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RADON AND WATER IN VOLCANIC GAS AT SURTSEY

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### Abstract

The radon concentration in volcanic gas at the oceanic volcano Surtsey, off the southern coast of Iceland, was found to range from 120 to 170 pC/l of noncondensing gases. The release of gas from ascending magma is briefly discussed and the suggestion made that radon could be used as a tracer for calculating the amount of water released from the erupted magma. According to this method the alkali-olivine basalt magma of Surtsey contained about 0.75 wt% of dissolved water.

Radon might possibly also be used for estimating the amount of other gas components, if data on the solubility of the volatile components in magma were available.

## Introduction

During the eruption of Surtsey, 1963 to present, many attempts have been made to sample volcanic gases released from the basaltic melt. Samples for chemical analysis, the measurement of D/H-ratio in hydrogen gas and water vapor, and the measurement of radon were taken simultaneously and with the same sampling apparatus. The results of the chemical analysis and the deuterium measurements are reported in the accompanying papers by Sigvaldason and Elisson (1967) and Arnason and Sigurgeirsson (1967). The present paper reports the results of radon measurements and discusses the possible use of radon as a natural tracer for estimating the water content of the erupted magma.

## Sampling of the volcanic gas

A detailed description of sampling locations and the apparatus and procedures for collecting the gas is given in the accompanying paper by Sigvaldason and Elisson (1967). The gas samples used for radon measurement are listed in Table 1. The samples are numbered in accordance to the numbers of simultaneous samples taken for chemical analysis and deuterium measurements.

## Measurement of radon

The activity of radon in the gas samples is measured by conventional ionization chamber techniques (e.g. Evans 1935). The arrangement of the apparatus is schematically shown in Fig. 1. The walls of the 4-liter-ionization chamber are held on + 900 volts by a stable dry battery supply. The central electrode is mounted in two teflon insulators, which are separated by a grounded guard ring to prevent stray currents in reaching the electrode. The ionization current is measured by a vibrating reed electrometer (Victoreen model 475 A). The background current in the chamber is equivalent to a radon activity of 2 pC.

The amount of gas contained in the gas sampling tube is determined by adjusting the gas pressure to one atmosphere with the aid of a niveau-flask containing saline water and measuring the volume of gas at this pressure and room temperature.

Two  $\text{Mg}(\text{ClO}_4)_2$  traps and one Ascarite-trap are used for drying the gas and removing most of the  $\text{SO}_2$  and  $\text{CO}_2$  before the sample is introduced into the chamber.

After reading the background current the ionization chamber and the purifying traps are evacuated down to about 60 mm Hg pressure and the gas sample sucked into the chamber through the drying and absorbing agents. The chamber is then slowly filled up to atmospheric pressure by blowing bubbles of  $\text{N}_2$  through the sampling tube for about 15 minutes. In this way the bubbles flush out more than 97% of the radon dissolved in the water in the tube. (See Lucas 1964).

The ionization current is read about three hours after the sample is introduced into the chamber, i.e. when radon has attained a radioactive equilibrium with its decay products.

The equipment was calibrated with the aid of a standard 0.099  $\mu\text{C}$  radium solution using the same method as above for transferring radon from the standard solution into the chamber.

## Results

### Radon

The results of the radon measurements are shown in Table 1. The radon activity in pC/l of noncondensing gases is shown in column 7. The amount of condensed water accompanying each liter of noncondensing gases is listed in column 6. According to deuterium measurements (Arnason and Sigurgeirsson 1967) this water has been released from the magma and is not evaporated sea water as one might perhaps suggest in an oceanic volcano. Column 5 shows volume percent of nitrogen and inert gases which serves as an indicator of atmospheric contamination

of the samples. According to Sigvaldason and Elísson (1967) the samples from February 21, 1965 are practically free from atmospheric contamination. Nitrogen and inert gases constitute only 0.5 vol % of the noncondensing gases in these samples. In the last column of Table 1 the radon activity per liter of noncondensing gases has been corrected for atmospheric contamination assuming that all nitrogen and inert gases in excess of 0.5 vol % is of atmospheric origin.

### Radium

In order to calculate the initial concentration of radon in the magma an attempt was made to determine the radium concentration in a lava bomb which was ejected from the crater on February 21, 1965, when the best gas samples were obtained. About 5 g of lava were repeatedly dissolved in HF and fused with  $\text{Na}_2\text{CO}_3$  until the lava was completely dissolved.

The radon generated in the solution was measured with the aid of the ionization chamber. The sensitivity of the method was, however, not sufficient and it could only be ascertained that the radium concentration was less than  $0.4 \cdot 10^{-12}$  g Ra/g of lava.

This sample and another, taken from lava flowing on Sept. 8, 1966 were then sent to Dr. K.S. Heier at the Australian National University, who kindly assumed the task of determining the radium concentration in these samples.

Dr. Paul W. Gast, Lamont Geological Observatory visited Surtsey in August 1966 and collected several samples for radium measurements. He also obtained a piece of the lava bomb mentioned above and has kindly allowed us to cite his unpublished results. The results of the radium measurements are summarized in Table 2.

### Discussion of the results

Gas samples taken close to the crater indicate a radon concentration of 120 to 170 pC/l, whereas samples 26 and 28 taken from a lava flow, which had been exposed to the surface for a distance of about 100 meters, contain only 50 to 70 pC/l. These samples were also anomalous in chemical composition (Sigvaldason and Elísson 1967) and deuterium ratio (Arnason and Sigurgeirsson 1967) and do probably represent an advanced state in the degassing of the lava.

The radium measurements indicate a radium activity of 0.1 to 0.3 pC/g of lava. As radon attains radioactive equilibrium with its parent nuclide radium within 30 days we may conclude that the initial radon concentration in the magma was of the order of 0.1 to 0.3 pC/g of magma. The concentration corresponds to several thousand atoms of radon per gram. In this extreme dilution the radon gas is a tracer far from being in saturated solution. Release of radon from the magma will thus not occur unless the magma becomes saturated with some other gas component, which is then partly exsolved and creates gas bubbles in the magma. Radon will be released into the gas bubbles. The amount released will depend on the volume percentage of bubbles and the solubility of radon in the liquid magma.

It would be of considerable interest, if we could estimate to what extent radon is released with a certain amount of volcanic gas from a unit mass of magma. Knowing the amount of radon initially dissolved and e.g. the radon/water ratio in the volcanic gas, we might then be able to calculate the amount of water released from the magma.

In the following an attempt will be made to estimate the release factor of radon. Since water is the main component of volcanic gas, we will limit our discussion to the release of water and radon from basaltic magma, which saturates with water on the ascent to the crater.

### The release of water in ascending magma

The release of water in ascending magma depends primarily on the solubility of water in the magma.

According to experimental data obtained by Hamilton et. al. (1964), Russel (1957) and Kurkjian et. al. (1958) the solubility of water in basalt melts is proportional to the square root of pressure. Extrapolation of the data of Hamilton et. al. down to lower pressure indicates a solubility of 0.11 wt % in a basalt melt (50.71 wt % SiO<sub>2</sub>, 4.68 wt % MgO) at 1100°C and 1 atm pressure (Björnsson 1966). This estimate is supported by the evidence on water retained in Hawaiian lavas. According to Mac Donald (1963) the average combined water content (H<sub>2</sub>O+) in 51 recent analyses of tholeiitic basalts of Kilauea and Mauna Loa of historic age, by several different analysts, is 0.14 wt %. Older analyses give somewhat higher values, but the more recent analyses, by improved techniques, are considered to be more nearly correct.

We may thus conclude that magma containing water dissolved in excess of about 0.14 wt % will saturate during the ascent and the excess water will create vapor bubbles in the magma. At 1100°C and pressures below 100 atmospheres the specific volume of water vapor deviates less than 1 % from that of an ideal gas (Kennedy et. al. 1966). The total volume of bubbles released from one gram of magma containing initially 0.1 wt % of excess water will be 6.25 cm<sup>3</sup> at 1100°C and 1 atm pressure. The volume of bubbles will be about 95% of the total liquid-gas volume. This means that magma containing only 0.1 wt % of excess water will be changed into a froth on the last tens of meters before it reaches the surface in the crater. This extensive frothing of the magma must greatly facilitate the release of other gas components into the vapor phase, as shown in the following discussion for radon.



The release of radon from magma

If  $R_r$  denotes the amount of radon released into vapor bubbles from a unit mass of magma and  $R_m$  the amount of radon initially dissolved in that unit mass, the release factor of radon is defined by the equation

$$k = R_r / R_m \quad (1)$$

The solubility of radon in the magma is defined as

$$\alpha = C_m / C_r \quad (2)$$

where  $C_m$  and  $C_r$  are the volume concentrations of radon in the liquid magma and the vapor bubbles respectively.

If  $V$  is the volume of bubbles of water vapor released from a unit mass of magma and  $v$  the specific volume of liquid magma the release factor  $k$  may under equilibrium conditions also be expressed as

$$k = \frac{V}{V + \alpha v} = \frac{1}{1 + \alpha v / V} \quad (3)$$

If  $W_r$  denotes the amount of water in grams released from one gram of magma, we may substitute  $W_r$  for  $V$  using the relation  $V = 6.25 \cdot 10^3 W_r$  which is valid at  $1100^\circ\text{C}$  and 1 atm pressure. Assuming further a density of  $2.9 \text{ g/cm}^3$  of the liquid magma, we obtain the expression

$$k = \frac{1}{1 + 5.5 \cdot 10^{-5} \alpha / W_r} \quad (4)$$

Thus the release factor of radon depends on the solubility of radon and the amount of water released. If  $W_r \gg 5.5 \cdot 10^{-5} \alpha$  practically all radon will be released.

The amount of water dissolved in the magma

We may now attempt to calculate the amount of water released from the magma using the data on the initial radon concentration in the magma,  $R_m$ ; the observed radon/water ratio in the volcanic gas,

$$\beta = (R_r / W_r) \quad (5)$$

and an estimate of the solubility of radon in the magma,  $\alpha$ . Combining equations (1), (4) and (5) and solving for  $W_r$  we obtain

$$W_r = \frac{R_m}{\beta} - 5.5 \cdot 10^{-5} \cdot \alpha \quad (9/9) \quad (6)$$

or if we express the amount of water released in weight percents

$$X_r = \frac{100 \cdot R_m}{\beta} - 5.5 \cdot 10^{-3} \cdot \alpha \quad (\text{wt}\%) \quad (7)$$

Only the last term of this equation depends on the solubility of radon. The solubility is not known but a reasonable assumption seems to be that the concentration of radon in the liquid is not greater than in the vapor i.e.  $\alpha \leq 1$ . But even for  $\alpha = 10$  this term will only be about 0.05 wt % which is insignificant for the present discussion.

In Table 3 the radon/water ratio,  $\beta$ , and the initial radon concentration in the magma  $R_m$  are tabulated from the data in Table 1 and 2. We may remark that the radon/water ratio is in general not affected by atmospheric contamination. Water added due to the burning of hydrogen is insignificant. As the radium concentration in the lava flowing on March 31, 1967 has not been determined, the concentration in the flow on September 8, 1966 is used for calculating the initial radon concentration on March 31, 1967. Solving equation (7) for this data and neglecting

the last term, we obtain 0.64 wt % of water released on February 21, 1965 and 0.56 wt % on March 31, 1967. If we add the amount of water retained in the magma, which in Hawaiian lavas was found to be 0.14 wt % on the average, we obtain a total of 0.78 wt % and 0.70 wt % respectively.

Using radon as a tracer we have thus come to the conclusion that the alkali-olivine basalt magma of Surtsey (46.6 wt %  $\text{SiO}_2$  and 9.3 wt % MgO, S. Steinhorsson, personal comm.) contained about 0.75 wt % of dissolved water.

It is of interest to compare this result to the findings of Moore (1965). In deep-sea tholeiitic pillow lavas (48.9 wt %  $\text{SiO}_2$  and 13.2 wt % MgO) dredged from the submarine part of the east rift zone of Kilauea, Hawaii, he found an average water content of  $0.45 \pm 0.15$  wt %  $\text{H}_2\text{O}+$ . The water content was roughly constant between 500 and 5000 meters depth and was regarded to represent the amount of water dissolved in the erupting magma.

To conclude, the measurement of radon in volcanic gas seems to offer an useful method for estimating the amount of water dissolved in magma. More data on the solubility of water and radon in magma at atmospheric pressure would give the method a sounder basis. It might also become possible to estimate the amount of other gas components but at present data on the solubility of these components is far too incomplete to justify a sincere attempt in that direction.

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Legends for figures.

Fig. 1. Schematic drawing of apparatus for measurement of radon in gas samples.

- 1 Ionization chamber, 2 Vacuum gauge,
- 3 Electrometer, 4 Drying agent ( $\text{Mg}(\text{ClO}_4)_2$ ),
- 5 Ascarite, 6 Drying agent ( $\text{Mg}(\text{ClO}_4)_2$ ),
- 7 Gas sampling tube

Table 1

Results of radon measurements in volcanic gas at Surtsey

Date of sampling	Distance from crater	Sample No.	Volume of non-condensing gases	Nitrogen + inert gases	Water	Radon activity	Radon activity corr. f. atm. contamination
	m		ml	Vol %	g/l	pC/l	pC/l
Nov. 25, 1964	30	14	250	90	2.8	12 ± 25%	120 ± 50%
Feb. 21, 1965	0	20	189	0.5	4.7	136 ± 5%	136 ± 5%
	0	23	206	0.5	4.7	120 ± 5%	120 ± 5%
	0	24	208	0.5	4.7	119 ± 5%	119 ± 5%
Sept. 2, 1966	150	26	166	58	2.7	30 ± 20%	71 ± 20%
	150	28	193	35	2.7	34 ± 20%	52 ± 20%
March 31, 1967	20	29	383	10.0	6.2	155 ± 5%	171 ± 5%
	20	30	472	41.4	6.2	90.4 ± 5%	159 ± 20%



Table 2

Radium in Surtsey lava

Date of flow	Radium $10^{-12}$ g/g	Author	Method
(Su 1, not dated)	0.26	} P.W. Gast 1967	
(Su 2, not dated)	0.27		
Su 3, Feb. 21, 1965	0.17		
Feb. 21, 1965	<0.4	S. Björnsson 1966	Ionization chamber. Radon generated in solution.
Feb. 21, 1965	$0.17 \pm 0.01$	} K.S. Heier 1967	Gamma-ray spectrometry 1.76 MeV Bismuth- 214 peak
Sept. 8, 1966	$0.11 \pm 0.01$		

Table 3.

Water content of erupted magma at Surtsey

Date of sampling	Distance from crater	Sample No.	Radon/water $\beta$	Radon $R_m$	Released water, $x_r$	Total water, $x$
	m		pC/g	pC/g	wt %	wt %
Feb. 21, 1965	0	20	29.0	0.17	0.64	0.78
	0	23	25.3			
	0	24	25.3			
March 31, 1967	20	29	25.0	(0.11)	0.56	0.70
	20	30	14.6			

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Schematic drawing of apparatus  
for measurement of radon in  
gas samples

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