

Radon in magmatic gas at Surtsey and its possible
use for determining the content of water in the magma

by

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Abstract.

The average radon activity of magmatic gas sampled on 21 February 1965 at the lava crater of Surtsey was found to be $125 \pm 5\%$ pC/l of gas. The radium content of the lava was found to be less than $0.4 \cdot 10^{-12}$ g/g. The radioactivity of a 5 g sample prepared was less than the background activity in the ionization chambers used for the measurements. An attempt for determining the radium content by more sensitive methods is in preparation.

A method is suggested for estimating the water content of the magma by calculating the radon content of the magma before eruption from its radium content and measuring the radon/water ratio in the liberated magmatic gas.

The radium content of the Surtsey lava is not known at present, but calculations based on an average radium content in similar basalts and the observed radon/water ratio in the magmatic gas gave a water content of 0.9 wt. % in the erupted magma.

Introduction.

During the lava phase of the Surtsey eruption several attempts were made to sample magmatic gas from the active crater. Gas samples were taken for chemical analysis, the measurement of the D/H - ratio in H_2 - gas and water vapor and the measurement of radon.

The radon content of magmatic gas is of considerable interest, because the determination of radon in the gas and its parent

nuclide, radium, in the lava may supply information for estimating the content of water in the magma.

Sampling of gas.

The gas samples for radon measurement were taken simultaneously with samples for chemical analysis and deuterium measurements using a common sampling apparatus. The sampling procedure is described by Arnason (1,2) and Sigvaldason and Elísson (3,4).

Measurement of radon.

The activity of radon in the gas sample was measured by conventional methods (e.g. Evans (5)) using a 4-liter-ionization chamber and a Victoreen 475 A vibrating reed electrometer for reading the ionization current of radon in equilibrium with its decay products. The equipment was calibrated with the aid of a standard 0,099 μ C radium-solution.

The background current in the ionization chambers was equivalent to a radon activity of 2 pC. The radon activity of samples containing less than 2 pC could therefore only be determined with a limited accuracy.

Measurement of radium in lava.

In order to estimate the initial concentration of radon in the magma an attempt was made to determine the radium content of the issued lava. The sample used for the radium analysis was a lava-bomb, which was ejected from the crater, while the gas samples were taken on 21 February 1965.

The chemical preparation of the sample was performed by B. Arnason at the Physical Laboratory of the University of Iceland.

The preparation was similar to that described by Hudgens et al. (6). 5 g of lava were repeatedly dissolved in HF and fused with Na_2CO_3 , until the lava was completely dissolved. The solution was then sealed off in a glass gas-washing-bottle identical to the bottle containing the standard-radium-solution. Great care was taken that no materials, which absorb radon (e.g. grease, rubber) were in contact with the radon generated in the bottle. After 30 days the radon had attained equilibrium with its parent nuclide. The radon was then transferred into an evacuated ionization chamber by blowing bubbles of N_2 through the solution until the radon was completely washed out of the solution. The method is described in detail by Lucas, (7).

The same method was used for transferring radon from the standard radium solution into the chamber for calibration.

Results.

Radon. The results of the radon measurement are shown in Table I. The samples taken on 25 November 1964 and 18 January 1965 were heavily contaminated with atmospheric air. Chemical analysis (see Sigvaldason and Elísson (3,4)) indicated a 90 - 95% contamination. If sample 1 and 2 are corrected for contamination, they indicate a radon concentration of the order of 100 pC/l.

On 21 February far better samples were obtained. They contained less than 0.75 % N_2 , which indicates less than 1% atmospheric contamination, if any at all. Samples 5, 7 and 8 gave consistent values for the radon concentration with an average of $125 \pm 5\%$ pC/l.

Radium. The radium content of the solution containing 5 g of dissolved lava was found to be less than 2 pC. An accurate determination of the radium content was not possible, because the radon activity generated by the prepared sample was less than the background activity in the ionization chamber.

Discussion of the results.

Radon and radium. In uncontaminated samples of magmatic gas the average radon content was $125 \pm 5\%$ pC/l.

The radium content of lava issued at the same time was found to be less than $0,4 \cdot 10^{-12}$ g/g. This indicates a radon content in the magma less than 0,4 pC/g. Further attempts to determine the radium content of the lava are necessary. If the measurements are made with present apparatus, about 50 g of lava must be dissolved for a radium analysis.

The basalt of Surtsey is of the alkali - olivine type (Steinthorsson (8)). In alkali - olivine basalts in Japan Heier and Rogers (9) found an uranium content of $0,48 \pm 20\%$ ppm and $0,57 \pm 13\%$ ppm respectively. If a radioactive equilibrium between uranium and radium is assumed this would correspond to a radium content of $0,16 \cdot 10^{-12} \pm 20\%$ g/g and $0,19 \cdot 10^{-12} \pm 13\%$ g/g respectively.

In a review article on the radioactivity of basic rocks Heier and Carter (10) found an average value for the uranium content of 14 analyses of island basalts from Hawaii, Japan and the Mid Atlantic ridge to be 0,46 ppm, which corresponds to an equilibrium value of radium of $0,16 \cdot 10^{-12}$ g/g.

The upward limit for the radium content of the Surtsey lava given above, is consistent with this average value.

Liberation of gas from magma.

Before the results above are used for estimating the water content of the magma a short discussion of the mechanism of the liberation of gas from magma is necessary. As the magma ascends the solubility of gases decreases with falling pressure, and the magma may become oversaturated for some of its gas components. Bubbles of gas will then be created in the magma. In rapidly ascending magma great oversaturation may occur before any bubbles

are formed. After the formation of bubbles each component of the gas will strive to establish an equilibrium between the liquid and gas, determined by its solubility in the magma. Because of the high viscosity of the magma (Einarsson (11)) travel of bubbles relative to the magma is negligible and gas will therefore not escape before the bubbles reach the surface of the lava lake in the crater.

The composition of the gas accumulating under the roof of a lava lake will be the same as in the bubbles and will depend on the amount of each component initially dissolved in the magma and the solubility of these components at about 1100°C and 1 atm pressure.

Water.

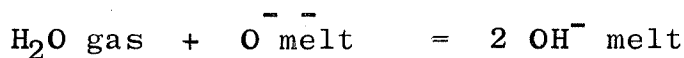
The solubility of water in magma has been thoroughly investigated. See e.g. Goranson, (12), Khitarov et al. (13) and Hamilton et al. (14).

Russel, (15) and Kurkijan et al. (16) have observed that a plot of the solubility (mol %) of water in molten alkali silicates versus the square root of pressure resulted in a straight line relationship in the pressure region below one atmosphere. Similar plots of the data obtained by Hamilton et al. (14) for basalts at 1100°C in the region of 1000 - 6000 bars pressure (1 kp/cm² = 0.98 bars) show that the solubility (mol %) is linearly related to the square root of pressure. The straight line passes through the origin and has a slope of 0.33 (mole %/(bars)^{1/2}). The solubility in moles % versus pressure in bars in basalt at 1100°C will be expressed by the formula

$$S = 0.33 \sqrt{P}$$

According to Hamilton et al. (14) this straight line relationship between solubility and the square root of pressure

might be interpreted to indicate that water entering the solution is fixed in the liquid structure as hydroxyl ions. Accordingly, each mole of water is visualized as entering the melt in the following manner:



It then follows, at constant temperature, that

$$\frac{[\text{f}_{\text{OH}^{\bar{\text{m}}\text{elt}}}]^2}{[\text{f}_{\text{H}_2\text{O gas}}] [\text{f}_{\text{O}^{\bar{\bar{m}}\text{elt}}}] = K$$

Therefore, as a first approximation, the concentration (the solubility) of $\text{OH}^{\bar{\text{m}}\text{elt}}$ in the melt at a given temperature varies as the square root of the water pressure.

The experimental data of Hamilton et al. (14) do not reach below 1000 bars pressure, but the facts that their empirical straight line goes through origo and that this relationship has also been observed for pressures below one atmosphere, indicate, that extrapolation of this line to lower pressures will give us a good estimate for the solubility of water in basaltic magma at 1100°C down to pressures of about one atmosphere.

In this way we obtain a solubility of 3.3 mole % (1.1 wt %) at 100 bars pressure, 1.05 mole % (0.35 wt %) at 10 bars and 0.33 mole % (0.1 wt %) at 1 bars pressure. 100 bars pressure corresponds approximately to 380 m depth in magma.

From these results we may deduce that magma, containing less than 0.1 wt % of water, would never saturate and no bubbles would be formed by the water vapor. If it contains 0.35 wt %, saturation will occur at about 40 m depth and about 70% of the water will be expelled at the surface. If the water content is 1 wt % the magma will saturate at 350 m depth and 90% of the water vapor will be liberated. Since the magma is rapidly ascending, bubble formation

may first occur after a some degree of oversaturation but once the bubbles are formed, liberation of the water will rapidly proceed.

Argon and radon.

It is well established by the workers of the K - Ar - method that fresh lava contains no detectable amount of argon. We may therefore conclude that argon has a negligible solubility in magma at 1 atm pressure and 1100°C. Radon is an inert gas like argon and the only difference in their solubility will be due to their different atomic mass. The solubility of radon under these circumstances is therefore likely to be of the same order as that of argon and we may expect practically all radon in the magma to be given off into bubbles and liberated.

Other volatile components.

The solubility of other volatile components in magma is less known. Their liberation will greatly depend on their degree of saturation in the magma. The solubility of those components, which approximately obey the Henry's law, will decrease approximately linearly with decreasing pressure. If some of these components are in saturated solution in the magma at pressures higher than 10 bars, they will be liberated into bubbles to more than 90% and thus their proportions in the sampled gas will be approximately the same as initially in the magma.

If the solubility of the components is proportional to the square root of the pressure as for water, saturation at 100 bars pressure is necessary for attaining 90% liberation.

Other components, which are so rare in the magma that they never reach saturated solution, in spite of the greatly reduced solubility at lower pressure, will not be liberated in the same proportions as other components.

Basaltic magma is believed to come from magma chambers below 30 km depth or pressures above 8000 bars. Gas components, which obey the square root law as water and do not reach saturated solution on their ascent with the magma, will have had a concentration in the magma chamber which was less than 1/90 of the saturation value at these depths. Components obeying the Henry's law will have had a concentration less than 1/8000 of the saturation value.

Evidence is lacking for estimating, if these low concentrations are realistic.

Water content of the magma.

After these considerations of the liberation of gas from magma, we may now proceed in our attempt to estimate the water content of the magma with the aid of radon. In the magmatic gas at Surtsey each liter of volatile gases was accompanied by about 5 g of water (Sigvaldason and Elísson (4)) and carried a radon activity of 125 pC. This radon activity is equal to that contained in 625 g of magma, if we take a radium content of $0.2 \cdot 10^{-12}$ g/g as a probable value for the radium content of the lava in Surtsey.

As already discussed above practically all radon will be liberated from the magma. Regarding the rapid ascent of magma and the great force of the escaping gases, we may assume that the gas is sampled a relatively short time after its liberation from the magma and therefore the decay of radon after liberation is negligible. (Radon decays with an half life of 3.8 days). Thus we come to the conclusion that 625 g of magma have liberated 125 pC of radon. As radon and water vapor are equally distributed in the gas, the 5 g of water which accompanied 125 pC of radon must also have been liberated from 625 g of magma. The magma has then given off 0.8 wt % of water. The solubility of water in the basaltic magma at 1100°C and 1 bar pressure was above estimated to be 0.1 wt %. If this amount is added to that liberated, we obtain a water content of about 0.9 wt % in the magma.

It must be emphasized that this result is still uncertain. The radium content used in the calculations above was an average value for alkali-olivine basalts and not the actual radium content of the Surtsey-lava, which is not known at present. The method is also based on estimates of the solubility of radon and water in magma at 1100°C and 1 bars pressure. A direct measurement of the solubilities would give a sounder basis.

Nevertheless, the radon method seems to be of value for estimating the water content of magma in future eruptions.

Comparison with earlier observations.

It would be interesting to compare this result to the findings of other workers, but actual estimates of the water content in erupted magma seem to be scarce.

MacDonald, (17) has estimated the content of gas in the magma erupted in the Mauna Loa summit eruption of 1940. The total volatile content was found to be approximately 1 wt % of the lava extruded during the first few hours of the eruption and 0.5 wt % of the lava being extruded a week later. The result was based on estimates of the amount of erupted magma and the amount of gas being liberated. According to MacDonald (17) it is probable that the average volatile content of the magma erupted in this eruption was less than 1 wt %.

Einarsson (18) investigated the physical and chemical properties of the lava erupted by the volcano Hekla in 1947-48. According to his observations the lava appeared tranquilly flowing in the crater in a spongy state having a specific weight of 0.6 - 0.8 and still retaining practically all its gas content. The water contained in the vesicles was less than 0.03 wt % of the lava and analyses of the lava which gave 0.35 wt % water on the average therefore showed practically the whole primary water content of the magma at great depths. Bombs and pumice thrown out at the beginning of the eruption had practically the same specific weight

and the same water content as the tranquilly flowing lava.

Einarsson (18) has investigated scoriae at many volcanic centers in Iceland and found them to be of the spongy type and similar in density to the Hekla material. In his opinion this indicates that as a rule the lava became oversaturated with gases (water) only just before it reached the surface, and that therefore the water content of magma is as a rule quite insignificant, 0.5 wt % or less.

Friedman (19) has measured the water content of a series of basaltic pumice and lava samples collected during the 1959-1960 summit and flank eruptions of Kilauea volcano, Hawaii. Samples of finely vesicular pumices of density ~ 0.3 g/cc from the summit eruption of Kilauea Iki were found to contain 0.064 to 0.099% H₂O by weight, but similar samples from the Kapoho flank eruption had water contents of 0.086 to 0.104 wt % H₂O.

Friedman (19) believes that the water extracted from the finely vesicular glassy pumices represents the water present in the magma just prior to eruption. Errors due to uptake of additional water from the surroundings were minimized by (1) a skin of non-vesicular lava on the surface of the pumice lumps, (2) rapid collection of the material shortly after eruption, and (3) the selection of large pieces of pumice for analysis.

Samples of coarsely vesicular pumice and lavas and also crystallized lavas were also analyzed; however, the results obtained (on these samples) varied widely from replicate to replicate.

According to Friedman (19) it is evident that during the eruption of these materials, water was lost from the large open vesicles. In addition varying amounts of water were lost during the partial or total crystallization of the lava during cooling.

It is interesting to note that the water content of the Hawaiian pumices found by Friedman (19) is very similar to the

saturation value of 0.1 wt %, which we found by extrapolating the data of Hamilton et al. (14) down to 1 bar pressure. On the other hand Einarsson (18) found a water content of 0.35 wt % in the Hekla lava. Similar results have been obtained by Tolstikhin (20) for the water content of the Quarternary effusives of Kamchatka and the Kurile Islands, estimated from 265 rock analyses. The average water content in basic and intermediate rocks was found to be 0.40 wt %, ranging from 0.1 wt % to 3 wt %.

TABLE IRadon activity in magmatic gas at Surtsey

Sample of gas	Volume of sample ml	Activity of radon in liter of sample pC/l	Atmospheric contamination of sample	Activity of radon in liter of magmatic gas (corrected for atm. cont.) pC/l
25. Nov. 1964 Sample 1	250	12 \pm 25%	90%	120
18. Jan. 1965 Sample 2	250	6 \pm 40%	90-95%	40-160
Sample 3	250	nil.		
21. Feb. 1965 Sample 5	189	136 \pm 5%	nil.	136 \pm 5%
Sample 7	208	119 \pm 5%	nil.	119 \pm 5%
Sample 8	206	120 \pm 5%	nil.	120 \pm 5%

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