PROCEEDINGS, Fourteenth Workshop on Geothermal Reservoir Engineering
Stanford University, Stanford, California, January 24-26, 1989
SGP-TR-122

ANOMALOUS BEHAVIOUR OF HYDROGEN IN STEAM FROM VAPOUR-DOMINATED GEOTHERMAL SYSTEMS

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ABSTRACT

The ratios of gases in dry steam produced from the vapour-dominated reservoir of the Unit 15 steam field at The Geysers in Northern California, USA, can be explained by a model incorporating mixing between original reservoir vapour and vaporised reservoir liquid. However, hydrogen analyses are anomalous in that they are apparently enriched in the produced steam compared with levels predicted by this model. This enrichment appears to be caused primarily by fluid-rock interactions during boiling in the reservoir, although well casing corrosion reactions during production are also contributary. Hydrogen enrichment also appears to be present in the northwestern area of The Geysers and at the vapour-dominated geothermal systems of Larderello, Italy, suggesting its occurrence may be widespread. The calculation and interpretation of steam fractions and water-rock interaction models could be significantly affected by this hydrogen enrichment phenomenon.

INTRODUCTION

The Geysers (northern California, USA) and Larderello (Tuscany, Italy) are the largest and most heavily exploited vapour-dominated geothermal systems in the world. They have been extensively studied from both reservoir engineering and geological viewpoints which have provided pre- and post-production conceptual models (eg White et al, 1971; Pruess, 1985; Walters et al, 1988). In recent years the importance of geochemical and isotopic studies has been recognised through the evaluation of the effects of recharge and reinjection, and the interest in reservoir liquid-vapour ratios (eg Panichi et al, 1974; D'Amore and Truesdell, 1979; D'Amore et al, 1982, 1983, 1987; Truesdell et al, 1987; Haizlip and Truesdell, 1988).

Due to the larger volume of published data, most geochemical research has concentrated on the Larderello field. No previous studies of The Geysers have involved a detailed examination of steam gas compositions although a general model for the field has been proposed (Truesdell et al, 1987). In contrast, this present study presents some results from a major investigation of steam gas data collected by GEO Operator Corporation (GEOOC) from Unit 15 at The Geysers (Figure 1). The data were evaluated by GeoScience Limited and GEOOC, both wholly owned subsidaries of Geothermal Resources International Inc. This investigation has produced a conceptual model of the area involving the production of steam from a mixture of both vaporised liquid and vapour* sources (McCartney and Haizlip, 1989). Some anomalies to the model exist although the most evident involves hydrogen which appears to be enriched in steam produced from low gas steam

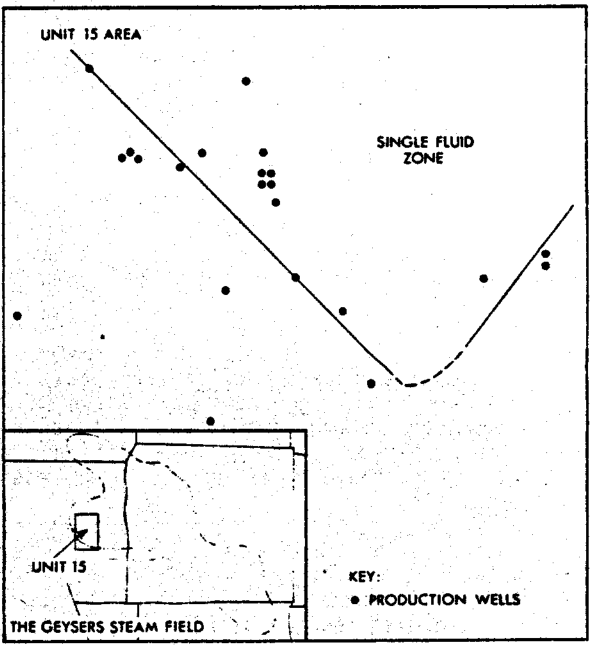

FIGURE 1 LOCATION OF THE UNIT 15 AREA AT THE GEYSERS AND THE SUGGESTED ZONE OF PRODUCTION FROM THE SINGLE FLUID SOURCE.

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* 'Vapour' in this text refers to vapour which has equilibrated with reservoir liquid and rock at the reservoir temperature. The term 'steam' is used generically to refer to a vapour phase (eg it can be used where the fluid is a mixture of vaporised liquid and 'vapour').

wells. Unpublished data from the northwestern area of The Geysers, and of published data from Larderello (D'Amore et al, 1983, 1987), indicate that this might be a widespread phenomenon.

The purpose of this paper is to present the evidence for hydrogen enrichment in steam produced from low gas wells at The Geysers and Larderello, examine the possible causes of this anomalous behaviour, and offer an explanation for the observations. As hydrogen is commonly used in the calculation of reservoir temperatures and steam fractions, the implications of hydrogen enrichment to such calculations are also discussed.

EVIDENCE FOR THE ANOMALOUS BEHAVIOUR OF HYDROGEN AT UNIT 15

Truesdell et al (1987) noted that the total gas concentrations in steam from The Geysers range from approximately 150 to 65 000 ppmw. The general order of dominancy in the gas phase is:

$$CO_2 > H_2 > CH_4 > NH_3, H_2S > N_2$$

CO_2 is typically 50-70 mole percent of the dry gas whilst the proportion of hydrogen usually ranges from 10 to 20 mole percent. At Unit 15, a similar order of gas dominancy is present with steam total gas concentrations ranging from approximately 1000 to 15 000 ppmw. All gas concentrations increase with increases in CO_2 concentration. The average proportions of CO_2 and H_2 are approximately 60 and 16 mole percent in the dry gas, respectively.

McCartney and Haizlip (1989) selected 290 steam samples, which had been collected from Unit 15 wells between 1979 and 1988, for examination. These were collected under conditions of full, stable production flow and are believed to be free of significant air contamination as all samples contained less than 5 mole percent N_2 in the dry gas. Their interpretation suggests that a number of wells from a geographically defined area at Unit 15 (Figure 1) derive their steam from a single, two-phase, fluid type* under similar reservoir conditions.

The distribution of a gaseous constituent between a liquid and a vapour phase can be described through the use of a laboratory derived distribution coefficient (Bi, where i is a gaseous constituent) which is the ratio of the gas molality in the vapour phase to the gas molality in the liquid phase. In terms of the constituents normally analysed in produced steam, the order of affinity for the vapour phase based on distribution coefficients at temperatures observed at The Geysers and Larderello (~240-260°C) is (see D'Amore and Truesdell, 1986, for a summary; Haizlip and Truesdell, 1988):

$$N_2 > H_2 > CH_4 > CO_2 > H_2S > HCl > NH_3 >> H_3BO_3$$

where, N_2 is the least 'soluble' gas and only H_3BO_3 has a distribution coefficient less than unity (ie partitions preferentially into the liquid phase).

Given that (a) the single fluid type is produced from the reservoir as steam, (b) no mass loss occurs through condensation between the production source and the wellhead, (c) no gas-gas or water-rock interaction occurs during or after boiling of the liquid phase, and (d) the gases obey their distribution coefficients, the change in gas composition with change in fraction of produced steam derived from vaporised liquid is qualitatively, if not quantitatively, predictable. As the proportion of vaporised liquid increases, gas concentrations decrease, and the fraction of the more liquid soluble gases increases in the produced steam phase. As an example, because BCH_4 is greater than BH_2S, the H_2S/CH_4 mole ratio of steam is higher at lower gas concentrations where the fraction of vaporised liquid in the produced steam is dominant over the fraction of original vapour. Single fluid type wells at Unit 15 show such a trend (Figure 2) which can be explained by assuming mixing between vaporised liquid and original vapour (Equation 1; after D'Amore et al, 1982) given the above conditions.

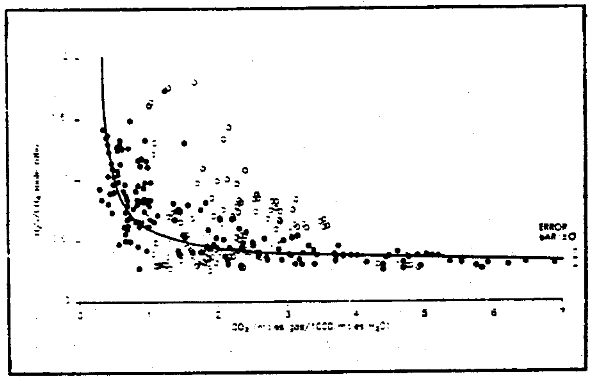

FIGURE 2 VARIATION OF H_2S/CH_4 MOLE RATIO WITH CO_2 CONCENTRATION FOR UNIT 15 STEAM SAMPLES. SOLID LINE REPRESENTS A BOILING/MIXING MODEL FIT THROUGH SINGLE FLUID SOURCE SAMPLES (SOLID CIRCLES). OPEN CIRCLES REPRESENT SAMPLES FROM ANOMALOUS WELLS.

$$C_s = C_v.y + C_l.(1-y) \qquad (1)$$

where, C_s = Concentration of gas in produced steam (moles/1000 moles H_2O)

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* A fluid type may be single- or two-phase and is defined as one which has a distinct and identifiable chemical composition. Where both liquid and vapour fractions are present, both are assumed to be in chemical and phase equilibrium in the reservoir.

C_v = Concentration of gas in reservoir vapour (moles/1000 moles H_2O)

C_l = Concentration of gas in reservoir liquid (moles/1000 moles H_2O)

y = Fraction of produced steam which is reservoir vapour

CH_4, H_2S, CO_2 and NH_3 data appear to support the proposed Unit 15 model but H_2 data do not. This can be illustrated by the variation in H_2/CH_4 mole ratios and CO_2 concentrations for single fluid type samples at Unit 15 (Figure 3). Whilst the distribution coefficients predict an almost constant H_2/CH_4 ratio during boiling/mixing, the field data indicates that H_2 is relatively enriched in the low gas wells and possibly also in the higher gas wells (the H_2 enrichment may not be large enough to take the H_2/CH_4 ratio out of the range of the error bars in the higher gas wells). All other gas ratios involving H_2 show the same anomaly. Although we realise that the anomaly could be caused by relative depletions in all the other gases we think that their strong correlation with the liquid-vapour mixing model makes it more likely that only hydrogen is enriched. In the following section we examine the possible causes of this effect on the single fluid type wells.

FIGURE 3 VARIATION OF H_2/CH_4 MOLE RATIO WITH CO_2 CONCENTRATION FOR UNIT 15 STEAM SAMPLES. SOLID LINE REPRESENTS BOILING/MIXING MODEL FIT THROUGH SINGLE FLUID SOURCE SAMPLES (240°C, SOLID CIRCLES). OPEN CIRCLES REPRESENT SAMPLES FROM ANOMALOUS WELLS (SEE TEXT). SAMPLES IN AREA A ARE BELIEVED TO BE AFFECTED BY CONDENSATION.

INTERPRETATION

Distribution coefficients

The trend in H_2 data (Figure 3) may be explained by the incorrect determination of laboratory H_2 distribution coefficients. That is, H_2 has a greater affinity for the reservoir liquid phase than indicated by published data (see D'Amore and Truesdell, 1986). If the 'true' distribution coefficient is in the region 30-60 (ie similar to that of H_2S) as opposed to 597 at 240°C, the single fluid data are closely fitted by the above model. Although variations in calculated partition coefficients exist between experimental results, they are not large enough to produce H_2 distribution coefficients as low as 30-60 at 240°C. Likewise, temperature variations which may occur in the reservoir as a result of boiling cannot explain the H_2 enrichment if all gases are in phase equilibrium. This is because the order of distribution coefficients noted above is maintained across the range of interest (ie 170-240°C).

Condensation

The low gas wells generally produce steam at low flow rates which may make them prone to wellbore condensation. If condensation occurs between the reservoir steam production site and the wellhead sampling point, all gases will become concentrated in the steam phase and the proportion of less liquid soluble gases in the produced steam will increase as long as all the gases obey their distribution coefficients. Thus, it is possible that the H_2 enrichment over CH_4 could result from condensation of steam which originally had a gas content much lower than that observed. However, simulations of Raleigh-type condensation (after D'Amore and Truesdell, 1979) indicate that this process is unlikely to explain the H_2 data. Even starting with gas concentrations less than 1 ppmw total gas, and condensing more than 99.9% steam causes an insignificant increase in the H_2/CH_4 mole ratio.

Boiling/mixing conditions

As long as the model criteria discussed above are obeyed, and only a single fluid type is present, variations in the boiling or mixing conditions in the reservoir do not affect the overall ratios of gases observed at different CO_2 concentrations. For example, consider the case where (a) steam (vapour or a vapour/vaporised liquid mixture) is continuously produced from a site, (b) liquid continuously boils in reponse to pressure decline, and (c) the gases in the remaining liquid and steam phases always satisfy the distribution coefficients (McCartney and Haizlip, 1989). Even in this case, as the proportion of vaporised liquid increases relative to the proportion of vapour in the produced steam, the more liquid soluble gases will always appear enriched relative to the less liquid-soluble gases, and vice-versa.

Gas transfer kinetics

Glover (1970) found that H_2S and CO_2 do not necessarily obey their gas distribution coefficients during flashing which is probably a result of relatively (for the process) slow gas transfer kinetics between liquid and steam phases. Likewise he found that each gas was affected to a different extent suggesting that the rates of transfer vary between gases. This effect might extend to other processes and provides a possible mechanism of H_2 enrichment in the steam phase. For example, given the condensation model discussed above, if H_2 entered the condensate at a slower rate than CH_4 then the H_2/CH_4 ratio in the remaining steam phase would increase to a greater extent than suggested by our previous calculations. However, further calculations indicated that even if H_2 does not enter the condensate at all, the H_2/CH_4 mole ratio in the steam phase increases only slightly, primarily because of the poor solubility of CH_4 in the liquid phase. Thus, slow H_2 uptake by the liquid phase during condensation could not have induced the observed H_2 trend.

H_2 might leave the liquid phase at a slower rate than other gases during boiling such that the remaining liquid, when completely vapoarised, produces low gas steam anomalously enriched in H_2. Although this process might occur we do not favour it as a cause of H_2-enrichment. Conceptually, any liquid which starts boiling will continue to do so until it is all vaporised. The presence of superheating in wells from this area shows that this has happened. Likewise, steam from a number of sites in different states of boiling will probably supply a well. Linking this conceptual model with the gas kinetics hypothesis would suggest that hydrogen-enriched and -depleted steam will enter each well at the same time. This is inconsistent with the fact that low gas wells have produced only H_2-enriched steam for at least nine years.

Fluid inhomgeneities

Inhomogeneities might exist in the H_2 composition of the reservoir fluid whilst the CO_2, H_2S and CH_4 compositions remain homogenous. For example, this could be caused through mineralogical variations or through mixing with unequilibrated reinjected fluids; there is no evidence for natural recharge at Unit 15 (GEOOC, unpublished data).

Wells affected by reinjection would tend to produce low gas steam due to the higher liquid content of the source fluid and because the reinjected liquids may react with the reservoir rocks prior to vaporisation and production they may potentially become enriched in H_2 over all other gases. Increases in deuterium, oxygen-18 and tritium in produced steam, since reinjection started at Unit 15, indicate that up to 60% of production from some wells comes from reinjected fluid, and the proportions are highest in some of the low gas wells. However, samples collected from the low gas wells before reinjection began show the same H_2 enrichment as observed in later samples suggesting that reinjection is not the cause of this effect. Although reinjection might perhaps add to the general scatter of the data about the boiling/mixing model trend in Figure 3, the fact that more recent data follows the gas mole ratio distributions of earlier samples may indicate that re-injected fluids equilibrate to a large extent in the reservoir prior to production.

Steam produced from a single fluid type will have almost constant H_2/CH_4 mole ratios over a range of CO_2 concentrations if the distribution coefficients and the model conditions discussed above are obeyed. If a range of reservoir fluid types exist in the reservoir where only the H_2 composition varies and CO_2, H_2S and CH_4 are constant, then the vapour, liquid and produced steam phases may have a range of H_2/CH_4 mole ratios. Thus, the low gas samples might represent samples of high vapoarised liquid content, produced from such a range of reservoir fluid types (Figure 4). There are, however, two factors which suggest that this is not the case. Firstly, although Unit 15 wells have been monitored under varying production conditions for over nine years there is little evidence of a range of vapour-liquid mixing lines which would support the presence of a range of H_2 fluid types (see Figures 3 and 4). Many of the data which might represent examples of these fluids are in fact low gas samples affected by wellbore condensation (samples marked A in Figure 3, McCartney and Haizlip, 1989). Secondly, we have no evidence of mineralogical variations across the reservoir which might explain why H_2 concentrations should vary in the single fluid type, whilst leaving H_2S, CH_4 and CO_2 concentrations constant.

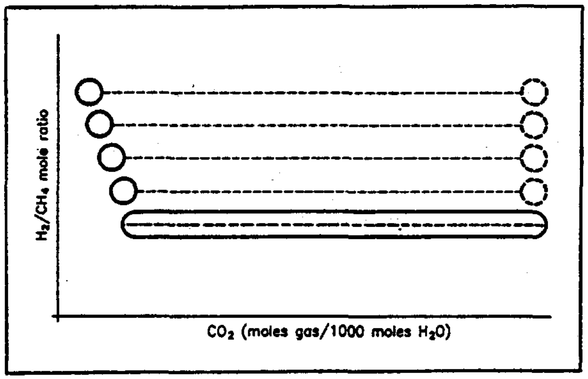

FIGURE 4 EXAMPLE OF THE DISTRIBUTION OF DATA EXPECTED FROM A FLUID SOURCE WITH INHOMOGENEOUS H_2 CONCENTRATIONS BUT HOMOGENEOUS CO_2, H_2S AND CH_4 CONCENTRATIONS. DASHED CIRCLES REPRESENT HYPOTHETICAL VAPOUR COMPOSITIONS, SHADED AREAS REPRESENT UNIT 15 DATA, AND DASHED LINES REPRESENT BOILING/MIXING LINES CONNECTING VAPOUR AND VAPOURISED LIQUID SAMPLES. NOTE THAT BECAUSE THE DISTRIBUTION COEFFICIENTS OF H_2 AND CH_4 ARE SIMILAR, THE MIXING LINES ARE ALMOST HORIZONTAL.

Fluid-solid interactions

Pruess et al (1985) presented evidence for the active production of CO_2 after the onset of boiling in the Larderello reservoir. Boiling produces a decline in gas partial pressures and results in disequilibrium between the residual liquid and the host rock. Whilst liquid remains, there is a thermodynamic driving force for gas production. If H_2 is produced at Unit 15 in this way, or H_2 is produced at a faster rate than other gases, then such a process would result in the H_2 enrichment we observe. Given the remarkable consistency of the CH_4, H_2S and CO_2 data over time to the single fluid model, we believe that H_2 is the only gas which might be significantly produced 'in excess', if the process is occurring at all. The reaction of prehnite/pyrrhotite to epidote/pyrite (Equation 2) is an example of the type which might be generating the excess H_2. This reaction has been proposed to buffer H_2 in liquid-dominated systems by Arnorsson and Gunnlaugsson (1985) and is also compatable with the mineralogy observed at The Geysers (Sternfeld, 1981).

$$4FeS + 2Ca_2Al_2Si_3O_{10}(OH)_2 + 2H_2O =$$

$$2FeS_2 + 2Ca_2FeAl_2Si_3O_{12}(OH) + 3H_2 \qquad (2)$$

Produced steam may be enriched in H_2 generated during corrosion of well casing and the steam gathering system. Minor, and in some cases more significant, condensation is common at Unit 15 and corrosion is localised where steam condenses and the condensate contacts carbon steel (eg Giggenbach, 1979). Analyses of corroded pipe indicate the presence of high levels of H_2 whilst the most common corrosion product is magnetite. This suggests that a reaction such as that in Equation 3 is important and the poor solubility of H_2 in the condensate will favour partitioning of excess H_2 into the produced steam phase.

$$3Fe + 4H_2O = Fe_3O_4 + 4H_2 \qquad (3)$$

Pyrite and pyrrhotite are also corrosion products at Unit 15 and their formation could generate H_2 (Equations 4 and 5). However, their occurrence is much less frequent than that of magnetite, and the lack of evidence for H_2S depletion in the lower gas wells suggests that Reaction 3 is the dominant mechanism of H_2 production during well casing and pipe corrosion.

$$Fe + 2H_2S = FeS_2 + 2H_2 \qquad (4)$$

$$Fe + H_2S = FeS + H_2 \qquad (5)$$

Despite the presence of corrosion products, we have performed mass balance calculations based on Reaction 3, observed corrosion rates of well casings, and cumulative steam production, which indicate that only 5-15% of the excess H_2 is likely to have been derived from well casing corrosion reactions.

EVIDENCE FROM OTHER AREAS

We have also examined steam gas mole ratio data from GEOOC's lease in the northwestern area of The Geysers and some from Larderello (D'Amore et al, 1983, 1987) where gas levels are much higher than at Unit 15. Total gas concentrations in the former area are in the higher ranges for The Geysers (quoted earlier) whilst at Larderello they typically range from approximately 17 000 to 20 0000 ppmw. As at The Geysers, CO_2 is by far the dominant gas over H_2 at Larderello (typically > 90, and 1-2, mole percent in the dry gas respectively).

We have not, as yet, performed detailed studies on either area and therefore do not suggest that the conditions present in these localities are similar to those at Unit 15. However, mole ratio data from both areas display a distribution with respect to CO_2 concentration which is similar to that seen at Unit 15 (eg H_2 appears to be enriched in lower gas wells; see Figures 5 and 6). One exception is that both H_2 and CH_4 appear to be enriched over CO_2 in lower gas steam from Larderello (Figure 6). It is unlikely that these apparent enrichments are the result of CO_2 depletion in these samples because Pruess et al (1985) have presented strong evidence to suggest that 'excess' CO_2 is present in steam from Larderello due to fluid-rock interaction during boiling.

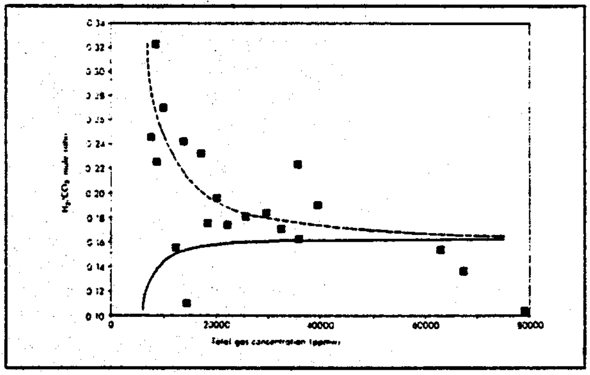

FIGURE 5 VARIATION OF H_2/CO_2 MOLE RATIO WITH CO_2 CONCENTRATION FOR STEAM SAMPLES FROM THE NORTHWESTERN AREA OF THE GEYSERS. EACH SOLID SQUARE REPRESENTS THE AVERAGE VALUE FOR A WELL. SOLID LINE REPRESENTS THE TREND EXPECTED FOR DATA DERIVED FROM A SINGLE FLUID TYPE. DASHED LINE SHOWS THE OBSERVED TREND.

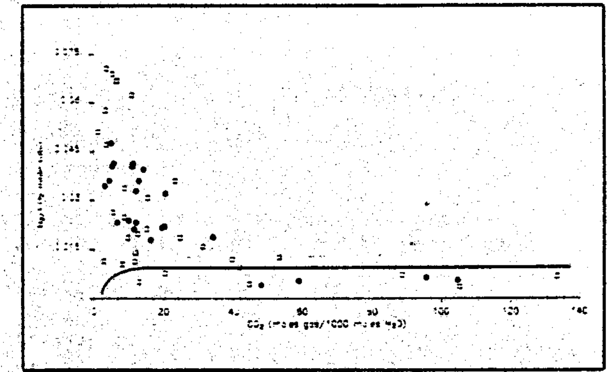

FIGURE 6 VARIATION OF H_2/CO_2 MOLE RATIO WITH CO_2 CONCENTRATION FOR LARDERELLO STEAM SAMPLES. FILLED CIRCLES (D'AMORE ET AL, 1983) AND OPEN SQUARES (D'AMORE ET AL, 1987). SOLID LINE REPRESENTS THE TREND EXPECTED FOR DATA DERIVED FROM A SINGLE FLUID TYPE.

Any of the processes discussed above for Unit 15 might be responsible for H_2 generation in the northwestern area of The Geysers and

Larderello. However, at this stage there is only evidence supporting H_2 generation through corrosion of well casings and production pipelines in the northwest Geysers. At this site magnetite, pyrite and pyrrhotite have been identified as corrosion products suggesting that the the reactions shown in Equations 3 to 5 are occurring during production. We have no information on corrosion products at Larderello but given the fact that 'excess' CO_2 appears to be generated by fluid-rock interactions in the reservoir (Pruess et al, 1985), it is possible that H_2 is also generated by such reactions. The same process would explain the behaviour of CH_4 in the lower gas wells.

CONCLUSIONS

H_2 enrichment of steam gas has been identified at Unit 15, The Geysers, and may also be occurring in other areas of both The Geysers and Larderello. At Unit 15 we have evidence that this effect may be caused, in part, by steam-well casing interactions during production although the major source of 'excess' H_2 appears to be fluid-rock interactions which take place during boiling in the reservoir.

Given the potential widespread presence of H_2 enrichment, it is evident that field data should be carefully evaluated prior to using them in H_2 influenced steam fraction, geothermometry or fluid-rock interaction calculations. In the case of Unit 15 data, we estimate that approximately 14 ppmw H_2 has been added to the steam through one of the processes considered above and this should be subtracted from low gas wells, and possibly also higher gas wells prior to use in such calculations. Errors will be largest if use is made of low gas mole ratio data incorporating H_2. If in doubt about the validity of H_2 data, we suggest non-H_2 influenced calculations are performed.

ACKNOWLEDGEMENTS

We thank Geothermal Resources International, Inc, and its wholly owned subsidiaries GeoScience Limited and GEO Operator Corporation for permission to use previously unpublished data. Thoughtful reviews by Drs A S Batchelor and J Combs are greatly appreciated. We also thank P Davies and J Pearson for help in producing the paper. We acknowledge the work of Drs A H Truesdell and F D'Amore for stimulating our interest in the behaviour of gases in vapour-dominated geothermal systems.

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