

## PETROGRAPHY AND ALKALI-REACTIVITY OF SOME VOLCANIC AGGREGATES FROM ICELAND

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### ABSTRACT

Petrographic examinations were made of volcanic aggregates from Iceland, based on microscopy, XRD and EPMA analysis of reactive minerals, to interpret early IBRI data of potential reactivity according to ASTM C227 mortar bar and C289 chemical tests. Basaltic rocks were found non-reactive when consisting of fresh basalt glass, but they can be reactive when they contain rhyolitic interstitial glass, and are even highly reactive when altered to contain secondary opal, besides cristobalite and rhyolitic glass. It was also revealed that volcanic rocks oversaturated with silica contain cristobalite and/or tridymite, and likely present pessimum phenomena. These relationships between the mineralogy and potential reactivity of Icelandic volcanic rocks are remarkably similar to those reported from volcanic aggregates in New Zealand and Japan.

**Keywords:** Alkali-reactivity, petrography, reactive minerals, volcanic rocks

### INTRODUCTION

Concrete aggregates in Iceland are mostly basaltic rocks which contain both alkali-reactive and non-reactive types (Gudmundsson & Asgeirsson 1975, Helgason 1981). However, petrographic methods to distinguish these aggregates with full mineralogical explanations, have not been well established (Thaulow & Olafsson 1983), unlike volcanic aggregates in the circum-Pacific region such as New Zealand and Japan (Katayama & Kaneshige 1986, Katayama et al. 1989).

In order to identify reactive minerals in these Icelandic volcanic rocks, detailed petrographic examinations were made by means of XRD and EPMA, and correlated with results of ASTM C227 & C289 tests obtained earlier at the Icelandic Building Research Institute (IBRI). This paper shows a possibility of mineralogically interpreting the reactivity of volcanic rocks from Iceland, like those done in the circum-Pacific region.

### GEOLOGICAL SETTING

Iceland consists essentially of basaltic rocks belonging to younger geologic ages since the Miocene Tertiary time. Subglacial eruptions during Pleistocene Quaternary produced hyaloclastites, dominated by water-granulated basalt glass called sideromelane (Saemundsson 1979). Sand and gravel deposits used for concrete aggregates in Iceland are rich in these glassy basalts.

These rocks have undergone extensive burial diagenesis intermediate between weathering and regional metamorphism, which alters sideromelane through palagonite into an assemblage of secondary minerals, smectite-zeolite-opal (e.g. Jakobsson 1979). Since this process is common to Japanese volcanic rocks of the same geologic ages (Katayama & Kaneshige 1986), Icelandic volcanic rocks examined were classified here into three alteration zones: the unaltered, the slightly altered and the smectite zones, as defined in Japan (*Table 1*).

## PETROGRAPHY OF VOLCANIC ROCKS

Several rock types representing western and southwestern Iceland, such as basalt, andesite, dacite, both deleterious and non-reactive basaltic sands, were examined petrographically to identify reactive minerals in these rocks. Summarized results are given below concerning the content of silica minerals, determined by microscopy (Table 2) and a phosphoric acid treatment coupled with XRD analysis (Fig. 1), as well as chemical compositions of interstitial glasses and silica minerals analyzed by EPMA (Tables 3, 4).

**Olivine tholeiite:** This is almost a fresh basalt undersaturated with silica, and none of the silica minerals was detected. EPMA analysis revealed that the rock contains less than 5% of rhyolitic interstitial glass with SiO<sub>2</sub> >80% (Fig. 2). Small amounts of iddingsite is found within olivine phenocrysts.

**Tholeiite:** This is a typical tholeiite oversaturated with silica, which is present as cristobalite (Fig. 3) and quartz in the groundmass. It is a well-crystallized basalt containing a trace amount of rhyolitic interstitial glass, along with accompanying smectite of a nontronite variety that replaces the glass (Table 3). Secondary calcite occurs in veins.

**Basaltic andesite:** This rock contains nearly 10% of rhyolitic interstitial glass with SiO<sub>2</sub> of 77%, with a trace amount of cristobalite. The glass has been slightly altered to a disseminated smectite (nontronite).

**Table 1 Geologic origin of Icelandic volcanic rocks**

Geologic age	Rock type	Origin	Alteration zone	Locality
Pleistocene	Olivine tholeiite	Lava flow	Unaltered zone	Lake Leirvogsvatn
Pleistocene	Basaltic sand	Spit*	Unaltered zone /Slightly altered zone	Raudamelur
Pliocene	Tholeiite	Dike	Slightly altered zone	Mt. Brekkufjall
Pliocene	Basaltic andesite	Lava flow	Slightly altered zone	Mt. Raudahnukur
Pliocene /Pleistocene	Basaltic sand	Glacio-marine*	Smectite zone /Unaltered zone	Hvalfjordur
Tertiary	Dacite	Plug	Smectite zone	Stora Borg

\* Hyaloclastite dominates in the Pleistocene parent rock

**Table 2 Petrography of some Icelandic volcanic rocks**

	Chemical			XRD of residue <sup>3)</sup>			Microscopy <sup>4)</sup>									
	SiO <sub>2</sub> <sup>1)</sup>	Q <sub>2</sub> <sup>2)</sup>	Res <sup>3)</sup>	Silica minerals			Silica minerals					Glass				
				cr	tr	qz	cr	tr	op	cd	qz	sd	pg	rh	sm	
Olivine tholeiite	48.0	0	0.0	0.0	0.0	0.0	0	0	0	0	0	0	0	0	3	0
Tholeiite	50.5	6	4.0	2.1	0.0	1.9	1	0	0	0	<1	0	0	2	1	
Basaltic andesite	52.6	6	0.1	0.1	0.0	0.0	<1	0	0	0	0	0	0	9	2	
Dacite	63.5	17	8.1	0.8	3.5	3.8	1	3	0	0	5	0	0	20*	5	
Raudamelur sand			0.0	0.0	0.0	0.0	0	0	0	0	0	15	3	1	0	
Hvalfjordur sand			1.4	0.1	0.0	1.3	<1	0	<1	<1	<1	3	1	1	6	

1) After Fridleifsson (1973) and Franzson (1978)

2) Normative quartz as oversaturated silica

3) Residual silica, extracted by phosphoric acid treatment

4) cr: cristobalite, tr: tridymite, op: opal, cd: chalcedony, qz: quartz  
sd: sideromelane, pg: palagonite, rh: rhyolitic glass, sm: smectite  
\* : devitrified to a cryptocrystalline groundmass

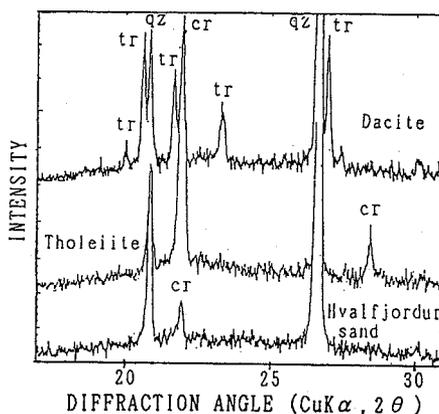
**Dacite:** This is an altered rock containing smectite. About a half of the oversaturated silica is present as silica minerals tridymite, cristobalite and quartz (*Table 2*), but the other half is hidden in a devitrified glass with a crypto- to microcrystalline texture. Tridymite occurs as wedge-shaped crystals, while cristobalite forms imbricated round patches in the groundmass. Some of these primary silica minerals have been inverted to secondary quartz leaving pseudomorphs of their original outlines (*Fig. 4*). Secondary quartz also occurs as irregular-shaped, sponge-like patches in the devitrified groundmass, formed through recrystallization of interstitial glass. The mineral assemblage of smectite-tridymite-cristobalite suggests that the rock has undergone a moderate degree of alteration, comparable to the smectite zone defined in Japan.

**Table 3** Volcanic glasses in Icelandic volcanic rocks

An. no:	Olivine tholeiite		Tholeiite	Basaltic andesite
	Rhy glass 98	Rhy glass 207	Nontro-nite* 209	Rhy glass 116
SiO <sub>2</sub>	80.75	79.75	43.54	77.19
TiO <sub>2</sub>	0.53	0.40	0.19	0.84
Al <sub>2</sub> O <sub>3</sub>	10.38	9.68	4.32	11.55
FeO	1.57	1.79	30.81+	2.35
MnO	0.00	0.03	0.21	0.04
MgO	0.02	0.17	6.66	0.02
CaO	1.47	0.27	1.34	0.68
Na <sub>2</sub> O	3.55	2.93	0.14	1.11
K <sub>2</sub> O	0.68	4.13	0.35	3.65
Total	98.95	99.15	87.56	97.43

\* Altered from glass, + as Fe<sub>2</sub>O<sub>3</sub>

**Fig. 1** XRD patterns of silica minerals extracted



**Fig. 2** Rhyolitic interstitial glass in olivine tholeiite



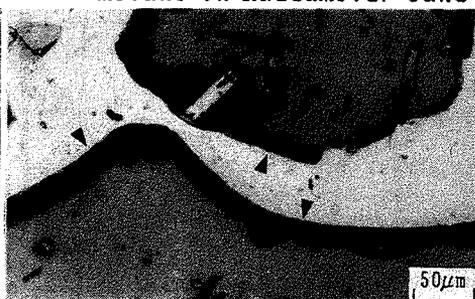
**Fig. 3** Interstitial cristobalite in tholeiite



**Fig. 4** Pseudomorphic quartz after tridymite in dacite



**Fig. 5** Palagonite rim to sideromelane in Raudamelur sand



**Raudamelur sand:** This sand, roughly a fresh basaltic hyaloclastite, has been used as a concrete aggregate for over two decades with good results. EPMA analysis revealed that a full range of interstitial glasses from basaltic to rhyolitic compositions are present in the basalt grains, depending on their crystallinity (*Table 4*). The sand with a grain size 0.6–1.2mm, consists of fresh basalt (55%), basalt glass (sideromelane, 30%), altered basalt glass (palagonite, 10%), plagioclase, olivine and other rock fragments. Sideromelane represents average bulk compositions of fresh basalt, while a palagonite rim to sideromelane indicates an initial alteration of this glass (*Fig.5*).

**Hvalfjordur sand:** This was a sea-dredged aggregate used as a sole sand source for concrete in Reykjavik from the 1960's to 1970's, usually unwashed. Extensive damage to concrete houses was caused by this aggregate (Olafsson 1989). It is an altered basalt containing smectite, and the fraction of 0.6–1.2mm consists of altered basalt (80%), fresh basalt (10%), rhyolite, andesite, basalt glass (sideromelane), secondary minerals and shell fragments.

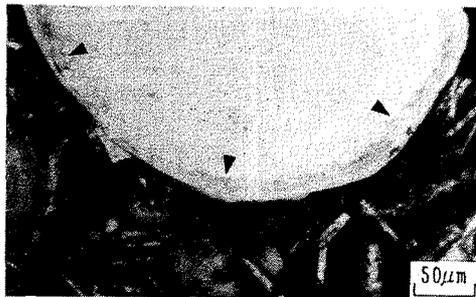
Vesicles of altered basalt are often filled with smectite (nontronite), forming amygdale textures, or occasionally lined with secondary opal (*Fig.6*) or chalcedony. In the altered basalt, cristobalite occurs as rounded patches, surrounded by smectite that replaces interstitial glass (*Fig.7*), and zeolite (alkali-heulandite, *Table 4*) may also be present. The mineral assemblage of opal-cristobalite-smectite-zeolite is common to moderately altered volcanic rocks from Miocene to Pliocene Tertiary in Japan, defined as the smectite zone (Katayama & Kaneshige 1986). Secondary opal and smectite are also common to a deleterious Tertiary basalt from Queensland, Australia (Shayan & Quick 1988).

**Table 4 EPMA compositions of mineral phases in the basaltic sands**

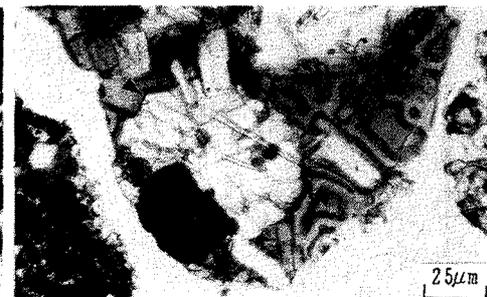
An. no:	Raudamelur sand					Hvalfjordur sand						
	Fresh basalts		Alt. bas			Fresh basalts			Altered basalts			
	Unaltered zone	Dac	Rhy	Pala-	Slt. alt. z.	Unaltered zone	Unaltered zone	Smectite zone	Opal	Non-	Heu-	
	Bas glass	And glass	Dac glass	Rhy glass	Pala-gonite	Bas glass	And glass	Rhy glass	Cristo-balite	Opal	Non-tron.	Heu-land.
140*	50.09	54.42	67.35	70.53	33.09	49.82	57.11	80.06	97.09	95.7	50.44	63.96
133	1.54	2.67	0.42	2.12	2.81	1.08	2.49	0.74	0.09	1.3	0.23	0.03
138	13.15	10.53	10.24	11.71	26.94	13.88	9.30	9.98	2.00	0.3	8.65	18.07
128	11.20	11.59	12.36	6.93	19.28	11.13	16.19	1.51	0.07	2.2	21.67+	0.24
202	0.19	0.18	0.00	0.17	0.20	0.23	0.43	0.02	0.00	0.0	0.02	0.01
	8.08	3.05	0.33	0.03	0.45	8.58	3.26	0.00	0.00	0.3	3.49	0.09
	12.17	13.69	3.36	4.83	0.94	13.11	7.96	0.25	0.07	0.1	0.63	0.96
	1.99	0.79	1.94	2.25	0.10	1.89	1.28	1.28	0.36	0.0	0.38	2.38
	0.37	0.53	1.84	0.72	0.16	0.21	1.41	4.46	0.02	0.2	2.82	4.39
Total	98.78	97.44	97.83	99.30	83.97	99.92	99.41	98.30	99.70	100.0	88.32	90.11

\* Sideromelane, \*\* total normalized to 100%, \*\*\* altered from glass, + as Fe<sub>2</sub>O<sub>3</sub>

**Fig.6 Opal in altered basalt in Hvalfjordur sand**



**Fig.7 Cristobalite surrounded by smectite in Hvalfjordur sand**



## POTENTIAL REACTIVITY OF ICELANDIC VOLCANIC ROCKS

The results of the ASTM C289 and ASTM C227 tests of Icelandic volcanic rocks performed at IBRI (e.g. Helgason 1981), are shown in *Table 5* and *Figs. 8 & 9*.

**Chemical test:** Rocks containing rhyolitic glass (olivine tholeiite & basaltic andesite) with low dissolved silica (Sc) & low reduction in alkalinity (Rc), classify as deleterious, whereas rocks with silica minerals cristobalite and tridymite (dacite, tholeiite & Hvalfjordur sand) gave higher Sc & Rc in the potentially deleterious field, or near the boundary. These tendencies are similar to volcanic rocks of the same geologic ages in Japan & New Zealand (Katayama et al. 1989). Experiences show that Icelandic deleterious volcanic rocks are often misjudged as innocuous in this test, due to unreliably high values of Rc, so that only values of Sc > 100 mmol/l have been used as a deleterious indication (Thaulow & Olafsson 1983). Smectite in altered rocks is probably the cause of misjudgement of marginally reactive rocks as innocuous, because this mineral raises Rc values by cation exchange (Morino et al. 1987).

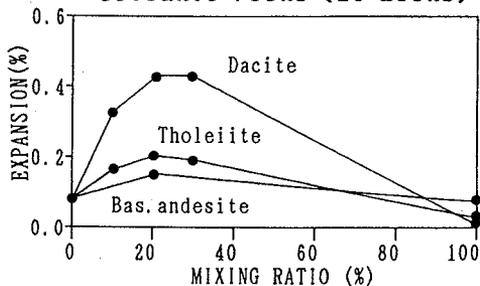
**Mortar bar test:** Olivine tholeiite tested as innocuous in the ASTM C227 at 26 weeks, and was mixed as 'inert' aggregate to examine pessimum phenomena of other rock types (*Table 5*). Rocks containing cristobalite and/or tridymite (dacite & tholeiite) exhibited a marked pessimum proportion at 20% mixing, whereas basaltic andesite containing mainly rhyolitic interstitial glass, did not show such a marked pessimum (*Figs. 8, 9*). Tridymite and cristobalite are considered responsible for the pessimum phenomena of some Icelandic volcanic rocks, which is similar to the experiences in Japan and New Zealand (Katayama et al. 1989). The olivine tholeiite is marginally reactive at one year (0.095%/52 weeks), if judged by a criterion (0.1%/1 year) by Mielenz et al (1947) that divides the innocuous & deleterious fields in the ASTM C289. As discussed later, the reactivity of this rock is attributable to the rhyolitic glass.

**Table 5 Potential reactivity of Icelandic volcanic rocks**

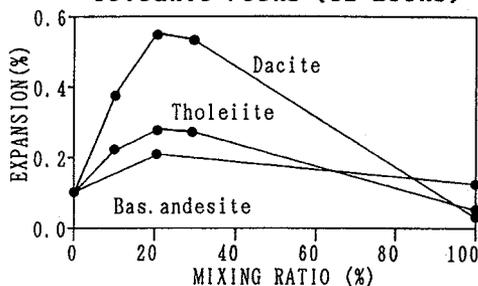
Rock	Reactive phase	ASTM C289 <sup>1)</sup>		ASTM C227 (26w) <sup>1)</sup>				ASTM C227 (52w) <sup>1)</sup>			
		Sc	Rc (mmol/l)	10%	20%	30%	100%	10%	20%	30%	100%
Olivine tholeiite	Rhy. gl	86	18 D	-	-	-	0.08 I	-	-	-	0.10 (D)
Raudamelur sand	(Bas. gl)	96	91 PD	-	-	-	-	-	-	-	0.04 I
Basaltic andesite	Rhy. gl, cr	167	151 D	-	0.14	-	0.07 D	-	0.20	-	0.12 D
Hvalfjordur sand	Cr, opal	204	179 PD	-	-	-	-	-	-	-	0.3-0.5D
Tholeiite	Cr	268	272 (I)	0.17	0.20	0.19	0.02 D	0.21	0.27	0.26	0.04 D
Dacite	Tr, cr	340	320 PD	0.33	0.43	0.43	0.01 D	0.38	0.55	0.54	0.03 D

<sup>1)</sup> IBRI data, using high-alkali cement; gl:glass, cr:cristobalite, tr:tridymite D:deleterious, PD:potentially deleterious, I:innocuous

**Fig. 8 Pessimum phenomena of volcanic rocks (26 weeks)**



**Fig. 9 Pessimum phenomena of volcanic rocks (52 weeks)**



## REVIEW OF POTENTIAL REACTIVITY OF REACTIVE MINERALS

A literature review was made on the potential reactivity of silica minerals and volcanic glasses (Table 6), to support interpretations of the results of the ASTM chemical test and ASTM mortar bar test of Icelandic rocks.

**Silica minerals:** References show that silica minerals have marked pessimum proportions, which tend to decrease with increasing the potential reactivity, i.e. opal, cristobalite, tridymite and chalcedony present pessimum proportions from less than 5% to 20% in the mortar bar test, and occupy the potentially deleterious field in the chemical test. Cryptocrystalline quartz and microcrystalline quartz are less reactive and their pessimum ranges from about 50% to 80%, or possibly 100%. The boundary line between the potentially deleterious field and the deleterious field in the ASTM C289 was presumably drawn at a pessimum proportion of 50%, based on the data by Mielenz et al. (1947) and Chaiken and Halstead (1959).

**Volcanic glasses:** Early American data suggest that obsidians, a natural rhyolitic glass, have pessimum proportions (Mielenz et al. 1947). However, it is necessary to check the presence of cristobalite and tridymite in these glassy rocks by a XRD analysis, because these minerals are often hidden in the obsidians and likely missed under the microscope. Nowadays, volcanic glasses are not considered to exhibit pessimum phenomena unless silica minerals such as cristobalite and tridymite are present (e.g. CCANZ 1991). Fresh volcanic glasses become particularly reactive when their SiO<sub>2</sub> content exceeds 65%, i.e. dacitic composition (Katayama et al. 1989, Katayama 1992).

For example, deleterious Egmont andesite in New Zealand containing dacitic glass (SiO<sub>2</sub> 65.4%, Katayama et al. 1989) and a glassy bronzite andesite in Japan containing rhyolitic glass (SiO<sub>2</sub> 78.0%), did not show a pessimum (StJohn 1988, Yoshioka et al. 1985). By contrast, less glassy bronzite andesite from Japan, with glass plus cristobalite, showed a pessimum proportion between 20 and 40% (Yoshioka et al. 1985). Likewise, an altered basalt from Queensland, Australia, containing dacitic glass (SiO<sub>2</sub> 67.3%) plus secondary opal and smectite (nontronite), showed a pessimum (Shayan & Quick 1988). Thus, rhyolitic glass in the Icelandic rocks is not believed to show a pessimum phenomenon.

**Table 6 Alkali reactivity of silica minerals and volcanic glasses**

Reactive mineral	ASTM C289 chemical test		ASTM C227 mortar bar test	
	Sc(mmol/l)	Result Material	Pessimum	Material Result
<b>Silica minerals</b>				
Opal	>1000	PD Opal(1,2)	< 5%	Opal(1), opaline chert(3) D
Cristobalite	500-1000	PD Synthetic(4)	<10%	Syn. (5), andesite(6)*** D
Tridymite	500-1000	PD Andesite(4)*	<10%	Syn. (5), andesite(6)*** D
Chalcedony	500-1000	PD (1, 2, 7)	20%	Chalcedony(1,7) D
Cryptocrystal. qz	150-300	PD/D Chert(2,8)**	50%	Chert(8,9)** D
Microcrystal. qz	70-150	D Chert(2)	>80%	Miscellaneous(10)**** D
<b>Volcanic glasses</b>				
Rhyolitic(SiO <sub>2</sub> >70%)	70-90	D Synthetic(4)	100%	Andesite(gl. SiO <sub>2</sub> 78.0%)(11)D
Dacitic (62-70%)	70-90	D Synthetic(4)	100%	Andesite(gl. SiO <sub>2</sub> 65.4%)(12)D
Andesitic (52-62%)	30-50	I/D Synthetic(4)		Syn. gl(SiO <sub>2</sub> 61.8%)(13) (D)
				Basalt(gl. SiO <sub>2</sub> 52.1%)(4) I
Basaltic (<52%)	15-30	I Synthetic(4)		Glassy basalt(1) I

\* Mixed with cristobalite, \*\* mixed with chalcedony

\*\*\* JIS A 5308 mortar bar test, \*\*\*\*CSA A 23.2-25A accelerated mortar bar test

(1)Mielenz et al.(1947), (2)Katayama & Futagawa(1989), (3)Stanton et al.(1942)

(4)Katayama et al.(1989), (5)Gaskin et al.(1955), (6)Ishida et al.(1988)

(7)Andriolo & Sgarboza(1986), (8)Morino(1989), (9)Goldman & Klein(1959)

(10)Grattan-Bellew(1992), (11)Yoshioka et al.(1985), (12)StJohn(1988)

(13)Stanton(1942), D:deleterious, PD:potentially deleterious, I:innocuous

## CONCLUDING REMARKS

From the foregoing considerations on the geology, petrology and potential reactivity of Icelandic volcanic rocks, along with the information from reviewed references, the following points can be summarized.

- Icelandic volcanic rocks have undergone stratigraphic alterations due to burial diagenesis, in a similar mode to that of Japanese volcanic rocks of the same geologic ages (Neogene Tertiary & Quaternary). The rocks examined can be classified into the unaltered, the slightly altered, and the smectite zones from upper to lower horizons.
- Deleterious basaltic sand came from altered strata of the Neogene Tertiary, belonging to the smectite zone. The high reactivity of this sand is due to the presence of secondary opal and chalcedony, combined with primary cristobalite and rhyolitic glass. Smectite in the altered rocks may give an unreliable result in the chemical test, causing high Rc and a misjudgement of potentially deleterious rocks as innocuous.
- Basaltic rock is usually non-reactive when it is glassy and contains fresh basaltic glass, but may show a potential reactivity when it is highly crystalline and contains rhyolitic interstitial glass as a residual melt. Rocks containing rhyolitic glass tend to test as deleterious in the chemical test, which suggests little possibility for these rocks to show a pessimum phenomenon in the mortar bar test.
- Volcanic rocks oversaturated with silica often contain the reactive silica minerals cristobalite and tridymite. They show high values of Sc & Rc in the chemical test, and exhibit a marked pessimum phenomenon. All these tendencies are common to volcanic aggregates of the same geologic ages in the circum-Pacific region, such as New Zealand and Japan.

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