form of hot spring at the center of the volcanic until nowadays. It has strongly influenced the formation of radioactive minerals.

Radioactive minerals occurrences in the alteration zones and hydrothermal process zone also create the natural dissolution of the radioactive elements. Uranium as mobile element will leave the immobile element (thorium) (Ohnuki et al., 2004). This condition may contribute to the forming of the supergene enrichment with high immobile elements and lateritization process of mobile elements. Exogenic factors such as weathering, oxidation and groundwater level will contribute highly control the occurrences of radioactive minerals and the enrichment of radioactive elements. The SEM-EDS analysis performed on several samples which are MJU 21, MJU1 and Botteng 1 showing it. Hydrothermal has high influence on the radioactive element deposit formation and mobilization of uranium from radioactive minerals. Result of SEM-EDS analysis showing the thorianite mineral has high content of thorium and low content of uranium (Figure 9). Meanwhile, based on point analysis in the mineral fractures, this area has higher uranium content than in the mineral body (Figure 10). These results prove the influence of hydrothermal process in radioactive mineral formation.

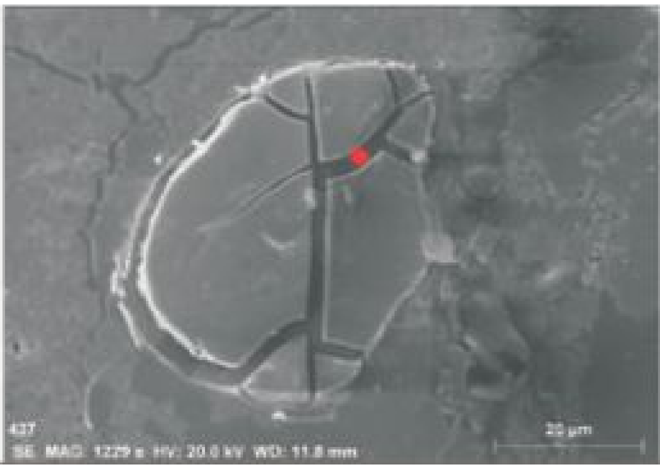
Hydrothermal process had greatly effected Malunda, Tapalang and Amapalas complexes. This condition occurred because the rocks on these complexes were older than others in the area. High radiometric value formed in thickness of red soil as product of hydrothermal alteration and supergene enrichment of radioactive minerals.



Spectrum: Acquisition 56

Element	Series	unn. C [wt.%]	norm. C [wt.%]	Atom. C [at.%]
Carbon	K-series	1.11	1.38	5.56
Oxygen	K-series	13.92	17.37	52.42
Fluorine	K-series	0.16	0.20	0.51
Aluminium	K-series	0.64	0.80	1.43
Titanium	K-series	2.06	2.57	2.59
Iron	K-series	6.15	7.67	6.64
Gold	L-series	13.02	16.25	3.98
Thorium	M-series	31.14	38.85	8.09
Uranium	M-series	1.16	1.45	0.29
Phosphorus	K-series	2.61	3.25	5.07
Sulfur	K-series	6.77	8.45	12.72
Calcium	K-series	0.00	0.00	0.00
Manganese	K-series	0.11	0.14	0.12
Silicon	K-series	0.01	0.01	0.01
Yttrium	L-series	0.00	0.00	0.00
Lanthanum	L-series	0.46	0.57	0.20
Cerium	L-series	0.84	1.05	0.36
	Total:	80.17	100.00	100.00

Figure 9 Point analysis in thorianite minerals from sample MJU 21 showing thorium content much higer than uranium.



Spectrum: Acquisition 57

Element	Series	unn. C [wt.%]	norm. C [wt.%]	Atom. C [at.%]
Carbon	K-series	0.50	0.71	2.61
Oxygen	K-series	6.61	9.41	26.12
Fluorine	K-series	0.00	0.00	0.00
Aluminium	K-series	0.67	0.95	1.57
Titanium	K-series	0.62	0.88	0.82
Iron	K-series	7.38	10.50	8.36
Gold	L-series	11.27	16.03	3.62
Thorium	M-series	8.62	12.25	2.35
Uranium	M-series	4.24	6.03	1.13
Phosphorus	K-series	0.68	0.96	1.38
Sulfur	K-series	2.14	3.04	4.22
Silicon	K-series	5.44	7.74	12.25
Calcium	K-series	0.00	0.00	0.00
Manganese	K-series	0.14	0.20	0.16
Yttrium	L-series	0.00	0.00	0.00
Lanthanum	L-series	0.12	0.17	0.06
Cerium	L-series	0.00	0.00	0.00
Potassium	K-series	21.88	31.12	35.37
	Total:	70.32	100.00	100.00

Figure 10 Point analysis in fracture of thorianite minerals showing the uranium content higer than in the minerals body

2.7 Geological Structure

Geological structure controls the radioactive minerals distribution especially in Adang complex. This factor is very important as favorable zone of radioactive mineral deposits. Besides the weak zone as the pathway of hydrothermal fluid flow, the structure zone is the zone where surface water seep into the ground. Both of these functions are significant factor in radioactive mineral deposits formation.

Mamuju area is an area that has intensity of tectonics influenced. These reflected in complexes pattern of geological structure in this area. The geological structures that have high value of radioactivity as radioactive minerals traps are faults with northeast-southwest, north-south and east-west pattern.

3 Conclusion

High radioactivity in Mamuju area comes from natural materials (NORM) as a mineral composition in volcanic rocks and their soil products. High radioactivity distributed in the high K-alkaline volcanic rocks of Adang Volcanic Formation. Radioactive minerals occurring the volcanic rocks are davidite and thorianite as primary minerals, meanwhile gummite and autunite deposited as secondary minerals in rocks pores, fractures, and soil. Radioactive minerals deposits were controlled by the lithology, repetition of volcanic stages, geological structures, hydrothermal process, supergene enrichment and lateritization. The minerals distributed in the Adang, Tapalang, Ampalas, and Malunda complexes. These complexes are developed as future exploration target for radioactive minerals in Mamuju Area.

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