ESTIMATION OF UNDERGROUND TEMPERATURES FROM THE SILICA CONTENT OF WATER FROM HOT SPRINGS AND WET-STEAM WELLS†

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ABSTRACT. The amounts of silica in solution in hot spring pools are not accounted for by the solubility of amorphous silica, which is the precipitated silica phase under surface conditions. The existing data on compositions of hot spring solutions and solubilities of silica phases suggest that the solubility of quartz at depth is the major control. Hot spring and geothermal steam well waters ascending rapidly to the surface become supersaturated with respect to quartz because of rapid cooling, separation of steam, and sluggish deposition of quartz and other crystalline SiO_2 phases.

The silica content of a boiling water discharged at the surface can be used to estimate the underground temperature of last equilibrium with quartz, provided a correction is made for the steam that forms during ascent of the solutions from the relatively high pressure environment at depth. Where measurements have not been made of the percent of steam that separates from a given amount of solution, estimates of this quantity may be made using steam tables and assuming adiabatic cooling along the vapor pressure curve either at constant enthalpy or constant entropy. Using such steam corrections, curves were constructed showing dissolved silica measured in water discharged at the surface versus underground temperatures of last equilibrium with quartz. The method was applied to three wet-steam wells, and good agreement was obtained between the estimated and measured maximum temperature at depth.

INTRODUCTION

It is the object of this paper to show that the silica content of many boiling or near boiling hot springs is controlled by the solubility of quartz at depth, rather than by the solubility of amorphous silica at and near the surface of the ground. In such springs, the silica content of the surface waters may allow an estimate of the temperature at which the water was last in equilibrium with quartz at depth.

Bödvarsson (1960) used the silica content of hot spring waters as a crude indicator of relative temperatures at depth. However, the method was not quantitative until the solubility of quartz had been measured over the full range of temperatures and pressures likely to occur in hot spring systems. Recently, Mahon (1966) has had remarkably good success using the quartz solubility curve of Morey, Fournier, and Rowe (1962) to estimate maximum temperatures at depth from the "enthalpy corrected" silica content of water in wet-steam produced from wells drilled at Wairaki, New Zealand. Mahon's results support the hypothesis that quartz controls the silica content of hot spring waters deep underground. His results also suggest that the dissolved constituents common in chloride types of hot spring waters do not significantly change the solubility of quartz from what it is in pure water.
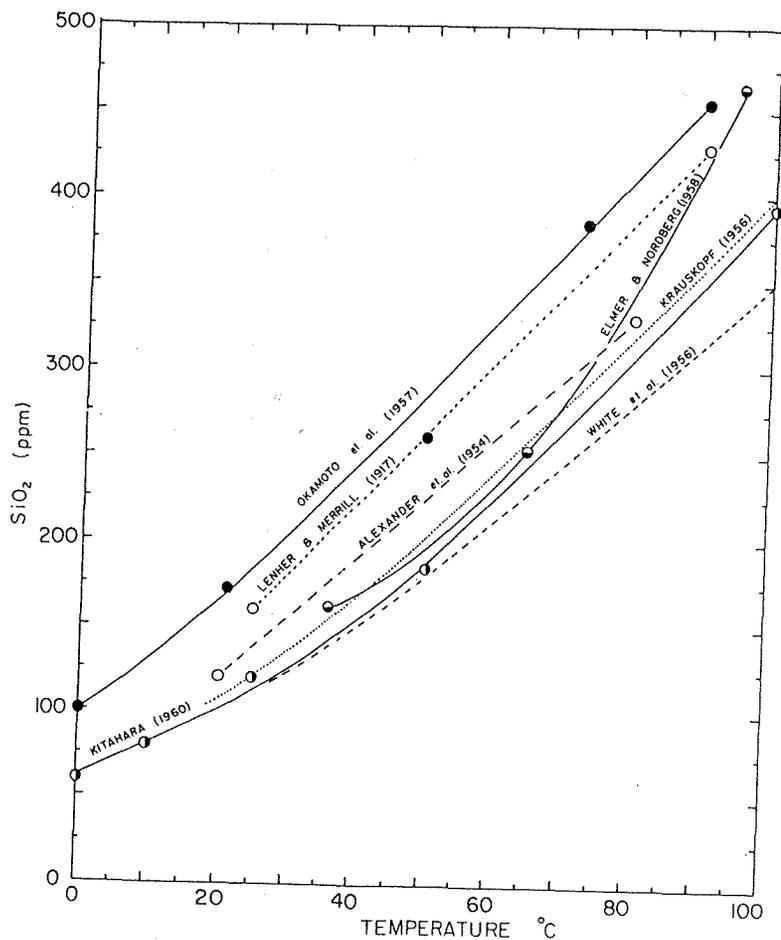
Fig. 1. Results obtained by various workers for the solubility of amorphous silica in water between 0 and 100°C.

SILICA IN SURFICIAL HOT SPRING WATERS

It is a common observation that silica deposited from hot spring waters in surface pools is amorphous. In view of this fact it is interesting to compare the dissolved silica content of hot spring waters with the results of laboratory studies of the solubility of amorphous silica. Some of the more recent work on the solubility of amorphous silica in water at temperatures between 0 and 100°C is shown in figure 1. A great diversity of results obtained by different investigators is apparent. Morey, Fournier, and Rowe (1964) considered this range of experimental data and concluded that the results of Kitahara (1960a) combined with the higher temperature results of Hitchen (1935) give the best values for the solubility of amorphous silica.

In 1947, Murata found that the quantity of silica determinable by standard colorimetric methods in aged-water samples from Steamboat Springs, Nevada, was much less than the total silica content determined gravimetrically (White, Brannock, and Murata, 1956). In contrast, it was later found by Murata and White that the values for colorimetrically determinable silica in fresh samples were approximately equal to those for total silica determined gravimetrically. A series of detailed observations and measurements on the state of silica in hot spring waters were then carried out in the field by Brannock and White (White, Brannock, and Murata, 1956). In 1961, Morey and others reported upon the colorimetrically determinable silica in water from several hot springs and geysers in Yellowstone National Park, Wyoming. This work was continued in 1962 and 1963. Many of the results are shown in figure 2, where the silica contents of various actively discharging hot springs and geysers are plotted versus the temperature of the water in the pools.[1] The circles indicate colorimetrically determined[2] silica measured at poolside immediately after collection. The triangles show the total dissolved silica content of each water sample.[3] A curve showing the solubility

[1] Tables of data from which figure 2 was constructed will be published elsewhere.
[2] The molybdenum blue method was used as described by Shapiro and Brannock (1956).
[3] Total silica in solution was determined after treating the solution with NaOH to convert all polymeric silica to colorimetrically reactive silica.
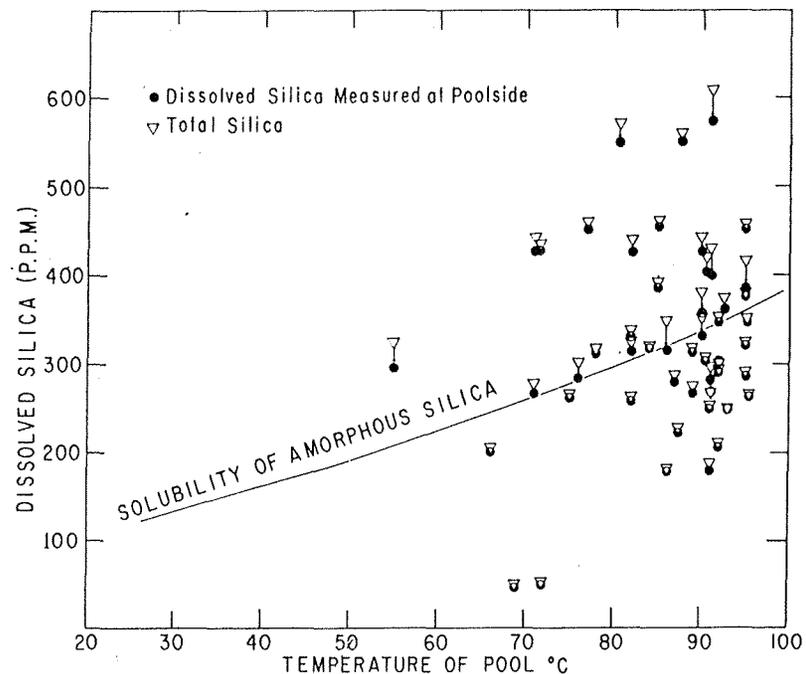

Fig. 2. Dissolved silica in water from hot springs and geysers in Yellowstone National Park, Wyoming.

of amorphous silica taken from Kitahara's (1960a) results also is included.

Inspection of figure 2 shows that many hot spring and geyser waters in Yellowstone are undersaturated with respect to amorphous silica at the temperatures of the pools. Thus, it is difficult to account for the dissolved silica contents of the waters on the basis of equilibrium with amorphous silica. Furthermore, many springs, such as the two listed in table 1, now have compositions essentially identical to those reported by Gooch and Whitfield in the year 1888. This indicates that either equilibrium or steady-state conditions have prevailed at depth for a long time and suggests that the dissolved silica in the water is controlled by silica-bearing phases other than amorphous silica.

In view of the occurrence of pools containing waters that are unsaturated with respect to amorphous silica, how did the amorphous opaline sinter, which lines many of these pools, originally form? There are many possible explanations. Most of the undersaturated waters plotted in figure 2 are from geysers; amorphous silica may deposit around geysers where silica-bearing waters cool and evaporate during the interval between geyser eruptions. Other, non-erupting springs may have undergone temporary periods with little or no water discharge. When this happens, temperatures in surface pools are likely to decrease markedly and silica precipitates from the cooling water. The authors have observed this behavior in a few pools in Yellowstone Park. Amorphous silica-lined pools may have formed in other hot springs at an early stage in their history: in volcanic terrain, hot water surging up along a newly formed crack may encounter unaltered volcanic glass. Very large concentrations of silica may dissolve from such glass until the glass becomes converted to crystalline phases and/or protected from further attack by a coating of newly formed minerals such as quartz and calcite. Thereafter, the silica content of the hot spring water may remain fairly constant at a lower concentration controlled by the dissolution of crystalline silicates.

TABLE 1

Partial chemical analyses of waters from Yellowstone National Park
(concentrations in parts per million)

	Artemisia Geyser 92°C (boiling)			Great Fountain Geyser 95°C (superheated)		
	1888*	1935**	1961***	1888*	1935**	1960***
SiO_2	275	268	272	318	316	325
Na	389	422	390	335	366	334
K	16	16	16	15	15	14
Cl	300	289	293	351	343	340

* Gooch and Whitfield (1888)
** Allen and Day (1935)
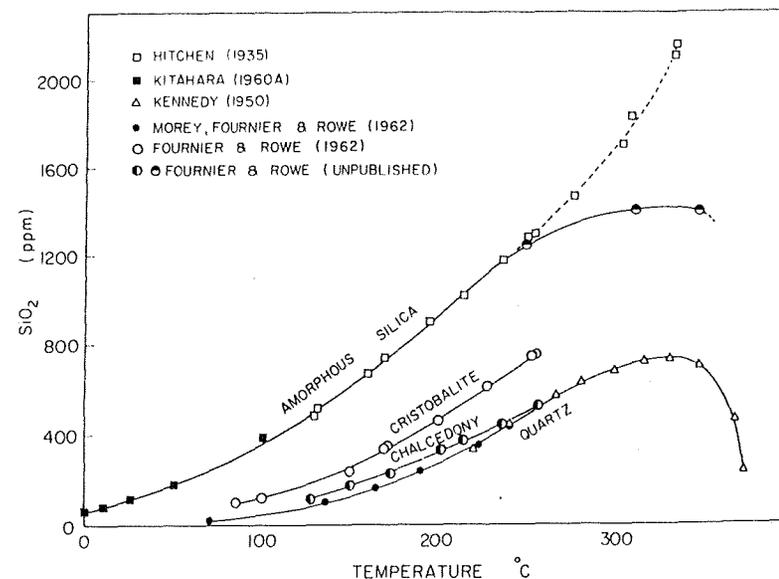*** Unpublished results of Fournier, Rowe, and Morey

Fig. 3. The solubilities of quartz, chalcedony, cristobalite, and amorphous silica at the vapor pressure of the solution.

STATE OF SILICA AT DEPTH

Drilling in numerous hot spring localities has shown that temperatures commonly increase rapidly with depth, and increased temperatures greatly increase the rate at which amorphous silica is converted to cristobalite, chalcedony, and quartz (Carr and Fyfe, 1958; White and Corwin, 1961). In Yellowstone most of the hot spring systems are hundreds, perhaps thousands of years old (Allen and Day, 1935), and amorphous silica is unlikely to persist at depth in such systems. In core from a drill hole 406 feet deep in the Upper Geyser Basin, a mixture of quartz and amorphous silica was found at a depth of only 7 feet (Fenner, 1936). Opal is dominant near the surface but does not occur below about 86 feet (White, 1955). Quartz is mainly the fine-grained chalcedonic variety near the surface, but megascopic crystals of quartz line cavities below about 92 feet, and veins of quartz and adularia are present in much of the core. Similarly, at Steamboat Springs, Nevada, amorphous silica generally occurs only at depths shallower than 50 to 90 feet; at greater depths B cristobalite, chalcedony, and quartz occur instead of amorphous silica (White, Thompson, and Sandberg, 1964).

EXPERIMENTALLY DETERMINED SOLUBILITIES AT HIGH TEMPERATURES

Figure 3 shows the solubilities of quartz and other silica phases at the vapor pressure of the solution. It has been pointed out by Kennedy (1950) that the solubility of quartz in water is strongly de-

pendent upon the density of the solvent and that at high temperatures along the vapor pressure curve, dissolved silica reaches a maximum and then decreases as the density of the solution rapidly decreases with further increase in temperature. Unpublished measurements by Fournier and Rowe (fig. 3) of the solubility of amorphous silica above 250°C follow a trend similar to the trend found for quartz and are much lower than the results of Hitchen (1935). Hitchen determined dissolved silica gravimetrically and thus was unable to distinguish truly dissolved silica from colloidal particles that may have been present.

Figure 3 shows that above 150 to 200°C very large concentrations of silica may dissolve from amorphous silica, and equally large concentrations of dissolved silica possibly may be leached from volcanic glass. The maximum solubility is about 1400 parts per million (ppm) at 330°C. However, as was previously pointed out, glass and opal are eventually converted to crystalline phases at such high temperatures, and their control of dissolved silica at depth is likely to be relatively short-lived relative to the ages of most hot spring systems.

The maximum concentration of silica likely to be found at depths where quartz controls the dissolved silica is about 725 ppm at 330°C. Kitahara (1960b) reported greatly increased quartz solubility in 1 to 7 percent NaCl solutions above 300°C. However, Kitahara had silver in contact with the NaCl solutions, and we found that NaCl will react with silver above 300°C to produce silver chloride, sodium hydroxide, and hydrogen. Thus, Kitahara's (1960b) high solubility results reflect the effective addition of NaOH to the system. The present writers have checked the effect of NaCl on the solubility of quartz, using a gold-lined pressure vessel, and find that at the vapor pressure of the solution, solubility results in 2M NaCl solutions are essentially identical to results obtained using pure water.

BEHAVIOR OF A SOLUTION AS IT MOVES TOWARD THE SURFACE

Let us assume that an increment of solution at some arbitrary high temperature below the critical temperature starts to move toward the surface. As it moves upward, the hydrostatic head will decrease, and at some place the pressure will become low enough for boiling to occur, provided little or no heat is lost by conduction into the wall rock. A certain amount of steam or gas will form, and the system will decrease in temperature owing to the latent heat of vaporization required to form the gas phase. The process will continue as the increment of solution moves toward the surface and boiling will take place at lower and lower temperatures in response to a steadily decreasing hydrostatic head. Eventually the solution may issue forth at the surface at the boiling temperature for the prevailing atmospheric pressure. The non-volatile constituents of the solution, including silica, will have become concentrated in the residual liquid; the extent of non-volatile concentration will vary depending on the manner of cooling.

If equilibrium is maintained between the liquid and gas as the solution moves toward the surface, and no heat is lost to the wall rock, reversible adiabatic cooling will occur at constant entropy. This may be thought of as reversible boiling in response to small changes in pressure applied very slowly. On the other hand, if equilibrium is not maintained between the liquid and gas, and no heat is lost to the wall rock, irreversible boiling and expansion of the Joule-Thompson type may occur. In this event the water will cool adiabatically, and the final enthalpy of the system will be the same as the initial enthalpy (Zemansky, 1951, p. 216-219). In old hot spring systems, where the wall rock has been heated for a long time, the removal of heat from the hot waters by conduction into the wall rock probably is negligible in comparison to the decrease in temperature caused by boiling. This is particularly true where the rate of flow from a hot spring is large. Thus, adiabatic cooling at either constant entropy or enthalpy may be approximated in underground hot spring systems.

Figure 4 shows the silica concentration attained in the residual liquids as three solutions, initially at 210, 260, and 330°C respectively, are cooled either at constant entropy or at constant enthalpy. For instance, a solution underground in equilibrium with quartz and vapor
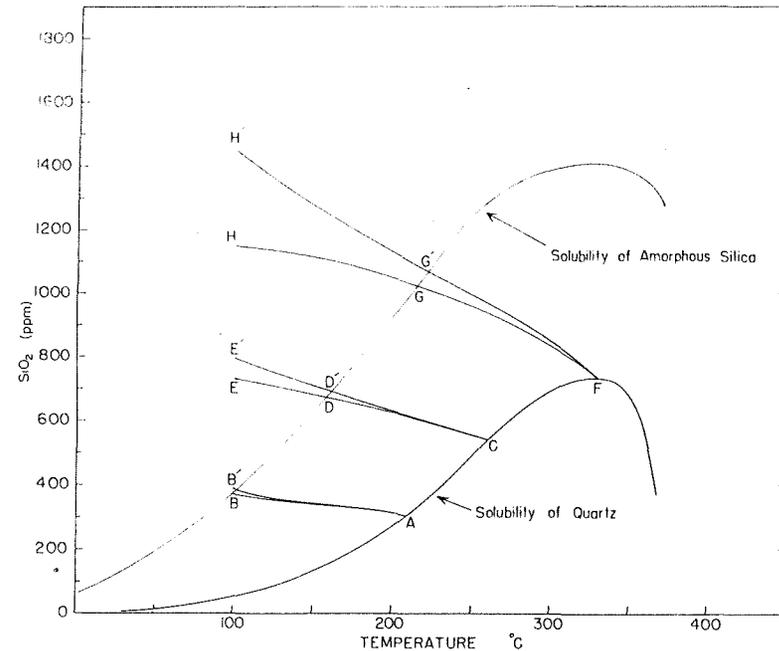

Fig. 4. Silica concentrations in residual liquids attained by adiabatically cooling three solutions initially in equilibrium with quartz at the vapor pressure of the solution and 210, 260, and 330°C respectively. Curves AB, CDE, and FGH are for cooling at constant entropy, and curves AB', CD'E', and FG'H' are for cooling at constant enthalpy.

at 260°C would contain about 540 ppm dissolved silica (fig. 4, point C). If quartz or other solid silica phases do not precipitate as the solution moves from there to the surface, the concentration of silica in the residual liquid will increase as steam continuously separates and the solution cools by adiabatic expansion. If steam forms at constant entropy (reversible expansion), the silica concentration in the residual liquid will follow the curve CDE, and the final silica concentration in hot spring water emerging at 100°C would be about 730 ppm. In the process about 26 percent of the initial liquid water would be converted to steam. On the other hand, if steam forms at constant enthalpy (irreversible expansion), the silica concentration will follow the curve CD'E', and the final silica concentration at 100°C might attain about 795 ppm after formation of about 32 percent steam. At point D (or D') the solubility curve for amorphous silica is intersected, and below about 159 to 162°C the solution becomes increasingly supersaturated with respect to amorphous silica.

Similarly, for a solution originally containing 725 ppm silica at 330°C (fig. 4, point F), the final concentration at 100°C might reach 1149 ppm after formation of 37 percent steam by adiabatic expansion at constant entropy (point H). Expansion at constant enthalpy might yield about 1430 ppm at 100°C after 50 percent steam formation. The solution would become supersaturated with respect to amorphous silica at temperatures below about 215 or 221°C (point G or G'), and gelatinous silica would probably precipitate in channelways underground owing to the very great degree of supersaturation that would be attained there.

Water in equilibrium with quartz underground at about 210°C would become just saturated with respect to amorphous silica at 100°C. Starting at 210°C, it makes little difference whether steam forms at constant entropy (fig. 4, curve AB) or at constant enthalpy (curve AB'). Underground water in equilibrium with quartz at less than 210°C would yield hot spring water at the surface that was undersaturated with respect to amorphous silica.

In the foregoing discussion it was assumed that quartz and other crystalline phases of silica stop precipitating as a solution cools below some arbitrary high temperature during its ascent from a hot region underground to a cooler environment near the surface. This assumption is supported by chemical analyses showing that most hot spring waters are greatly supersaturated with silica in respect to the solubility of quartz and other silicates, and by drill hole data showing that veins of hydrothermal quartz occur at depth but not near the surface. The temperature of "last quartz precipitation" should be different for each hot spring system and should depend on many factors including: the underground thermal environment, the rate of flow of the solution, and poorly understood possible effects of various dissolved constituents which may either promote or hinder the precipitation of silica.

Drilling has shown that in many hot spring systems temperatures steadily increase with depth along the boiling point curve until the temperature levels off at some value, which is different for each hot spring system, and little additional increase in temperature is found at greater explored depths (White, 1961). Thus, an increment of solution rising from depth might remain at a fairly constant high temperature for a relatively long time compared with the time that it takes that solution to continue to the surface after first encountering boiling conditions. Such a solution could attain equilibrium with quartz owing to a long "residence time" at the constant high temperature deep underground. Later, little or no precipitation of silica might occur during the quick trip to the surface after boiling and cooling commenced. This certainly happens with water produced from many steam wells. Dissolved silica in the solutions entering the wells at depth appears to be controlled by equilibrium with quartz, but we know of no examples of quartz deposition on the walls of producing steam wells. However, amorphous silica may deposit where solutions become greatly supersaturated in respect to that phase.

ESTIMATION OF UNDERGROUND TEMPERATURES ASSUMING EQUILIBRIUM WITH QUARTZ AT DEPTH

To use the silica content of water from a hot spring or wet-steam well to estimate an underground temperature it is necessary to correct for the amount of steam that separates from the water as it rises from a region of high pressure underground to atmospheric pressure at the surface. Ideally, the proportion of discharged steam to discharged water should be measured. This is not always practical or possible, particularly for hot springs, and estimates must be made.

One way to deal with this problem is to assume adiabatic cooling at either constant enthalpy or constant entropy and then utilize steam tables such as those of Keenan and Keyes (1936) to determine the amount of steam that would form upon transporting water from boiling conditions at high pressure to boiling at atmospheric pressure. Combining these data with the data for the solubility of quartz at the vapor pressure of the solution as shown in figure 3, it is possible to determine what the final silica content of a water would be after cooling from any given high temperature. For instance, upon adiabatic cooling at constant enthalpy from 260°C and 46.3 atm to 100°C and 1 atm, 32 percent of the initial water would be converted to steam. The solubility of quartz at 260°C and 46.3 atm is about 540 ppm. After losing 32 percent of the water portion of this solution the concentration of silica in the residual solution would be:

$$\frac{540}{0.68 \ (10^6 - 540) + 540} \approx \frac{540}{68000},$$

which is about 794 ppm. Values so derived for residual silica concentrations may be plotted in place of actual quartz solubilities on a
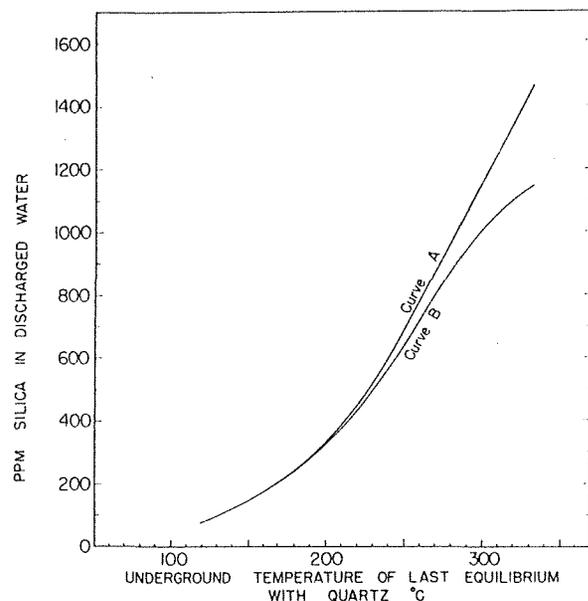
Fig. 5. Curves for estimating underground temperatures from the silica contents of boiling waters discharged from hot springs and wells. Curve A is for solutions cooled at assumed constant enthalpy, and curve B is for solutions cooled at assumed constant entropy.

dissolved silica versus temperature diagram. The resulting curve can then be used to estimate underground temperatures from the silica contents of waters discharged at the surface. Figure 5 shows one such curve for cooling at constant enthalpy and a second curve for cooling at constant entropy. According to figure 5, a boiling water discharging at 1 atm pressure and containing 700 ppm silica indicates an underground temperature between 250°C (cooling at constant enthalpy) and 257°C (cooling at constant entropy).

In using the curves shown in figure 5 to estimate underground temperatures, it must be remembered that the procedure is valid only for systems in which cooling is mainly by steam formation, and that the temperature obtained will be the temperature at which the solution was last in equilibrium with quartz. The underground temperature determined in this way will be too low if quartz continued to precipitate as the water moved toward the surface and cooled. The silica content of hot spring waters will also yield an anomalously low estimated temperature at depth if large quantities of gas such as CO_2, H_2S, et cetera, separate from an ascending solution because this gas will withdraw heat from the system and cause less steam to form. Of course, mixing with relatively dilute and/or cool near-surface water will result in low estimated temperatures at depth.

In Yellowstone National Park, the silica contents of the hot spring waters suggest a maximum temperature of last equilibration with quartz of about 245°C in the Norris Geyser Basin, and about 190 to 215°C in the Upper Geyser Basin. Drilling in the respective areas (Fenner, 1936) yielded temperatures of 205 and 180°C. However, drilling was terminated before the maximum temperature at depth was attained at Norris, and this may also be true in the Upper Geyser Basin.

Table 2 represents a comparison of estimated and measured temperatures at depth for three wells and a hot spring. The dissolved silica content of water from the three wells allows fairly good estimation of the actual underground temperatures. The estimate from hot spring number 24 at Steamboat Springs is too high, possibly due to dissolution of amorphous silica as the water encountered silica gel near the surface: the solubility of amorphous silica is 345 ppm at 94°C (the temperature of the pool), precisely the concentration of dissolved silica found in hot spring number 24. Alternatively, at 170°C the rate of growth of quartz may be so slow that solutions may become supersaturated with silica owing to a rapid rate of alteration of feldspar and hornblende to mica, montmorillonite, chlorite, and calcite with the concomitant release of silica. Schoen and White (1965) found such alteration products in core from diamond drill holes at Steamboat Springs, Nevada, and concluded that the present-day spring waters are producing that hydrothermal alteration.

CONCLUSIONS

In view of the foregoing discussion, the following generalizations are made. The silica content of waters from discharging hot springs

Table 2

Temperatures at depth and dissolved silica after cooling by steam formation

	Dissolved silica (ppm)	Estimated Temperature °C	Maximum measured temperature in drill hole °C
Well, Seltun, Iceland[1]	425	215 to 220°	220°
Well, Kairaki, New Zealand[2]	660	246 to 252°	250°
Well, Steamboat Springs, U.S.A.[3]	245**	178 to 180°	170°
Spring no. 24, Steamboat Springs,[4] U.S.A.	345	201 to 205°	170°

[1] Bödvarsson (1961)
[2] A. J. Ellis, oral communication.
[3] White (1961)
[4] White, Brannock, and Murata (1956)
** This value for dissolved silica is questionable because the analysis was not performed immediately after collection of the water sample (Donald E. White, oral communication).

and steam wells may be used to estimate temperatures at depth, provided that the water is cooled chiefly by steam formation, and *not* by conduction, convection, by separation of an abundant, non-aqueous gas phase, or by mixing with dilute, cooler water near the surface. If the silica content of a hot spring water is less than the solubility of amorphous silica at the temperature of the pool, amorphous silica is likely to dissolve from old amorphous silica deposits near the surface and cause an anomalously high estimate of temperature at depth. On the other hand, if the silica content of a hot spring water is greater than the solubility of amorphous silica, estimated temperatures of last equilibrium with quartz are likely to be fairly close to or lower than the maximum temperature at depth.

The silica contents of water discharged from steam wells yield estimated underground temperatures in good agreement with measured temperatures, even where enthalpies of the discharging water-steam mixtures have not been measured. In New Zealand, where enthalpies of discharging water-steam mixtures have been measured, W. A. J. Mahon (written communication, June 15, 1966) has found that the "silica" method consistently yields estimated temperatures that are within 2°C of the maximum measured temperature at depth.

ACKNOWLEDGMENTS

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