Mössbauer spectroscopy of samples from the 2010 Fimmvörðuháls/Eyjafjallajökull eruption

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Abstract Mössbauer spectra of samples from the 2010 Fimmvörðuháls/Eyjafjallajökull eruption are presented with determinations of the $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratios. Mössbauer spectra of time series of samples from the Eyjafjallajökull eruption show a change in the characteristics of the erupted material mid-way in the eruption, suggesting changing access of water to the eruption.

Keywords Mössbauer spectroscopy · Volcanic ash · Eyjafjallajökull eruption 2010 · Fimmvörðuháls eruption 2010

1 Introduction

The 2010 Eyjafjallajökull summit eruption (Iceland) produced large amounts of fine ash, disturbing air traffic across the North-Atlantic and within much of Europe (see e.g. [1]). The eruption phase started with a flank eruption on the 20th of March 2010 at Fimmvörðuháls, a glacier-free area between Eyjafjallajökull and Mýrdalsjökull. The expelled magma was olivine- and plagioclase-bearing mildly-alkaline basalt that

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Table 1 Eruption phases of the Fimmvörðuháls/Eyjafjallajökull eruptions 2010

Eruption phase	Dates	Description
0	20/3/2010–12/4/2010	Flank eruption at Fimmvörðuháls
1	14/4/2010–17/4/2010	First explosive phase at Eyjafjallajökull
2	18/4/2010–3/5/2010	Declining activity, lava flow at Eyjafjallajökull, coarse grained tephra erupted
3	4/5/2010–17/5/2010	Second explosive phase, new magma seems to have entered the system at 2–3/5/2010
4	18/5/2010–22/5/2010	Declining explosive activity at Eyjafjallajökull

This sub-division is given by papers in [1] and the Fimmvörðuháls eruption is added as phase 0

exhibits uniform and rather primitive whole-rock composition ($\text{SiO}_2 \sim 46\text{--}47$ wt.%) [2]. This eruption phase ended on 12th of April 2010.

On the 14th of April, 2010 a summit eruption started at the Eyjafjallajökull stratovolcano of benmoreitic to trachytic composition ($\text{SiO}_2 \sim 56$ wt.%) [3]. The summit eruption has been subdivided into four phases (Table 1).

Geochemical studies of the erupted material from the Eyjafjallajökull eruption [3, 4] shows mixing and mingling of two components, an evolved FeTi-basalt, derived from material similar in composition as that erupted at Fimmvörðuháls and remobilized silicic melt with composition similar to that produced by the 1821–1823 AD Eyjafjallajökull summit eruption ($\text{SiO}_2 \sim 69$ % [5]). Both the end-member-compositions and their mixing ratios changed with time, and mingling of the two was rapid and incomplete such that the two components are often preserved on a micrometre scale in samples obtained from the eruptions.

In this contribution we present Mössbauer spectra of samples from the eruptions and determine their $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratios.

2 Samples and experimental

Two tephra samples from the Fimmvörðuháls eruption are used in this study, labelled according to the collection date 22/3 2010 (coll. Þ. Kristjánsson) and 24/3 2010 (coll. Á. Höskuldsson). These samples are compared to a rock sample from the lava flow at Fimmvörðuháls (coll. 24/3 2010, Á. Höskuldsson).

Ten tephra samples from the Eyjafjallajökull eruption were used and also labelled after eruption date in 2010; 13/4, 14/4, 22/4 (a), 3/5, 7/5, 14/5, 20/5 (coll. Á. Höskuldsson and G. Larsen), 17/4 (coll. J. Merrison) and 22/4 (b) and 27/4 are samples EJ-3 and EJ-4 from [3]. All samples except the sample from 22/4 (a) exhibit a broad grain-size distribution and are brownish in appearance. The 22/4 (a) sample is darker coarse-grained material.

Samples of a lava bomb from the Eyjafjallajökull eruption were also studied. The sample was collected after the eruption, and is thought to be from the latest stages of the eruption (Á. Höskuldsson, private communication, 2011).

A sample from the 1821–1823 AD Eyjafjallajökull eruption was also measured for comparison.

The Mössbauer spectra presented here were measured at room temperature in transmission mode using a conventional constant acceleration drive system and 5–25 mCi $^{57}\text{Co}:\text{Rh}$ sources. Velocities and isomer shifts are given relative to the centre of the spectrum of $\alpha\text{-Fe}$ at room temperature.

Fig. 1 Mössbauer spectra of tephra samples from Fimmvörðuháls. Solid lines show the fitting components indicated with a bar diagram at the top and their sum (black line)

3 Results

3.1 Samples from the Fimmvörðuháls eruption

Figure 1 shows Mössbauer spectra of tephra samples from the Fimmvörðuháls eruption. The spectra were analysed in terms of five components, Fe(II) assigned to Fe^{2+} in glass, Fe(III) assigned to Fe^{3+} in glass, Ilm assigned to Fe^{2+} in ilmenite (FeTiO_3), Ol assigned to Fe^{2+} in olivine ($(\text{Fe}, \text{Mg})_2\text{SiO}_4$) and a broad magnetic hyperfine field distribution assigned to iron-titanium oxide (not shown in Fig. 1). The presence of this last component is barely seen in the spectra, but evidenced from lower transmission in the wings (-8 to -2 mm/s and 5 to 8 mm/s) compared to the background (< -9 mm/s and > 9 mm/s). The signal is too small to firmly identify this component, but in view of the overall relatively reduced state of the samples, it was assumed to be titanomagnetite and was simulated with average parameters of titanomagnetite ($\text{Fe}_{3-x}\text{Ti}_x\text{O}_4$) with $x = 0.6$ [6]. Doublet components were analysed with Voigt profiles, a convolution between a Lorentzian profile of full width at half maximum of Γ and Gaussian profile with standard deviation of σ . For Fe(II) the two legs of the quadrupole doublet had the same area, but different Gaussian broadenings. The spectral fraction of ilmenite (Ilm) was too small to allow its hyperfine parameters to be included as fitting variables, and therefore table values from the literature were used [7]. Average hyperfine parameters and spectral areas are gathered in Table 2.

Overall, the hyperfine parameters obtained are in good agreement with their assignment. The parameters obtained for Fe(II) are in good agreement with

Table 2 Hyperfine parameters and spectral areas obtained from the analysis of tephra samples from Fimmvörðuháls (Fig. 1)

	Fe-Ox ^a	Ol	Fe(II)	Fe(III)	Ilm
B_{hf} (T)	25				
δ (mm/s)	0.77	1.14(1)	1.05(5)	0.36(3)	1.06
$\Delta E_Q, 2\varepsilon$ (mm/s)	0	2.94(1)	1.98(9)	1.14(4)	0.68
Γ (mm/s)	0.45	0.27(2)			
σ_L (mm/s)		0.0(1)	0.23(1)	0.28(2)	0.1
σ_R (mm/s)			0.30(1)		
Area 22/3/2010 (%)	5(2)	16(2)	61(3)	14(1)	3(1)
Area 24/3/2010 (%)	4(1)	20(2)	59(3)	13(1)	4(1)

^aAverage hyperfine parameters of a distribution

The table lists the average magnetic hyperfine field (B_{hf}), isomer shift (δ), quadrupole splitting of doublets (ΔE_Q) and quadrupole shift (2ε) for Fe-Ox, and line-width parameters. Numbers in parentheses represent the error in the last digit, values with omitted errors were fixed and not included as fitting variables

Fig. 2 Mössbauer spectra of a rock sample from Fimmvörðuháls. The upper spectrum marked “Rock” represents the whole-rock sample and the lower spectrum marked “M-Rock” is that of a magnetic separate of a crushed fraction of the whole-rock sample. The *solid lines* show the fitting components indicated with bar diagram on top and their sum (*black line*)

average parameters of Fe^{2+} in glass from [8], suggesting that crystallisation of pyroxene before quenching of the melt has been negligible. There is no significant difference in the spectral areas of components in the two samples. Using 20(2) % higher f -factor of ferric Fe relative to ferrous Fe [9], $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio of the melt (using only Fe(III) and Fe(II)) is 15.6(6) % and 11.9(5) % for the whole rock.

A Mössbauer spectrum of a solidified rock sample from the Fimmvörðuháls eruption is presented in Fig. 2.

Fig. 3 Room-temperature Mössbauer spectra of samples from the Eyjafjallajökull eruption obtained at the dates indicated. *Solid lines* show the fitting components and their sum

The main difference between the rock sample and the tephra samples is the presence of magnetic sextets due to hematite (Ht) and Fe-Ti oxide (Fe-Ox) with parameters consistent with oxidized titanomagnetite. The area fraction of Fe^{3+} species in this sample is $\sim 41\%$ compared with $\sim 15\%$ of the tephra samples. This indicates that the magma oxidized when flowing over a wet (snowy) surface away from the crater. The rock sample would give a quite different oxidation state of the magma than the tephra samples, illustrating that care has to be taken upon interpretation of oxidation states.

3.2 Time series of samples from the Eyjafjallajökull eruption

Figure 3 shows Mössbauer spectra of samples acquired on different dates from the Eyjafjallajökull eruption. The spectra have been analysed in the same way as the spectra of the tephra samples from Fimmvörðuháls. The hyperfine parameters for Ol, Fe(III) and Fe-Ox were taken as global variables, for Ilm, literature values were used. Only the hyperfine parameters of the dominant Fe(II) component were allowed to vary between measurements. Table 3 lists the global hyperfine parameters and Table 4 lists the hyperfine parameters of Fe(II).

The isomer-shift of Ol seems slightly low compared to usual isomer-shifts of Fe in olivine (usually $\delta = 1.16(1)$ mm/s [7]). The reason for this apparent discrepancy is not known but a possibility is that this is due to trace foreign elements in the

Table 3 Global hyperfine parameters used in the analysis of the spectra in Fig. 3

	Fe-Ox ^a	Ol	Fe(III)	Ilm
B_{hf} (T)	32(2)			
δ (mm/s)	0.53(5)	1.135(3)	0.38(2)	1.06
ΔE_Q (mm/s)	0	2.892(6)	0.97(3)	0.68
σ (mm/s)		0.055(6)	0.251(5)	0.1

Values in parentheses represent the one sigma error in the last digit. Values with omitted errors were fixed and not included as fitting variables

^aAverage hyperfine parameters of a distribution

Table 4 Hyperfine parameters of Fe(II) obtained from the analysis of the spectra in Fig. 3

Date 2010	δ (mm/s)	ΔE_Q (mm/s)	σ_L (mm/s)	σ_R (mm/s)
14/04	1.079(3)	2.001(7)	0.205(5)	0.259(6)
15/04	1.092(4)	2.16(2)	0.216(7)	0.265(8)
17/04	1.071(4)	2.045(7)	0.222(5)	0.275(4)
22/04 (a)	1.077(4)	2.13(2)	0.352(6)	0.428(8)
22/04 (b)	1.059(6)	1.99(3)	0.24(2)	0.28(2)
27/04	1.089(6)	1.97(2)	0.20(2)	0.25(2)
03/05	1.094(4)	1.94(2)	0.184(8)	0.247(9)
07/05	1.099(5)	1.95(2)	0.179(9)	0.23(2)
14/05	1.101(5)	1.99(2)	0.19(2)	0.24(2)
22/05	1.099(3)	1.992(8)	0.176(6)	0.250(7)

structure (see supporting information of [4]). The average hyperfine parameters of Fe-Ox suggest oxidized titanomagnetite [6], and in $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ determinations it is assumed that 67 ± 5 % of the Fe-Ox component is due to ferric Fe.

The hyperfine parameters of Fe(II) show significant variation, which is difficult to relate to area fractions or eruption phases. They deviate in all cases from ideal glass parameters (see e.g. Table 2 or [9]) suggesting that the Fe(II) component is due to (at least) both Fe^{2+} in glass and (ortho/clino) pyroxenes in varying amounts. This means also that only whole-rock $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratios are reliable as it is not possible to distinguish the melt/glass component with certainty from crystalline minerals.

The time-dependence of spectral area fractions are shown in Fig. 4. The most clear change during the eruption is in the spectral fractions of the materials erupted between 22/4 and 27/4 2010. This is seen as an increased fraction of Fe-Ox and Ol, and a reduced fraction of Fe(II).

All samples from the Eyjafjallajökull eruption show the presence of Fe^{2+} in olivine. This can be regarded as a surprise; taking the average whole-rock analysis [3] with the $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratios found here; neither CIPW norm [10] nor MELTS calculations [11, 12] suggest that olivine should be found. This result is, however, in agreement with the observations of [3, 4] that the two components that make up the melts from Eyjafjallajökull eruption, did not fully mix to form a homogeneous melt. In that case, the olivine is a part of the mafic component.

The $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio, evaluated under the assumption that the f -factor of ferric Fe is 20(2) % higher than that of ferrous Fe is shown in Fig. 5. The ratio shows significant variations, from 21.5(16) % for the 22/4 (a) sample to 33.8(26) % for the 27/4 sample. An accurate determination of $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio is somewhat hampered by the relatively large error assigned to the ferric proportion of the Fe-Ox component, although the spectral fractions are determined more accurately. The Mössbauer

Fig. 4 Area fraction of the fitting components used for the analysis of the spectra in Fig. 3. The bar diagram at the top refers to the eruption phases outlined in Table 1.

Fig. 5 $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio obtained from Mössbauer measurements of a time series of samples from the Eyjafjallajökull eruption

spectrum of the most reduced sample 22/4 (a) is very distinctive from the others as seen for example in the line-width of the Fe(II) component (Table 4). To study this sample in greater detail, a major element analysis was performed.

Table 5 lists new whole-rock analyses of the 17/4 and 22/4 (a) samples compared with analyses available in the literature for Fimmvörðuháls samples and glass from tephra from the 1821–1823 AD Eyjafjallajökull eruption.

The analysis of the 17/4 sample is in rough agreement with average analyses from from the work of Sigmarsson et al. [3] of samples from the Eyjafjallajökull eruption.

Table 5 Major element analysis of samples from the Fimmvörðuháls/Eyjafjallajökull eruption

	FVH	22/4 (a)	17/4	1821-3
SiO ₂	46.3(4)	56.35	57.2(6)	69.13
TiO ₂	2.9(2)	1.75	1.72(8)	0.36
Al ₂ O ₃	15.0(2)	14.99	14.8(2)	14.1
FeO	10.6(4) ^a	8.57 ^a	7.0(3) ^a	4.29 ^b
Fe ₂ O ₃	1.60(9) ^a	2.16 ^a	2.7(2) ^a	
MnO	0.182(6)	0.25	0.24(1)	0.15
MgO	8.6(4)	4.86	2.6(3)	0.13
CaO	10.0(2)	6.16	5.4(3)	1.1
Na ₂ O	2.9(2)	4.57	5.15(7)	6.14
K ₂ O	0.61(5)	1.68	1.9(1)	3.5
P ₂ O ₅	0.41(2)	0.34	0.38(2)	
Sum	99.2	101.33	99.02	98.91

FVH = Fimmvörðuháls-data from [3], numbers in parentheses represent the standard deviation of 4 samples; 17/4 = average of 6 measurements, numbers in parentheses represent the standard deviation; 1821-3 = data from [5] on glass chemistry of Eyjafjallajökull 1821–1823 tephra

^aFeO and Fe₂O₃ calculated from Fe³⁺/Fe_{Tot} ratios determined here

^bTotal iron expressed as FeO

The chemistry of the 22/4 (a) sample is different, and specifically the differences in MgO and CaO content are of interest in view of the mixing model of Sigmarsson et al., (Fig. 3a in [3]). The CaO and MgO content suggests a larger amount of the mafic component compared to other samples from the eruption. It is therefore concluded that at least part of the variation presented in Fig. 5 is due to different mixing of the two end-members making up the eruptive materials from the Eyjafjallajökull eruption.

However, before firm conclusions on the Fe³⁺/Fe_{Tot} ratio can be reached, alternative explanations of the high oxidation state have to be explored. It was evident from the analysis of the rock samples from Fimmvörðuháls (Fig. 2) that the amount of ferric Fe can be due to secondary oxidation, and similar effects could be invoked here as an explanation of the high Fe³⁺/Fe_{Tot} ratios.

The obvious candidate explanation for raising the oxidation state is the interaction of the magma with water from the ice covered volcano. In order to address this in some detail, size-separated samples and a lava bomb were investigated.

3.3 Size-separated samples

There is evidence from scanning electron microscopy studies [13] that the fragmentation process in the beginning of the Eyjafjallajökull eruption was due to water-magma interaction. It would therefore be a logical interpretation that the more fragmented part of a sample (the smaller particles) had been more affected by interactions with water than the larger particles. In that case, one could expect to see a size dependence of the Fe³⁺/Fe_{Tot} ratio, where the largest particles would best preserve the original Fe³⁺/Fe_{Tot} ratio of the magma.

The sample from 17/4 2010 was separated using sieves. The sample <38 µm was further separated using sedimentation and Stokes law (see [14] for details on the method). Mössbauer spectra (not shown) were used to determine Fe³⁺/Fe_{Tot} ratios shown in Fig. 6.

Fig. 6 $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio determined from Mössbauer spectra of different size fractions. The *inset picture* shows the distribution of masses in the size bins from sieving

Fig. 7 Picture of a slice of the lava bomb. The original sample was close to spherical and ~ 20 cm in diameter. The surface appeared glassy, evidencing quenching. The *numbers* show subdivisions of samples

Only the sieved samples $<38 \mu\text{m}$ showed slightly elevated $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio of 29.4(20) %. However, this size fraction is less than 1 % of the weight of the whole sample, and has negligible effect on the $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio determined on the whole sample. From this, it can be concluded that the oxidation due to water/magma interactions has been short and that the $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratios determined in Fig. 5 are not affected by higher oxidation state of the finer particles.

3.4 Lava bomb samples

It can still be argued that the samples were affected as a whole by the interaction with water. To investigate that, a lava bomb was investigated. Lava bombs are clumps of lava, ejected during an eruption in a liquid or at least plastic state, which often takes on a near spherical shape in flight. Only the surface can be subjected to major interactions with water and/or atmosphere, and the interior can be regarded as representative of the melt.

A lava bomb was collected shortly after the eruption, and probably originates from late in the eruption (Á. Höskuldsson 2011, private communication). The original rock was ~ 20 cm in diameter (see Fig. 7) and had a glassy surface. Slower cooling of the interior was evidenced by more evolved gas bubbles in the centre of the bomb.

A slice of the bomb was subdivided into 4 samples depending on the distance from the surface, as indicated in Fig. 7.

Fig. 8 Room-temperature Mössbauer spectra of the lava bomb sample

Table 6 Area fractions obtained from the analysis of the spectra in Fig. 8

Sample	Fe(II)	Ol	Fe(III)	FeOx	Ilm
1	49.0(9)	7.0(5)	5.5(3)	35(1)	3.5(3)
2	45(2)	6.8(9)	7.7(6)	37(2)	2.0(5)
3	44(2)	8.1(6)	6.3(5)	38(2)	2.9(4)
4	45.1(9)	10.2(6)	6.1(4)	35(2)	3.1(3)

The Mössbauer spectra (Fig. 8) of different samples show only minor differences. The parameters are identical to those obtained for the time series (Tables 3 and 4) and spectral areas are given in Table 6.

The area fractions are approximately consistent with the area fractions of components observed in the last phase of the eruption. The most obvious trend in the dataset is the decrease of the Fe(II)/Ol ratio toward the centre of the bomb. The $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio of 27(2) % is also in reasonable agreement with the data in Fig. 5, and shows no increase toward the surface of the sample.

The important finding from the lava bomb is that interactions with water/atmosphere have not altered the ferric ratio significantly, allowing us to conclude that the whole-rock ferric ratios displayed in Fig. 5 are reliable.

3.5 Sample from the 1821–1823 eruption

According to [3, 4], the material that erupted in the Eyjafjallajökull eruption appears to be a mixture of two components, one akin the Fimmvörðuháls magma, the other that of the 1821–1823 Eyjafjallajökull eruption.

Fig. 9 Mössbauer spectrum of a tephra sample from the 1821–1823 Eyjafjallajökull eruption

The Mössbauer spectrum of a tephra sample from the 1821–1823 (Fig. 9) can be analysed in terms of the same components as before except that Ol is not present in this sample. The $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio is found to be 32.4(20) % using the same methods as above.

It should be emphasized that the $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio of samples from the Eyjafjallajökull (Fig. 5) are not a simple sum of the mixing proportions of Fimmvörðuháls-like material and 1821–1823 AD Eyjafjallajökull-like material. Both mixing end-members show evolution [3] that would have to be taken into account.

4 Discussion

The striking variation with time observed in the Mössbauer analysis of samples from the Eyjafjallajökull eruption is the abrupt change in the relative ratios of Fe(II) and Fe-Ox (Fig. 4). This seems not to be correlated with eruption phases in Table 1, and suggests a change in the eruption between 22/4 and 27/4 2010.

The relative increase in the Fe-Ox component, suggests more crystallization in the latter part of the eruption. Some of the Fe in the melt (part of Fe(II) and Fe(III)) combines to form inverse spinel-type iron oxides (Fe-Ox). This would suggest either lower quenching temperature and/or lower quenching rate of the melt in the latter part of the eruption. As water (glacier melt) is probably the factor that mainly controls the quenching of the melt, these changes suggest less access of water to the melt.

This is in accordance with the scanning electron microscopy observation of Dellino et al. [13] that reveals that ash produced during the first phase of the eruption shows key features of magma-water interaction while fragmentation was purely magmatic in the last phase of the eruption.

5 Conclusions

Mössbauer spectroscopy has been used to determine $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratios in samples from the Fimmvörðuháls/Eyjafjallajökull eruptions. For the Fimmvörðuháls sam-

ples, a whole-rock $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio of 11.9(5) % is determined whereas $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratio of the glass is found to be 15.6(6) %.

Samples from the Eyjafjallajökull eruption contain Fe^{2+} in olivine which is consistent with mingling and mixing of two components, a mafic component (with olivine) and silicic magma as found from studies of the chemical composition of samples [3, 4]. Samples from the Eyjafjallajökull eruption have $\text{Fe}^{3+}/\text{Fe}_{\text{Tot}}$ ratios ranging from 21.5(16) % to 33.8(26) %. At least part of this variation is explained by different mixing proportions of the end-members.

Mössbauer spectra of samples taken during the eruption suggest higher degree of crystallization in the latter part of the eruption, either due to lower quenching temperature of the magma and/or lower quenching rate, in both cases evidencing that access to water was reduced in the latter part of the eruption.

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