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"Estimation of the Distribution of Tritiated Water Between the Vapor Phase and Molten Rock in the Cavity Created by a Nuclear Explosion - With Special Reference to Recent French Data"

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29

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Estimation of the Distribution of Tritiated Water Between the Vapor Phase and Molten Rock in the Cavity Created by a Nuclear Explosion - With Special Reference to Recent French Data

R. G. Bedford

I. Consideration of the Distribution of Water Between the Vapor Phase and a Solution in Liquid Silicates

Previous investigations on the high temperature, high pressure solubility of water in molten silicates⁽¹⁾ and review of literature⁽²⁾ indicate that the solubility of water in molten silicates is essentially independent of temperature between 600° and 1500°C, and that the solubility is about the same for melts of most naturally occurring silicate materials. The solubility depends on the square root of the water vapor pressure. The equilibrium constant can be expressed in the form $K = \frac{C}{P^{1/2}}$ where C is the concentration of H₂O in the silicate and P is the vapor pressure of H₂O(g). If C is expressed in the units grams H₂O/gram silicate and P in atm, $K \approx 1.5 \times 10^{-3}$. Experimentally measured values of K have been found to be in the range 1.3×10^{-3} to 1.7×10^{-3} .^(1,2)

II. A Model for the Distribution of Tritiated Water in a Cavity Created by a Nuclear Detonation

We hypothesize the following process for introduction of tritium produced in an underground nuclear event into the molten rock. At high temperatures and correspondingly high water vapor pressures, as the vaporized rock condenses and additional rock is melted, the tritium produced in the event is incorporated in water vapor derived from molten rock. This treatment considers only tritium that is incorporated in water. In cases where the rock contains carbon, some tritium will also be found in hydrogen and in methane, as discussed by Taylor et al.⁽³⁾ The tritium-bearing water dissolves in the molten silicate at concentrations corresponding to relatively high partial pressures of water existing in the cavity when the temperature has just fallen below the boiling point of the rock. As the system cools and the water vapor pressure decreases, tritiated water escapes from the molten material until the rock freezes, or until the viscosity of the glass becomes so high that diffusion of water out becomes insignificant.

*Work performed under the auspices of the U.S. Atomic Energy Commission.

The following assumptions are made:

1. Only water originally contained in the rock which is melted, is involved. No water is lost by leakage or by chemical reaction, and no additional water is added from heated but unmelted material.
2. The volume in which the gas is contained equals the cavity volume.
3. Equilibrium is established and frozen at the melting point of the rock. Gas and rock temperatures are equal at that point and no significant transfer of water occurs between rock and vapor phase as further cooling takes place.
4. Water vapor obeys the ideal gas law at the equilibrium temperature and pressure.
5. The mass of rock melted and the volume of cavity produced scale linearly with device yield.

Using the assumptions listed above, we derived an equation based on the ideal gas law and the solubility expression which gives the fraction of water dissolved in the melt.

$$F = A \left[\left(\frac{2}{A} + 1 \right)^{1/2} - 1 \right]$$

where F = the fraction of tritiated water dissolved in the melt

and $A = \frac{2.27 K^2 MT}{VC_0} = 5.11 \times 10^{-6} \frac{MT}{VC_0}$.

K = the equilibrium constant for solubility of water in molten silicate (1.5×10^{-3});

M = the mass of rock melted per kt yield, g/kt;

T = the freezing temperature, °K;

V = the cavity volume per kt yield, cm³/kt;

C_0 = the average concentration of water in the original rock, gH₂O/g rock.

F vs. A is plotted in Fig. 1.

The only parameters of A which change significantly from one experiment to another in silicate media are V and C_0 . For contained shots, V varies from about 1.5×10^9 cm³/kt to about 1.5×10^{10} cm³/kt and C_0 from 0.003 to 0.3. For shallow shots of medium to high yield, particularly in wet media, V can

become much larger, or the shot can vent to atmospheric pressure leading to values of $F \ll 1\%$. But for contained shots in silicate media, F is expected to be in the range 0.1 to 0.75. Therefore, for experiments in which water is the principal hydrogen-containing substance, 10% to 75% of the tritium should be found in the glass.

Significant discrepancies between the model and physical reality are likely to be included in the assumptions.

1. At present not enough is known about cavity formation phenomenology to permit selection of the best value to use for the amount of water involved in the equilibrium. If significant amounts of hydrogen-containing species other than water are involved, the equilibria can be established by the methods discussed in Ref. (3). The tritium contained in these substances can thus be accounted for. It should also be noted that tritiated water in the gas phase at the time the equilibrium is frozen will subsequently be diluted by water extracted from unmelted rock so that the post-shot concentration of tritium in water is only indirectly related to the tritium content of the glass.
2. The volume occupied by the molten material has been neglected. Conceivably, in very deep shots the volume of rock melted could be a significant fraction of the cavity volume. In these cases an appropriate correction can be applied. A far more serious problem is the uncertainty of the volume produced by a given yield under a given set of conditions.
3. The third assumption is very idealized. In the actual nuclear event, as the melt cools and solidifies, the vapor phase is probably cooler than the freezing rock due to contact with cooler material at the cavity walls and debris from chimney formation. Since the rock cools fairly slowly during this period, water is probably lost from the melt after the gas has cooled substantially below the freezing point of the rock. This results in a smaller fraction of the tritium being contained in glass than that calculated. Also, neither the gas phase nor the liquid can be expected to be homogeneous in composition or in temperature throughout this period.
4. The fourth assumption probably does not contain any problems. Since the solubility of water depends on the square root of pressure, the uncertainties in the proper value for the effective equilibrium temperature are not serious.

5. The problems in the fifth assumption have been mentioned above. Accurate measurements of the mass of rock melted or of cavity volumes are not generally available. Therefore, appropriate values for V and M to be used in specific cases remain uncertain.

III. Application of the Model to Specific Examples

We have used the model to estimate the fraction of tritium (tritium contained in water at the time the cavity temperature is about 1000°C) expected in the solidified melt for a few specific examples, and plotted the results on Fig. 1. Since conditions are not known precisely, accurate values for tritiated water in glass cannot be calculated. Because of the approximations involved in the model, we expect the results to represent an upper limit to the tritium retained.

French papers presented at the 1970 American Nuclear Society Conference, Las Vegas, Nevada and other publications⁽⁴⁾ list the following average parameters for experiments in a granitic formation in the Sahara Desert:

$$M = 1300. \times 10^6 \text{ g/kt}$$

$$T = 1300. \text{ }^\circ\text{K (melting point of rock)}$$

$$V = 1.63 \times 10^9 \text{ cm}^3/\text{kt (cavity radius} = 7.3 \text{ W}^{1/3} \text{ meters)}$$

$$C_0 = 6. \times 10^{-3} \text{ g-H}_2\text{O/g-rock}$$

The French also reported orally⁽⁵⁾ that approximately 50% of the tritium produced in the events was found in the solidified melts.

The fraction of water dissolved in the melt calculated using the information given above is $F = 0.71$. The concentration of water in the solidified melt would be $4.3 \times 10^{-3} \text{ g-H}_2\text{O/g-glass}$ and the partial pressure of water in the cavity at the time equilibrium is frozen (1300°K) would be 8.2 atm.

Since the parameters are somewhat uncertain, we have calculated the distribution for a few other cases to show the effects of changing these values. If the water concentration of the original rock were $C_0 = 3. \times 10^{-3} \text{ g-H}_2\text{O/g-rock}$, the fraction in the solidified melt would be $F = 0.81$. If the cavity radius were $10 \text{ W}^{1/3} \text{ meters}$, the distribution would be $F = 0.55$ for $C_0 = 6. \times 10^{-3}$ and $F = 0.67$ for $C_0 = 3. \times 10^{-3}$. For the conditions listed by the French scientists, we estimate 75% as an upper limit to the tritium retained, which compares

favorably with their observation. Apparently, the crude model given above allows the distribution of tritium contained in H_2O to be estimated to better than a factor of 2.

The high fraction of tritium retention in the rock in the French experiments resulted from the small cavity radius per kt, the large mass of rock melted per kt, the low water concentration of the rock, and the small amounts of hydrogen and hydrocarbons produced.

It must be pointed out that most rocks (including those of most past and proposed Plowshare experiments) contain more than 4% water. Two examples are given of such cases. Typical parameters for events in turf and alluvium are:

$$M = 600 \times 10^6 \text{ g/kt}$$

$$T = 1300^\circ\text{K}$$

$$V = 16. \times 10^9 \text{ cm}^3/\text{kt} \text{ (cavity radius } \approx 15.5 \text{ W}^{1/3} \text{ m)}$$

$$C_0 = 4. \times 10^{-2} \text{ g-}H_2O/\text{g-rock}$$

The fraction of water dissolved in solidified melt in such an example would be $F = 0.11$ (the water pressure at equilibrium (1300°K) would be ~ 10.5 atm).

For events at greater depths or in denser media, such as Gasbuggy:

$$M = 600. \times 10^6 \text{ g/kt}$$

$$T = 1300.^\circ\text{K}$$

$$V = 4. \times 10^9 \text{ cm}^3/\text{kt} \text{ (cavity radius } \approx 10 \text{ W}^{1/3} \text{ m)}$$

$$C_0 = 4. \times 10^{-2} \text{ g-}H_2O/\text{g-rock}$$

For this case the fraction of water dissolved in solidified melt would be $F = 0.20$ and the water vapor pressure at the time the melt freezes would be about 28.4 atm. If more than 600×10^6 g/kt of rock were melted, the fraction water in solidified melt would be greater. This represents only the distribution of tritiated water. If a substantial amount of the tritium is in the form of hydrogen and hydrocarbons at the time melt freezes, the amount of tritium in the melt would be less.

IV. Comments

It is suggested that post-shot analyses be made for the water and tritium contents of solidified melts in Gasbuggy and other nuclear events so that correlations can be made involving cavity volume, mass of rock melted, post-shot water content of the glass, and tritium distribution. Such correlations would allow us to calculate the amount of steam in which the tritium was distributed at the time the melt froze. The results would be useful both for developing a model for cavity phenomenology and for allowing us to make predictions of tritium distribution in proposed nuclear explosions with greater confidence.

Fig. 1. Tritium Dissolved in Melt in Nuclear Explosions

- Calculated for French experiments (Ref. 4).
- ⊙ Estimated for French experiments from post-shot analyses.
- Calculated for typical Nevada experiments in tuff and alluvium.
- △ Calculated for conditions similar to those for Gasbuggy.

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