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Geology and Mineralogy of Zeolitic Tuff in Tulul Unuqar Rustum Ash Shmaliyya, NE JORDAN

Khalil M. Ibrahim*

Department of Earth & Environmental Sciences, Hashemite University, Zarqa, Jordan

Abstract

Detailed geological mapping and drilling campaign in Tulul Unuqar Rustum Ash Shamaliyya (URS) volcanic center have been carried out to evaluate the geology and mineralogy of zeolitic tuff. The most abundant rock type in the volcano is scoriaceous tuff and zeolitic tuff. The zeolites occur as cement to palagonite clasts and as filling inside the vesicles, making about 30% of the rock. Phillipsite is the most abundant zeolites mineral with spherulitic radiating aggregates or stout-prismatic crystals. The second zeolites mineral is chabazite which occurs as rhombohedra with simple penetrating twinning. Chemical analysis of the phillipsite indicates that they are K-rich type with similar Na/K and Ca/K ratios and has small variation in the Si/Al ratio. The zeolitic tuff samples have a wide range of SiO_2 content indicating wide range of alteration and formation of authigenic minerals. The model of zeolites formation in the URS volcano is believed to be an open hydrological system model, where soluble elements released from the fresh basaltic glass by the hydrolysis reaction due to the raised higher pH and saline conditions, re-deposited in the of form zeolites, smectite and calcite.

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Keywords: phillipsite, chabzite, zeolitic tuff, NE Jordan.

1. Introduction

Harrat Ash Shaam Basaltic Group (HAS) exposed in northeast Jordan is considered the largest volcanic field in the Arabian Plate (Ibrahim, 1993, Ibrahim et al., 2003, Shaw et al., 2003) (Fig. 1). The basaltic activity of the HAS was manifested in form of intermittent pulses of eruption started from about 26 Ma and continued to about 0.1 Ma ago (Illani et al., 2001). As a result, volcanic tuff deposits were extensively formed from isolated volcanic centers, which are confined to three volcanic fields as shown in Fig. 1 (Ibrahim et al., 2006). Locally, the black variety of the volcanic tuff is commercially exploited as pozzolanic tuff in cement industry. The estimated annual production of the black pozzolana from the HAS Basaltic Group is about 500,000 ton. Zeolites were discovered and reported in several volcanic centers associated with the volcanic tuff (Dwairi, 1987, Ibrahim, 1996; Al Dwairi et al., 2009; Khoury et al., 2015). A new occurrence of zeolites was reported in Tulul Unuqar Rustum Ash Shamaliyya (URS) area during an intensive geological exploration program to delineate the commercial pozzolanic tuff deposits. The URS is a cinder cone of Tlul Esh Shahba volcanic field (Fig. 1, Field 2), which is about 55 km east of As Safawi town along the Amman – Baghdad highway. It is situated about 20 km to the south of the highway. The study area is bordered by the coordinates 394900 – 395900 E and 180700 -181700 N; Palestine Grid. The main objective of this paper is to contribute to the geology, mineralogy and geochemistry of the zeolitic tuff in the URS area.

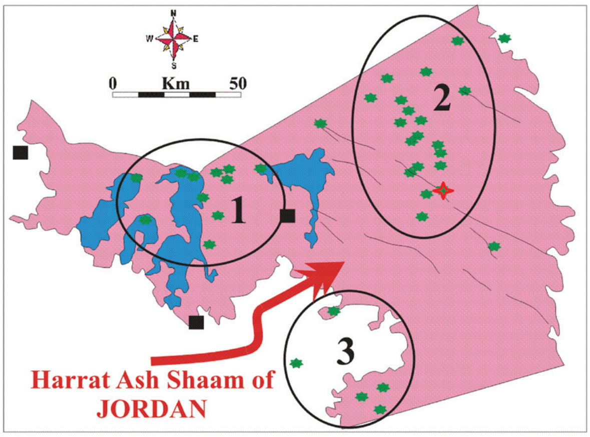

Figure 1: Location map of Harrat Ash Shaam Basaltic field and location of Tulul Unuqar Rustum Ash Shmaliyya (URS) volcanic center. (1): Asfar volcanic field, (2): Tlul Esh Shahba volcanic field, (3) Qatafi volcanic field.

2. Geological Setting

The HAS was classified by Ibrahim (1993) into five volcanic groups. As illustrated in Table 1, the URS volcanic center comprises rocks belong to the Asfar and to Rimah groups. The former is defined simply to include the basaltic rocks, which pre-date or associated with the Rimah Group (Ibrahim et al., 2006). The latter is subdivided into two formations of which the Aritayn Formation contains authigenic zeolites (Ibrahim, 1996). The second is Hassan Formation. The Aritayn Formation is developed from composite cinder

* Corresponding author. e-mail: Ibrahim_kh@yahoo.com

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strato-volcanoes. The formation consists of stratified, sorted, poorly cemented air-fall tuff and agglomerate, which are rarely intercalated with short lived lava flows from a central vent. Lithologicaly, it is made up of fine-grained ash, angular and spherical lapilli as well as volcanic bombs and blocks of different size and shapes cemented either by carbonate or zeolites and carbonate. However, non-cemented volcanic ash and lapilli is sometimes found forming tephra deposits.

Table 1: Late phases of Harrat Ash Shaam basaltic field (Ibrahim et al., 2006)

Eruption Phase	Age (Ma)
Bishriyya Group	1.80 – 0.15
RIMAH Group	2.19- 0.60
Late NW-SE dykes	3.09-1.64
ASFAR Group	4.52 -1.00

3. Methods of Investigations

Field work was conducted to execute detailed geological mapping and exploration drilling. A detailed geological map at a scale of 1: 5000 was prepared for the area (Fig. 2). Four exploration boreholes 15 to 25 m deep were drilled. Continuous core drilling was used to obtain representative samples. The cores were subjected to detailed and systematic description of their lithology, texture and color (Fig. 3). 20 representative samples were selected from the zeolitic tuff for further studies and analyses. Mineralogical and petrographical characterization of the core samples was carried out using the polarizing microscope. Normal thin sections were prepared for the representative samples. The samples were characterized using petrological, mineralogical, and chemical methods.

The analytical work included the minerals identification using X-ray diffraction analysis (XRD) and chemical analysis using X-ray fluorescence techniques (XRF). A Shimatzu X-ray diffractometer with CoKα radiation was carried out in the Royal Scientific Society according to (SOP) No. 131M03-010 test method. Whole rock 'random powder' samples were scanned with a step size of 0.02° 2 theta and counting time of 0.5 s per step over a measuring range of 2 to 65° 2q. Powdered samples were analyzed using a XRF machine (PANanlytical-Axios and a PW2400 spectrometer). Samples are prepared by mixing with a flux material and melting into glass beads. After mixing the residue with 5.0 g lithium metaborate and 25 mg lithium bromide, it is fused at 1200 °C for 20 min. The calibrations are validated by analysis of Reference Materials. Blank samples and several certified reference materials (CRM) were used for the correction procedures. The beads were analyzed by wavelength dispersive x-ray fluorescence spectrometry (WD-XRF). To determine loss on ignition (LOI) 1000 mg of sample material are heated to 1030 °C for 10 min. To investigate crystal forms and the paragenetic relationships of the zeolites and non-zeolites, scanning electron microscope (SEM) and Energy-Dispersive X-ray (EDX) study of selected samples has been carried out using a PANanlytica based in Mango Center, University of Jordan.

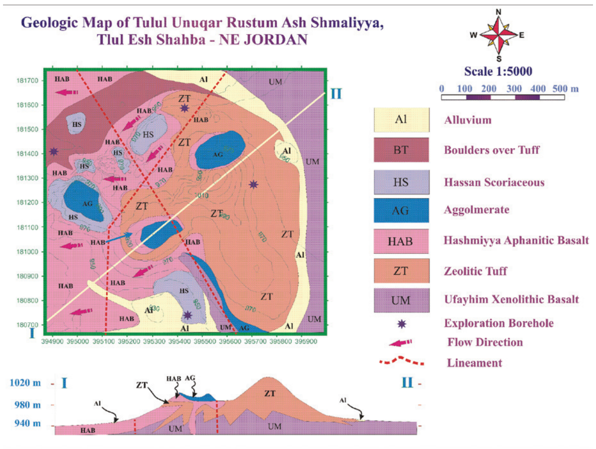

Figure 2: Geological map of Tulul Unuqar Rustum Ash Shmaliyya volcanic center

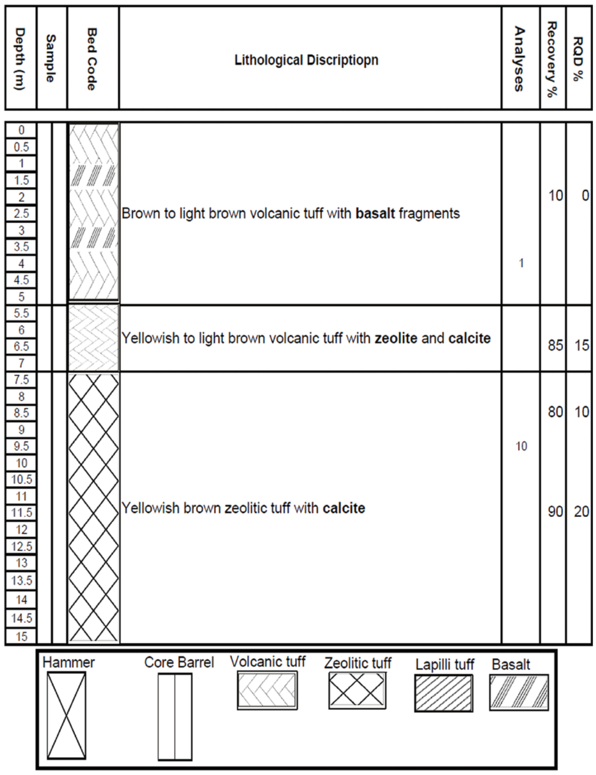

Figure 3: Lithological description of selected borehole drilled in the study area.

4. Results

4.1. Geology and Volcanology

The URS is a conical crescent shape volcano (Fig. 4a & 4b) that is about 750 m wide and 950 m long. It consists of three summits surrounding a depression, facing the southwest (Fig. 4a). The topographic relief in the volcanic cone is about 70 m, varying from altitude about 940 m in the low laying ground surrounding the cone to altitude 1030 m at the north eastern summit of the volcano (Fig 4a & 4b).

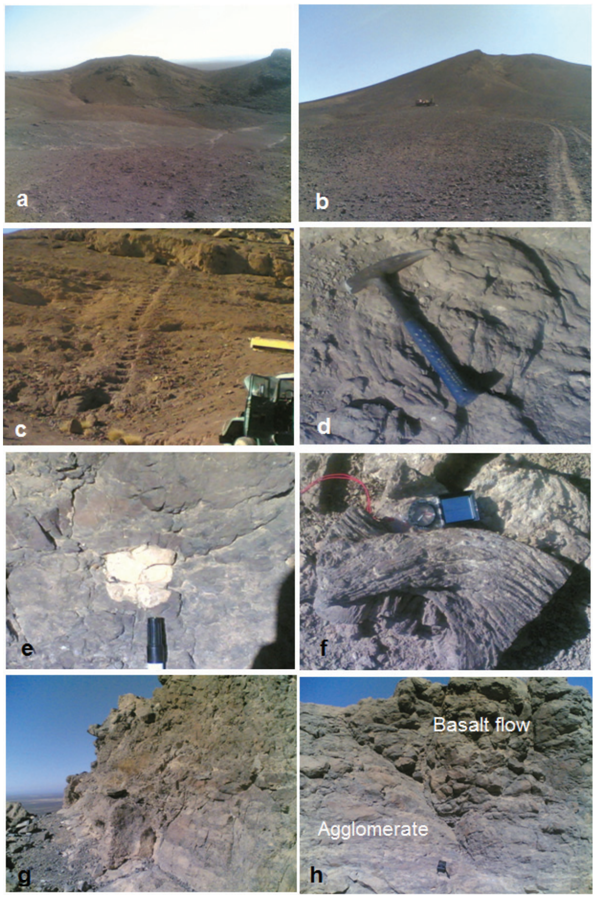

Figure 4: Lithology and morphology of URS volcanic center. (a): Morphology of volcanic center; (b): Zeolitic tuff unit (c): Hassan scoriaceous unit; (d): Sandstone inclusions in Ufayhim basalt unit; (e) Sandstone inclusions in Ufayhim basalt unit; (f): Volcanic bombs; (g): Agglomerates unit; (h): Basaltic flow over agglomerate.

Detailed geological mapping of the URS (Fig. 2) indicate presence of several lithological rock units outcropping in the URS area. The Ufayhim Xenolithic Basalt is the oldest unit. It displays closely spaced horizontal jointing, which appears as thin laminations. It contains huge amounts of lherzolite and dunite xenoliths sometimes forming up to 50-60%. They also host large pyroxene xenocrysts. The xenoliths and xenocrysts vary in size from few millimeters up to 15 cm, and are rounded. The xenoliths have brown reaction rims, comprising mainly granular, orange, olive brown olivine and lesser amounts of dark green tabular pyroxene.

The zeolitic tuff of the Aritayn Formation (Fig. 4c) and the Hassan scoriaceous tuff (Fig. 4d) are very similar in lithology. The difference is that the clasts in the former unit are cemented by zeolites and calcite. The zeolitic tuff consists of stratified, sorted, air-fall volcaniclastic (tuff, labillistone, volcanic breccia and agglomerate). The unit contains ash, lapilli, volcanic bombs and basaltic blocks of different size and shapes that are all cemented by zeolites and carbonate. The volcaniclastic layers are arranged in shower bedding maintaining an even thickness and exhibit graded laminations with particle size from 1 mm to 50 mm. zeolitization of the volcanic tuff is not consistent. Localities with high degree of zeolitization are characterized by presence of yellowish light brown color, soft and friable highly altered lapilli clasts. They are cemented by a thick coating of zeolites and calcite (Fig. 5). The exposed thickness of the zeolitic tuff in the URS is from 10 m to 30 m whereas the scoriaceous tuff is from 2 m to 15 m (Fig. 3). The scoriaceous tuff is characterized by presence of highly vesicular lappili and cinder clasts (grain size >4 mm) called scoria. The unit comprises bedded, friable, reddish brown to violet in color, poorly sorted, coarse-grained scoria (generally 0.5-20 cm long, and with average of 6 cm) making about 85% of the rock, along with volcanic bombs and blocks with various shapes and up to 50 cm-sized (Fig. 4f). Mantle xenoliths and lower crust lithic fragments are abundant in the zeolitic tuff and scoriaceous tuff units (Fig. 4e). They also host large pyroxene xenocrysts.

Figure 5: Hand specimen of the zeolitic tuff

The Hashimyya basalt is the second basaltic phase in the URS area. It covers most of the western and northern parts of the volcano, and makes about 25% of the URS area. It is less than 15m thick and comprises thin flow units, mostly between 2 m and 3 m thick, sometimes inter-bedded within the volcanic tuff units. The basalt is melanocratic, bluish-gray to medium-dark gray, aphanitic, holocrystalline olivine-phyric basalt with microporphyritic textures.

Agglomerate is a very hard pyroclastic unit with large clasts (Fig. 4g). It occurs as a massive, hard, about 5 -10 m thick unit consists of 2 –4 beds occupies the central part of the URS area. It gives weathering colors of brown, red brown and black. It includes bombs with various shapes (Fig. 4f). The clasts are from 15 cm to 50 cm-sized and with average of 20 cm. The unit is associated with thin basaltic dikes and flows (Fig. 4h).

Field evidence for faulting in the basalts of the mapped area is usually equivocal. Therefore, the structure interpretation is based mainly on: interpretation of satellite images, the alignment of volcanic centers and vents along with the abrupt variation in elevation of lava surface. In the mapped area, the faulting is influenced by the regional NW- SE tectonics related to Qitar El Abid dike and the associated fault system (Fig. 1), which is a regional structure extends more than 100 km with not less than 500 m wide (Ibrahim et al., 2003, Rabba' and Ibrahim, 2005). It appears as a normal fault with a possible strike-slip movement. The other important fault trend is the dominant N-S fault associated with the central volcanism of the Asfar Group occurring in the HAS in both the Asfar and Tlul Esh Shahba volcanic fields (Fig 1) (Ibrahim et al., 2006). The fault is indicated based on the alignment of the volcanoes and the associated volcanic vents, although direct evidence for vertical and/or horizontal displacement is lacking.

4.2. Petrographic Study

The studied samples comprise highly-altered volcanic tuff with variable amounts of secondary minerals including zeolites in the form of phillipsite and chabazite; smectite; calcite and gypsum. The volcanic clasts comprise highly vesicular dark brown, yellowish brown to deep red color palagonite. Color variability reflects the degree of alteration (i.e. palagonitization). The vesicles are either encrusted by rim of the secondary minerals or totally filled with the secondary phases. These vesicles are circular, subcircular and sometimes irregular in shape. The palagonite is characterized by its vitreous luster with conchoidal fracture. Micro-phynocrysts of anhedral olivine and iddingsite are common. Plagioclase is found as euhedral to subhedral phenocrysts or small laths in the groundmass with albite twinning.

All samples contain variable amounts of phillipsite crystals. It occurs mainly as colorless, radial, fan shape crystal aggregates or as spherulitic growth. Phillipsite crystals form thin rim encrusting walls of the vesicles or cementing the palagonite clasts. Chabazite appears as colorless sugar-like, rhombohedral crystals with twinning. Smectite appears as colorless, cloudy rim fringing palagonite granules and/or encrusting the vesicle walls with minute fringes in most of the samples.

4.3. Mineralogy

In addition to presence of primary minerals including olivine, pyroxene and plagioclase, the sample host secondary minerals including zeolites. The zeolites occur in form of phillipsite and chabazite. Quantitative determination of these two minerals was aided by XRD. As Show in Table 2, zeolites content is very high in specific areas among the mapped areas. The total zeolites content is up to 60% in the borehole No. 1 and close to 49% in the borehole No. 2. Phillipsite is the most dominant zeolites mineral which makes up to 49% in some samples. The average zeolites content in the zeolitic tuff in the URS is 30%.

Table 2: Qualitative mineralogical composition of the zeolite samples by (XRD).

Sample No.	BH No.	Depth (m)	Phillipsite (wt%)	Chabazite (wt%)	Total Zeolite (wt%)
1	1	3 – 4	21	12	33
10	1	9 – 10	45	15	60
8	2	5 – 6	28	21	49
9	2	24 – 25	49	0	49
11	2	16 - 17	44	3	47
5	3	5 – 6	8	0	8
6	3	6 – 7	6	0	6

4.4. SEM and EDX Results

The SEM study shows that phillipsite occurs either as spherulitic radiating aggregates or stout-prismatic crystals elongated along the a-axis (Fig 6a, 6b & 6c). Phillipsite forms pseudo-orthorhombic twinned prismatic crystals evident from the two-sided or four sided dome capping the end of the prism (Fig. 6b). Some of the phillipsite twins imitate a tetragonal prism. The spherulites are about 40µm, and range in length from 30µm to 100µm and in width from 10µm to 5µm. EDX study of phillipsite (Fig. 7) indicates that Si/Al ratio is between 2.45 and 2.65, Na/K ratio is 0.95 and Ca/K ratio is around 0.45 (Table 3). It can be concluded that URS phillipsite is K-rich phillipsite.

Chabazite occurs in colorless rhombohedra with simple penetration twinning (Fig. 6e & 6f). Crystals vary in size from 50 up to 250µm. Smectite occurs as colorless aggregates of tiny flakes (Fig. 6d) or as thin rim fringing palagonite clasts, or encrusting vesicle walls.

Table 3: Energy dispersive X-ray analysis of phillipsite crystals.

		BH1		BH2	
El	Z	(Wt %)	Atomic (%)	(Wt %)	Atomic (%)
O	8	43.29	57.71	40.22	54.49
Na	11	2.94	2.73	2.63	2.48
Al	13	13	10.27	13.42	10.78
Si	14	33.1	25.14	36.98	28.54
K	19	5.25	2.86	4.71	2.61
Ca	20	2.42	1.29	2.04	1.1
Si/Al			2.45		2.65
Na/K			0.95		0.95
Ca/K			0.45		0.42

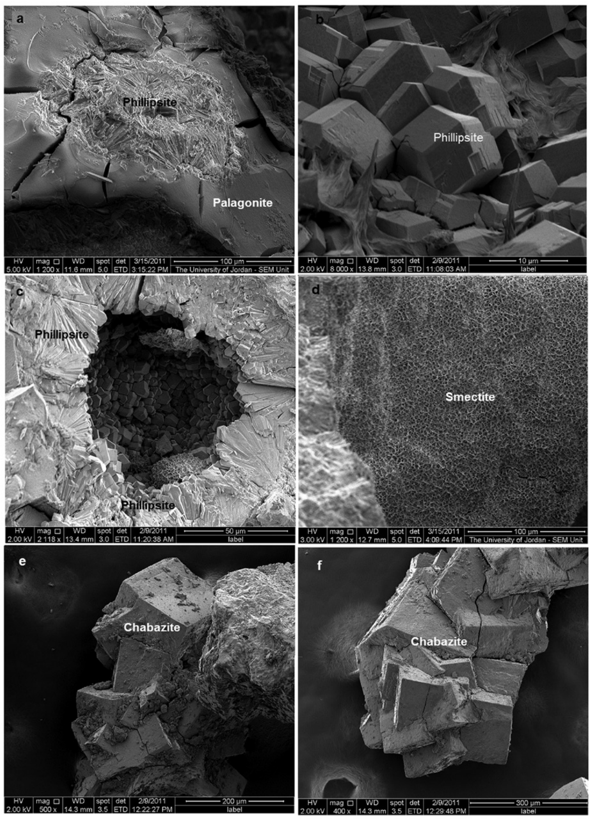

Figure 6: Scanning electron micrograph of the zeolitic tuff (a, b, c): phillipsite, (d): smectite and (e, f): chabazite.

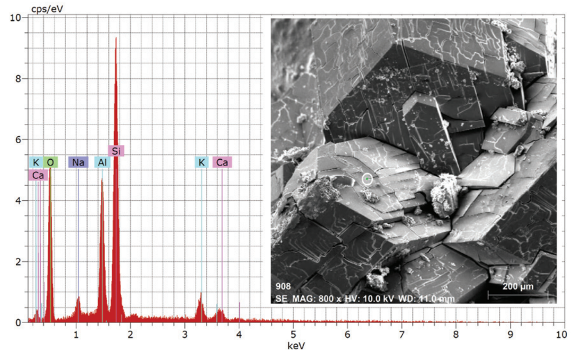

Figure 7: Energy dispersive X-ray analysis of phillipsite crystals.

4.5. Geochemistry

As Illustrated in Table 4, the samples of the zeolitic tuff are characterized by low SiO_2 content with a very wide range of variation. It varies from 21.6% to 40.10% decrease with increasing degree of alteration. The Al_2O_3 wt% in the studied samples is between 7.69% and 12.8%. As shown in Fig 8a and 8b, the Al_2O_3 wt% versus SiO_2 wt% and total alkalis (Na_2O + K_2O wt %) versus SiO_2 wt% display proportional relationships, indicating that they exhibit similar geochemical behavior under the zeolitization process. The studied samples are also typified by high value of LOI, with a very wide range of variation as well, ranging from 10.0 to 30.7 wt%. The inverse relationship between SiO_2 wt% versus LOI content is well illustrated in Fig 8c. This indicates that the samples are highly altered and display several stages of alteration. This is supported by field evidences, including occurrences of abundant zeolites and calcite in the volcanic tuff, occurring as cement in the volcanic clasts. The presence of very high CaO wt% in selected samples up to 24.9% indicates presence of abundant carbonates. Presence of minor amounts of Cl wt% and SO_3 wt% may indicate occurrence of traces of gypsum and halite. The other important constituents of the samples are the Fe_2O_3 and MgO. In detail, the Fe_2O_3 wt% is from 6.26%, to 13.5%, whereas the MgO wt% is from 4.04% to 8.05.

Table 4: Chemical composition of selected zeolitic tuff samples (wt%) by (XRF).

Sample No.	1	5	6	7	8	9	10	17	18
SiO_2	33.40	25.30	21.60	33.90	28.60	29.60	37.60	37.30	40.10
TiO_2	2.09	1.11	1.09	2.15	2.91	1.72	2.09	2.59	2.30
Al_2O_3	10.30	8.07	7.69	11.02	9.23	9.31	12.80	12.80	13.00
Fe_2O_3	11.10	6.26	6.04	11.10	14.80	10.40	11.30	13.50	12.50
MnO	0.13	0.07	0.07	0.13	0.16	0.11	0.13	0.16	0.15
MgO	6.56	4.04	3.31	8.05	7.52	7.84	6.90	6.58	8.47
CaO	8.72	23.20	24.90	6.89	5.11	7.21	8.00	7.21	9.90
Na_2O	1.43	0.13	0.13	1.22	0.88	0.42	1.32	0.85	0.90
K_2O	1.53	0.76	0.43	1.80	2.00	1.82	2.23	1.52	1.91
P_2O_5	0.55	0.15	0.16	0.37	0.71	0.59	0.56	0.67	0.59
SO_3	1.03	0.36	0.16	0.36	0.23	0.14	0.53	0.11	0.11
Cl	0.76	0.04	0.05	0.06	0.92	0.13	0.51	0.03	0.03
LOI	21.80	30.50	34.50	22.70	26.20	30.70	15.80	16.90	10.00

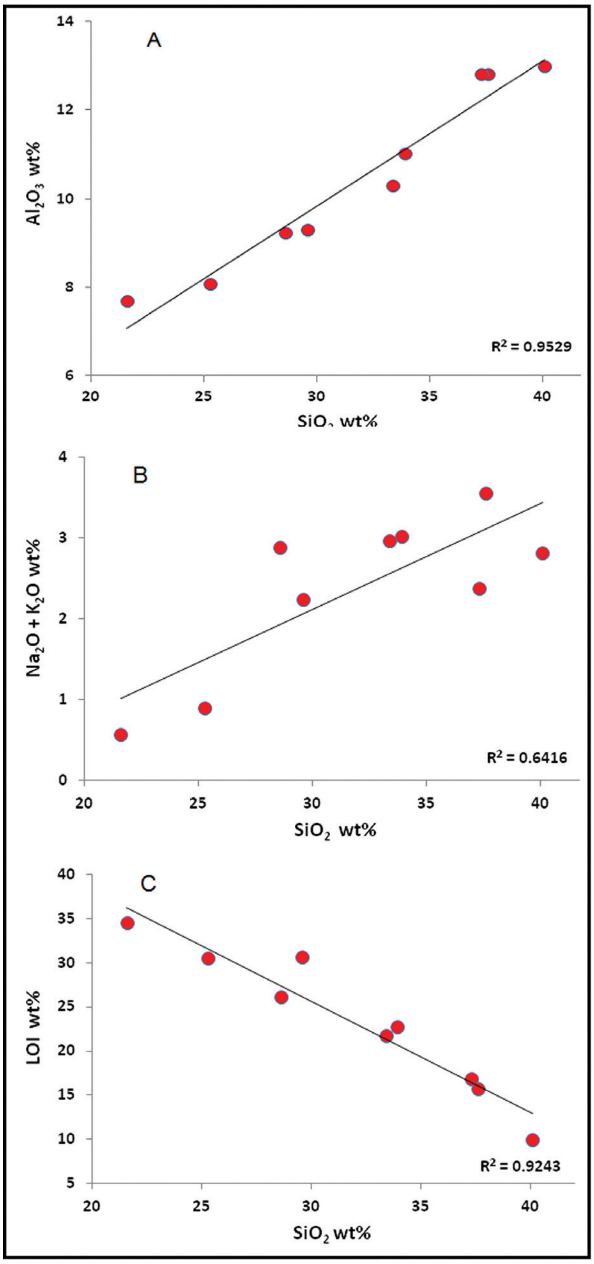

Figure 8: Variation diagram of selected major oxides in the zeolitic tuff, (a): SiO2 vs Al2O3; (b): SiO2 vs (Na2O+K2O) and (c): SiO2 vs LOI.

5. Discussion

The origin of zeolites in Jordan was thoroughly investigated by many authors (Dwairi, 1987; Ibrahim, 1996; Ibrahim & Hall, 1995, 1996; Khoury et al, 2015). The model of zeolites formation in the URS volcano is believed to be the open hydrological system model which was first described by Hay and Iijima (1968a & 1968b) at Koko Crater, Hawaii. This model was already adopted by Ibrahim (1996, Ibrahim and Hall (1995 & 1996) and Khoury et al. (2015).

Following the deposition of the fresh basaltic glass in the volcanic tuff pile, percolating water coming from rainwater interacts with the glass. The interaction of the percolating water with the fresh non-stable basaltic glass in the volcanic tuff increases the pH and salinity of the percolating water with depth. Thus, hydrolysis reaction of the basaltic glass takes place. This leads the volcanic glass to transform to palagonite.

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As a result of the transformation of the fresh basaltic glass to palagonite, smectite and different zeolites minerals start to form. Mass movement of the percolating water downwards through the volcanic tuff pile leads to development of vertical zonation of the zeolites minerals (Hay and Iijima, 1968a & 1968b, Ibrahim, 1996). The elements released from the fresh basaltic glass by hydrolysis reaction including Si, Al, Na, K, Ca and Mg have been re-deposited as authigenic minerals (smectite, zeolites and calcite) forming the intergranular cement and the amygdaloidal texture.

The degree of zeolites formation in the open hydrological system depends mainly in many factors including the chemical and mineralogical composition of the parent material (basaltic glass), chemistry of the percolating water, water /rock ratio, the physical properties of the host-rock including its porosity and permeability, hydraulic /topographic gradient and the climatic conditions including rainfall and temperature. Such conditions influence the chemical and structural nature of the zeolites minerals crystallization (Hay, 1986; Barth-Wirsching and Höller, 1989; De la Villa et al., 2001).

6. Conclusions

Exploration program including detailed geological mapping and drilling was executed in Tulul Unuqar Rustum Ash Shamaliyya volcanic centers of the Tlul Esh Shahba volcanic field in northeast Jordan for the purpose of evaluating the geology and mineralogy of zeolitic tuff. The geological mapping indicates that the most abundant rock type is the scoriaceous tuff and zeolitic deposits. The volcanic tuff contains zeolites occurring as cement and as filling inside the palagonite clasts, making about 30% of the rock. Two zeolites minerals were reported, which are phillipsite and chabazite. Chemical analysis of phillipsite indicates that they are K rich type with narrow variation in Si/Al ratio, and similar Na/K and Ca/K ratios. The zeolitic tuff samples have a wide range of SiO_2 content indicating wide range of degree of alteration which leads to formation of authigenic minerals. The model of zeolites formation is believed in the URS volcano is believed to be the open hydrological system model, where soluble elements released from the fresh basaltic glass by the hydrolysis reaction due to the high pH and saline conditions re-deposited in form of zeolites, smectite and calcite.

Acknowledgemnet

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