# CHARACTERIZATION OF ZEOLITIC TUFF FROM NORTHEAST JORDAN USING BOREHOLES DATA

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ABSTRACT. Zeolite, the hydrate tectosilicate minerals, composed of various minerals such as phillipsite, chabazite, faujasite and analcime. Zeolites have been used for many applications such as drying and purification. Zeolites are distributed throughout Jordan in volcaniclastic and tuff volcanic centers. Several zeolitic deposits were located in northeast areas, mainly in Al-Aritayn, Tlul Al-Shahba, Jibal Hmelan, Tal Rimah, Tell Hasna and other places. These deposits are associated with the Harrat-Asham volcanic activity. The zeolitic tuff is composed of different assemblages of minerals such as phillipsite, faujasite, chabazite and analcime with high amount of iron oxide as hematite, fresh sideromelane, palagonite and smectite. They can be associated mainly with the black and red scoria. For the purpose of this study many boreholes have been drilled for examination of main characteristics of the zeolitic tuff, with aim to get a better understanding about vertical and lateral distribution of these minerals below the surface. To achieved that, many samples from different boreholes have been investigated using XRF, XRD and thin sections. The results revealed huge reserves that can be approved for future detailed exploration and extraction. The Jordanian zeolites are considered to have many important applications such as: ion-exchange (water softening and purification), and in the separation and removal of gases and solvents, in agriculture, and in animal husbandry.

Keywords: Jordan, zeolites, tuff, deposits, reserves.

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Abstract. Zeolite , the hydrate tectosilicate minerals, composed of various minerals such as phillipsite, chabazite, faujasite and analcime. Zeolites have been used for many applications such as drying and purification. Zeolites are distributed throughout Jordan in volcaniclastic and tuff volcanic centers. Several zeolitic deposits were located in northeast areas, mainly in Al-Aritayn, Tlul Al-Shahba, Jibal Hmelan, Tal Rimah, Tell Hasna and other places. These deposits are associated with the Harrat-Asham volcanic activity. The zeolitic tuff is composed of different assemblages of minerals such as phillipsite, faujasite, chabazite and analcime with high amount of iron oxide as hematite, fresh sideromelane, palagonite and smectite. They can be associated mainly with the black and red scoria. For the purpose of this study many boreholes have been drilled for examination of main characteristics of the zeolitic tuff, with aim to get a better understanding about vertical and lateral distribution of these minerals below the surface. To achieved that, many samples from different boreholes have been investigated using XRF, XRD and thin sections. The results revealed huge reserves that can be approved for future detailed exploration and extraction. The Jordanian zeolites are considered to have many important applications such as: ion-exchange (water softening and purification), and in the separation and removal of gases and solvents, in agriculture, and in animal husbandry.

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## Introduction

The new occurrences of the zeolitic tuff deposits are belong to the Cenozoic continental basaltic rocks exposed in northeast Jordan (Fig. 1). Detailed geological mapping of the northeast Jordan was carried out by the Natural Resources Authority since 1988, which allowed to produce several geological maps of 1:50,000, 1:25.000 and 1:250.000. Ibrahim (1993) studied in detail the basaltic province, redefined and subdivided the exposed volcanics of the Harrat Ash Shaam Basaltic Super-Group into five groups: Wisad, Safawi, Asfar, Rimah and Bishriyya. Based on new K-Ar dating of the basalt Tarawneh et al. (2000, 2002), subdivided the Harrat Ash Shaam Basaltic Super-Group into three major phases. The first phase is of Oligocene age 26.0-21.0 Ma. The second phase is of late Miocene (12-8Ma) and the third phase is mostly of Pliocene –

Quaternary (6-<0.5Ma). Authigenic zeolites were first discovered and reported in the Aritayn Formation of northeast Jordan by Dwairi (1987); followed by Malabeh (1993); and Ibrahim (1996; 2004). New occurrences in northeast, central and south Jordan were later reported by Ghrir (1998); Rabba and Tarawneh (2001); Tarawneh (2003 and 2004); Al Dwairi and Sharadqah (2014) and Khuori et al., (2015). This research aims to study the spatial distribution of the new zeolitic tuff occurrences, investigate their mineralogy and chemistry and to understand the vertical and lateral distribution of the zeolite minerals regarding the data from 26 log which were drilled at four localities at Tlul Shahba Sheet/northeast Jordan (Fig. 1). For the exploration of zeolitic tuff the area was subdivided into four localities as the following:

A) The first locality (Mount Hamlan) lies about 12 km from the main road Safawi - Ruwaished within the coordinates: 1204-1207 N and 390-392 E.

- B) The second locality is located to the south of the first locality with coordinates: 1201-1202.5 N and 390-392.5 E.
- C) The third locality (Tlul Hasna) is located of about 3.5 km, north of the Ashgaf area within the coordinates: 1201-1202.5 N and 394.5-396.5 E.
- D) The fourth locality located northwest of the south Ashgaf and lies with coordinates: 1187-1188 N and 395-396.5 E.

The elevation of these localities ranges between 896-988 m ASL, and include volcanic hills of medium height, which are covered by basaltic flows intercalated with different types of pyroclastic materials. Soil is scattered throughout the area with many mudflats sediments. The area is accessible and very optimistic for future exploration and extraction of the ore deposits.



Fig. 1. Simplified geological map of northeast Jordan, arrow showing the sample localities, modified after Ibrahim (2004)

## Methodology

#### Field work

Field work was carried out in the north-eastern regions of the Kingdom for the last years, and it is represented by preparing of geological maps, with scale 1:250.000, 1:50.000 and 1:25.000, companied with petrographic studies, geochemical studies, mineral processing of the ores, and age dating of volcanic rocks. Many samples were collected from several sites. Based on the results of these samples, representative areas were selected for drilling at four localities for zeolitic tuff exploration.

#### **Drilling Program**

Exploration program was carried out by field studies and inspections of samples that indicate the presence of zeolite minerals, which are belong to volcanic tuff in the study area. To cover the whole area and to show the distribution of the zeolite minerals and the depth of zeolitic tuff beneath the surface, the exploration program was carried out by drilling of wells and opening of trenches at four localities. For this exploration program 26 wells were drilled at four sites. The depths of the logs ranged from 3 to 26 meters (Table 1). Total meters that were drilled reached up to 274 m. Most of the excavated core samples have good yield ratios (fig. 3 A,B).

#### **Analytical Methods**

More than 100 samples from boreholes and outcrops were examined by petrographic studies using polarizing microscope, X-ray diffraction and XRF analysis. Some samples also were observed by scanning electron microscope (SEM).

# **Results and Discussion**

Detailed field investigation indicated the presence of the vertical and lateral zonation with respect to the degree of alteration of sideromelane to a reddish-brown colours palagonite with very distinguish zoning in depth at all localities. This zoning was described by Ibrahim (1996) in many localities at northeast Jordan. It displays distinctive yellowish light brown, reddish and black colours. The highly zeolitized areas contain soft and friable highly altered lapilli clasts cemented by coating of zeolite and calcite. In many cases, zeolite minerals

filled the cavities between particles of scoria and volcanic tuff. The exposed thickness of the zeolitic zone ranges from 1 m to 20 m, while from the boreholes data it reaches up to 26m beneath the surface. The contact between zones are sharp and affected by the topography and structure of the area that has been cut by faults and dikes. Diagenesis involves palagonitization of sideromelane and the formation of authigenic minerals including zeolites and calcite. The amount of the former is generally proportional to the amount of palagonite in the volcanic tuff (Hay and lijima, 1968).

### Petrogrphic and Mineralogical studies

More than 100 samples of the zeolitic tuff were studied through polarizing microscope and XRD. It can be argued that the zeolitic tuff composed of assemblages of minerals that can be subdivided into zeolite minerals (phillipsite, chabazite, faujasite and analcime), volcanic glass (palagonite and sideromelane), associated with calcite, Fe-Ti oxides, xenolithes and phenocrysts of olivine, pyroxene (ortho- clinopyroxene) and spinel (fig. 4A).

Phillipsite occurs mainly as colourless, radiating crystal aggregates forming a thin rim on pyroclasts. It is also present as isolated euhedral prisms or as rosettes of radiating and spherulitic crystal form. Crystals are commonly less than 50µm long and rarely as long as 300µm (fig. 4B).

Chabazite occurs in transparent, isolated or clustered, equant, rhombic, crystals with a rhombohedral cleavage, simple penetration twining and zoning. Crystals vary in grain size from several microns up to  $300 \ \mu m$  (fig. 4C).

Faujasite is discovered by Ibrahim and Hall (1995). It occurs in colourless, equant, isotropic, isolated and aggregated crystals between 50  $\mu$ m and 100  $\mu$ m, grows directly on the vesicle walls or sometimes preceded by the smectite phase.

Analcime is very rare in the studied samples from boreholes and is identified by X-ray diffraction in some wells. It comes as euhedral well-formed crystals typical with trapezohedral form with size up to 150  $\mu$ m.

Smectite is present filling the cavities and voids and it occurs in the form of undulated flakes. Smectite crystals range in size between 0.2 $\mu$ m and 2  $\mu$ m. with an average size of about 0.5  $\mu$ m. Smectite form the earliest alteration product of the authigenic minerals. It appears as cloudy very thin rim fringing palagonite clasts and/or the vesicle walls (khoury, et al., 2015).

Calcite which represents the latest phase of authigenic minerals, occurs in the form of rim and blocky cement, filling central parts of vesicles and the intergranular space. Ibrahim and Hall (1995) indicated that authigenic minerals were deposited in the same sequence.

The volcanic tephra comprises massive, poorly cemented lapilli vitric tuff to coarse vitric ash tuff. The cementing material consist essentially of zeolite and calcite. In addition to fresh and palagonitized sideromelane clasts, the tuff contains lesser amounts of crystal of olivine, ortho- and clinopyroxene and spinel. The lithic clasts consist of the following types: vesicular, brown to dark brown, microcrystalline, olivine phyric basalt, and pyroxene olivine phyric basalt; upper crustal inclusions including sandstone, limestone, quartzite, argillite, chert, phosphorite and ultramafic xenoliths including all the spectrum of both the (spinel) peridotite and (spinel) pyroxenite, in addition to garnet pyroxenite and pyroxenite (enstatite) (Tarawneh and Abu Jassar, 1994).

Sideromelane comprises greenish light brown, hypohyaline, texturally uniform and smooth groundmass with fresh olivine phenocrysts or needles. Vesicles and voids are mostly rounded, but with different sizes and coated or filling by calcite or zeolites.

Palagonite is made of a groundmass of reddish brown to yellowish brown and golden red hydrated sideromelane with distinctive hydration polygonal cracks. The palagonite occurs either as smooth or mottled consists of tiny (less than 1 mm) massive, yellow-brown, isotropic ooidal spherulites. The palagonite granules and the vesicles inside always exhibit a darker colored mantle zone of palagonite, named by Dwairi (1987) as rim zone palagonite. Authigenic minerals usually coat both the granules and the vesicles. The chemical composition of palagonite, is rather variable and reflects the degree of the palagonitization. It was noticed by many authors that Fe and Ti content in the palagonite is proportional to the degree of the palagonitization of the sideromelane (Ibrahim, 1996). Lateral and vertical variation can be traced in most of the boreholes and this could be related to the degree of zeolitic processes and the rate of the transformation of the volcanic glass to zeolite minerals. This process is well described according to Ibrahim and Hall (1996) to the order of paragenesis of the principal authigenic minerals in the Aritayn Volcaniclastic is as follows:-

Fresh sideromelane  $\rightarrow$  palagonite  $\rightarrow$  Mg-clay  $\rightarrow$  faujasite  $\rightarrow$  phillipsite  $\rightarrow$  chabazite  $\rightarrow$  calcite.

#### **XRD Results**

For XRD methods, standard sample of 175.5 g from zeolitic tuff was prepared through grain size analysis (Table 1), XRD and point counting method using stereomicroscopic was used, with aim to identifies the percentage of zeolite minerals and other associated minerals (Table 1).

From the results of grain size destribution, it can be concluded that the zeolite minerals can be found in all grain size with different content. The point counting method indicated that the percent of the zeolite minerals ranges between 15 to 50.00%, while by XRD method indicated that the percent of the zeolite minerals ranges between 43.21 and 51.77% as shown in Table 1.

Table 1.

Gra	ain s	ize	dıstrii	butio	n of	the	zeoliti	c tuff	from	Mount	Hami	an

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Sample No.	Grain Size μm	Total wt (gm)	Zeolites, % counting	Calcite % XRD	Zeolites % XRD
PZ1	600	48.3	20-50	7.13	51.77
PZ2	500	35.7	15-50	7.26	46.55
PZ3	150	31.4	20-50	5.30	47.81
PZ4	75	32.9	30-50	9.97	43.21
PZ5	pan	24.5	-	5.17	45.02

More than 88 sample were analyzed using X-ray diffraction method with aim to determine the mineral components. The results of the XRD were used for quantification of zeolite and calcite minerals using the sizing and specific gravity methods of De Gennaro and franko (1979), and Mondale et al. (1988). Pure phillipsite, faujasite and chabazite were separated from samples and used for XRD standard. The XRD results indicated that the zeolite minerals are composed of phillipsite, faujasite and chabasite as shown in Table 2 and Figure 2.

The results of the quantified zeolites and calcite indicated that the percent of phillipsite varies from 18.44 to 44.10%, chabazite from 23.75 to 27.72%, faujasite up to 6.32%, calcite from 5.24 to 22.91% and the pyroclasts from 20.72 to 47.63%. The average percentage of the zeolite minerals in all localities is between 50.00% and 85.48% as shown in Table 3. The high content of zeolites were indicated at locality four.

Sample No.	Ph	He	Cc	Au	F	Fa	Ch	Sm	G
BH1	***	*	*	**	***	**	**	*	-
BH3	**	*	*	**	***	*	**	*	*
BH5	***	*	*	**	***	*	***	*	-
BH7	***	*	*	**	***	*	***	*	-
BH9	***	*	*	***	**	*	***	*	-
BH11	***	*	*	**	***	*	**	*	-
BH21	***	*	**	**	***	*	***	*	-
BH25	*	*	**	***	**	*	***	*	*

#### Table 2.

X-Ray Dilliaction Analyses for representative samples from som
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Ph-Phillipsite, He-Hematite, Cc-Calcite, Au-Augite, F-Feldspar, Fa-Faujasite, Ch: Chabazite, Sm-Smectite, G-Gypsum, Major: \*\*\*, Minor: \*\*, Trace: \*



Fig. 2: X-ray diffractogram of the zeolitic tuff with typical reflections of major zeolite minerals and other secondary minerals

Table 3.

Shows the Zeolite percent at four localities using XRD method

Locality	Zeolites%
First	54.16
Second	52.09
Third	50.00
Fourth	85.48

#### **Chemical Studies**

Chemical analyses of some selected samples from zeolitic tuff are shown in Table 4. More than 88 samples were analyzed from boreholes using XRF methods. The results of chemical analyses demonstrated that the most important components of the main oxides which are belong to zeolitic tuff exclusive of H<sub>2</sub>O, are SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, CaO, Na<sub>2</sub>O and K<sub>2</sub>O (Table 4).The average content of SiO<sub>2</sub> is 47.65%, K<sub>2</sub>O is 1.84%, Na<sub>2</sub>O

is 1.73%, Al<sub>2</sub>O<sub>3</sub> is 12.46%, CaO is 6.07%, MgO is 6.83% and Fe<sub>2</sub>O<sub>3</sub> is 11.99%. The variation of SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio varies between 2.93 and 3.8. The SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio controls the formation of zeolites (Cundy and Cox, 2005; De Silva and Sagoe-Crenstil, 2009). There is slight inverse relationship between CaO and SiO2. This is most probably related to the transformation of the primary sideromelane to palagonite and formation of both zeolities and calcite. The SiO<sub>2</sub> content of the studied samples is confined between 40.77% and 47.90%. There is a distinctive variation in Na<sub>2</sub>O and K<sub>2</sub>O contents and K<sub>2</sub>O/Na<sub>2</sub>O ratio between the different localities. A wide range of K<sub>2</sub>O/Na<sub>2</sub>O ratio occurs in the studied samples that ranges between 0.44 to 1.66%. This most probably is related to two main factors: differences in parental magma, and related to the dominating zeolite phases, whether these phases are Na-rich, K-rich and/or Ca-rich (phillipsite and chabazite) (Khourv. et al.. 2015).

Sample No.	L.O.I%	Fe <sub>2</sub> O <sub>3</sub> %	CaO%	K <sub>2</sub> O%	Na <sub>2</sub> O%	SiO <sub>2</sub> %	Al <sub>2</sub> O <sub>3</sub> %	MgO%
BH1	15.79	12.15	5.75	2.21	1.33	42.68	13.71	5.57
BH3	12.63	13.89	8.59	1.78	1.49	42.45	11.88	6.11
BH5	15.03	12.06	5.13	2.14	1.44	42.94	13.23	6.38
BH7	8.97	15.06	5.16	2.04	1.95	47.63	12.03	6.16
BH9	12.49	13.56	4.50	2.24	1.84	43.69	12.87	6.81
BH11	15.07	10.86	7.48	1.78	2.18	42.42	12.88	5.52
BH20	14.36	10.43	14.79	0.45	1.02	40.77	12.15	4.24
BH21	9.15	13.23	4.13	1.37	2.54	47.90	12.05	7.75
BH22	9.44	12.98	6.28	2.12	2.02	43.72	13.9	7.94
BH23	10.90	13.13	7.50	2.03	1.94	42.32	12.66	8.58
BH8	10.96	13.16	6.11	1.60	1.75	43.55	12.54	8.57
BH6	11.53	14.74	2.9	1.43	1.29	45.89	10.35	8.62
BH11	11.93	14.73	3.90	1.81	1.24	43.66	12.45	8.40
BH25	12.46	12.59	4.21	2.21	2.29	47.52	12.10	6.51
BH4	12.23	12.60	4.62	2.44	2.13	48.21	12.11	5.35

#### Table 4. XRF analyses shows the major oxides of the zeolitic tuff

# **Reserve Estimation**

The estimation of the reserves of zeolites and volcanic tuff was based and measured by the nearest neighbor method assigns grade values to blocks from the nearest sample point to the block composed of polygons each with a unique grade, and with influence area for each borehole, taken into consideration that all layers have lateral distribution, with specific gravity of 1.6 for the ore. From the boreholes data it can be concluded that the thickness of the zeolitic tuff at the first locality varies from 1.5m (BH 1) to 15.85m (BH 6), while the thickness of the overburden ranges between zero level (BH6) up to 6.4m (BH3) (Table 5). The thickness of the zeolitic tuff at the second locality varies from 7.2m (BH 18) to 17. 5m (BH 14), while the thickness of the overburden ranges between zero level (BH14) up to 1.3 m (BH13) (Table 6). The thickness of the zeolitic tuff at the third locality varies from 4m (BH 24) to 26 m (BH 21), while the thickness of the overburden ranges

between zero level (BH24) up to 1.3m (BH26) (Table 7). The thickness of the zeolitic tuff at fourth locality varies from 2m (BH 25) to 5.3 m (BH 26), while the thickness of the overburden ranges between zero level (BH25) up to 2.3 m (BH26) (Table 8).

It can be noticed that the depth of zeolitic tuff in the boreholes at four localities varies between 4.5 to 26 m. Lateral change in thickness can be traced at all boreholes, with some variation in the zoning of the zeolitic tuff that are related to changes in the colure, thickness, zeolite content, clastic materials and the overburden.

It can be assumed that the indicated volcanic tuff reserve estimation reaches up to 9.202.752 metric tons, while the measured zeolites reserves reaches up to 4.818.595 metric tons at all localities.

## Table 5.

# Reserves of the Zeolites and volcanic tuff at first location

BH No	Zeolites %	Volcanic Tuff (m.T)	Volume (m <sup>3</sup> )	Thickness (m)	Area (m <sup>2</sup> )	Zeolites (m.T)
TZ1	48.00	208399.20	130249.50	1.50	86833	100011
TZ2	51.58	118211.68	73882.30	3.10	23833	60973.59
TZ3	54.16	138665.28	86665.80	2.60	33333	75101.12
TZ4	49.34	148534.40	92834.00	2.00	46417	73286.87
TZ5	53.97	3454000.00	2158750.00	10.00	215875	1864123.8
TZ6	60.09	484376.00	3027350.00	15.85	191000	291077.68
TZ7	52.81	882000.00	551250.00	5.00	110250	465784.2
TZ10	51.82	22137.76	13836.10	8.30	1667	11471.344

## Table 6.

# Reserves of the Zeolites and volcanic tuff at second location

BH No	Zeolites %	Volcanic Tuff (m.T)	Volume (m <sup>3</sup> )	Thickness (m)	Area (m <sup>2</sup> )	Zeolites (m.T)
TZ1	55.53	234453.12	146533.20	10.20	14366	130180.09
TZ14	40.05	300076.00	187547.50	17.50	10717	120180.44
TZ15	49.57	2462.40	1539.00	4.50	342	1220.4886
TZ17	49.04	125030.40	78144.00	5.50	14208	61308.657
TZ18	51.28	19258.56	12036.60	2.70	4458	9875.7896
TZ19	59.14	183087.36	114429.60	13.80	8292	108277.86
TZ20	59.40	226000	141250	10.00	14125	134244

## Table 7.

Reserves of the Zeolites and volcanic tuff at third location

BH No	Zeolites %	Volcanic Tuff (m.T)	Volume (m <sup>3</sup> )	Thickness (m)	Area (m <sup>2</sup> )	Zeolites (m.T)
TZ21	48.79	1355452.80	847158.00	26.00	32583.00	661313.1
TZ22	49.94	466127.20	291329.50	11.50	25333.00	232774.6
TZ23	49.90	506676.80	316673.00	19.00	16667.00	252846.2
TZ24	49.21	41068.80	25668.00	4.00	6417.00	20209.96

Table 8.

Reserves of the Zeolites and volcanic tuff at fourth location

BH No	Zeolites %	Volcanic Tuff (m.T)	Volume (m <sup>3</sup> )	Thickness (m)	Area (m²)	Zeolites (m.T)				
TZ25	47.58	167734.40	104834.00	2.00	52417.00	79812.221				
TZ26	54.22	119000.00	74375.00	3.50	21250.00	64521.8				



Fig. 3. Core samples from borehole (A) and close up view of reddish zeolitic tuff (B)  $% \left( A^{\prime}\right) =0$ 

# Conclusions

The results indicated that there is a clear change in the proportions of the major oxides that are associated with zeolitic tuff in all boreholes, as a result of zeolitic process of transformation of glass material to zeolites, with the availability of the physical and chemical conditions to form zeolites minerals. Petrographically, the zeolitic tuff characterized by the presence of zeolite minerals, calcite, volcanic glass and iron oxides. Their presence and distribution in the studied samples from the boreholes is changeable and varies. Lateral and vertical changes can be traced through all localities, that is due to presence of wide variation of the zoning of zeolitic tuff at depth. It was also noticed that the oxides of silicon, aluminum, sodium and potassium ratios are high in the zeolite tuff with high content of zeolite minerals, compared with non-zeolitic tuff contained sidromelane and palagonite. It can be argued that the tuff with more palagonite characterized by decreasing in potassium, sodium and calcium oxides, and increasing in magnesium and iron oxides. It can be noticed also from the chemical results that the richest zeolite levels are characterized by high proportion of potassium oxide.



Fig. 4. Photomicrographs of the zeolitic tuff, showing phillipsite (Ph), Chabazite (Ch) and Palagonite (PI) (XPL,X20)

Results of XRD indicate the presence of zeolite minerals (Phillipsite, Chabazite, Faujasite and partially analcime), associated with calcite, and smectite and iron oxides as secondary minerals. The percent of the zeolite minerals range between 15 to 50.00% using point counting method, while by XRD method the percent of zeolite minerals ranges between 43.21 and 51.77%. It can be concluded that the depth of zeolitic tuff in the borehole is vary between 4.5 and 26 m. Lateral change in thickness can be traced at all boreholes with clear variation of the zoning of the zeolitic tuff which are related to the colure, thickness, zeolite content and the overburden. It can be assumed that the indicated volcanic tuff reserve estimation reaches up to 9.202.752 metric tons, while the measured zeolites reserves reaches up to 4.818.595 metric tons at all localities. The new zeolitic tuff deposits are of commercial value due to the high content of the zeolite minerals, contain large grain size zeolite minerals, relatively poorly lithified and porous. These properties are all favourable for successful beneficiation. Experimental investigations on the zeolitic tuff emphasized the importance of the Jordanian zeolites for use in wastewater treatment plants and as a soil conditioner and as slow-release fertilizer.

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