CONF-811026--7

MASTER

NOTICE

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PREDICTING DIAGENETIC HISTORY AND RESERVOIR QUALITY IN THE FRIO FORMATION OF BRAZORIA COUNTY, TEXAS AND PLEASANT BAYOU TEST WELLS[1]

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ABSTRACT

Good-quality geothermal reservoirs displaying secondary porosity exist on the upper Texas coast (Brazoria County), site of the Pleasant Bayou No. 1 and No. 2 wells, and are attributed to a moderately stable mineral assemblage, normal geothermal gradients, and low in situ pH. Major authigenic minerals are calcite, quartz, and kaolinite. Detrital feldspar has been extensively albitized at depth. Major diagenetic events overlapped, occurring in the general order--precipitation of calcite, formation of quartz overgrowths, albitization, leaching of calcite to form secondary porosity, and precipitation of kaolinite. Seventeen Brazoria County water analyses including two from the Pleasant Bayou No. 2 well were thermodynamically tested. Predictions based on equilibrium thermodynamics add new insight on mineral stabilities and are consistent with the paragenetic sequence developed from petrographic data. Early precipitation of calcite at shallow depths of burial is predicted. Low temperature and low in situ pH explain the general absence of chlorite. The use of solution-mineral equilibria as a predictor of reservoir quality is inconclusive, but well-defined clustering of waters indicates that identification of regional trends will probably be possible. Shallow waters may be the key to predicting reservoir quality at depth.

INTRODUCTION

Diagenetic studies of Gulf Coast Tertiary rocks, funded by DOE and conducted at the Texas Bureau of Economic Geology since 1977, have produced much data on the regional variations in detrital and authigenic mineralogy (Bebout et al, 1978; Loucks et al, 1979a, 1979b, 1980). Sandstone consolidation studies have progressed from broad regional reservoir quality assessment stressing sandstone petrography to detailed comparison of sandstone mineralogy and elemental composition of calcite from selected areas. The next logical step in the effort to understand diagenesis and predict reservoir quality is to investigate the role of pore fluids or aspects of water-rock interaction. Consequently, current work is more geochemical in nature than earlier work. The goal is to relate water-rock interaction to sandstone diagenesis by thermodynamically testing formation waters for stability with respect to reaction pairs involving authigenic and detrital minerals and for supersaturation with respect to authigenic minerals. In other words, a solution-mineral equilibria approach was used. Thermodynamic functions for nine key minerals and nine ionic species have been estimated in 25° or 50° C increments over the temperature range 25° to 350° C. Seventeen Brazoria County Frio Formation waters from two fields and the Pleasant Bayou No. 2 well were tested (fig. 1). This paper briefly presents preliminary results, subject to update of thermodynamic data.

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[1] Publication authorized by the Director, Bureau of Economic Geology, The University of Texas at Austin, 78712.

DETRITAL AND AUTHIGENIC MINERALOGY

Most Brazoria County Frio sandstones are lithic arkoses and feldspathic litharenites along with a significant number of subarkoses and sublitharenites (Loucks et al, 1980). Detrital constituents are mainly quartz, lesser amounts of feldspar and rock fragments, and minor matrix material. Plagioclase is the dominant feldspar, although minor K-feldspar (mostly microcline) is also present. Volcanic rock fragments are the dominant rock-fragment variety. Mudstone, shale, chert, and other sedimentary fragments occur less commonly. Depositional matrix, detrital material finer than 20 μm, is composed mainly of mixed layer smectite/illite. Diagenesis has modified the detrital mineralogy through precipitation of cements and alteration and dissolution of detrital grains and authigenic cements. Major authigenic minerals are calcite, quartz, and kaolinite. Minor phases are albite, chlorite, laumontite, and unidentified clay coats (Loucks et al, 1980). Ca-montmorillonite clay coats have been identified in the Catahoula Formation, the updip equivalent of the Frio Formation (Galloway and Kaiser, 1980).

DIAGENETIC HISTORY

Multiple generation of authigenic calcite is the rule in the Frio sandstones of Brazoria County. Major episodes of calcite precipitation occurred in several stages throughout the burial history. These episodes are reflected in the presence of multiple, distinctive calcite compositions at various depths (Loucks et al, 1980). At least four unique calcite cement and grain replacement compositions were distinguished in the Pleasant Bayou No. 1 and No. 2 wells. Two phases of non-ferroan calcite are present, one shallow, preceding quartz overgrowths, and the other at shallow to intermediate depths, following quartz overgrowths (fig. 2). Shallow Pleasant Bayou samples (less than 1,220 m) contain ferroan calcite as poikilotopic cement and non-ferroan calcite as grain replacements.

Leaching of authigenic calcite is also episodic and is responsible for most of the secondary porosity in these sandstones. The most intense leaching follows formation of quartz overgrowths which, based on oxygen isotope data, formed at temperatures of 75° to 80°C (Loucks et al., 1980). Petrographic evidence suggests that authigenic albite cement formed contemporaneous with quartz. Authigenic kaolinite occurs mainly as a filling in secondary pores displaying a vermicular habit. In many cases, it appears to have begun forming as a plagioclase replacement. Pore-filling kaolinite follows quartz overgrowths and much of the leaching; it formed at approximately 100°C, based on isotopic analysis, and appears to be the last major authigenic cement precipitated (fig. 2).

Diagenetic processes believed responsible for generating constituent components of authigenic cements or conditions necessary for their precipitation or dissolution involve reaction of detrital components with pore fluids--organic maturation, smectite/illite transition, and albitization of detrital feldspar. Organic maturation and the smectite/illite transition take place mainly in

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shales whereas albitization goes forward in sandstones. Based on isotopic data and trends in feldspar composition with depth, albitization is initiated at depths slightly deeper and presumably at slightly higher temperatures than the smectite/illite transition (Loucks et al, 1980), which according to Freed (1980) begins at approximately 90°C in Brazoria County. Microprobe data from Brazoria County show that albite (An 0-10) begins to appear in quantities greater than 25 percent at depths in excess of 3,580 m corresponding to a temperature of approximately 110°C, whereas K-feldspar remains largely unaltered to this depth. Below about 3,720 m virtually all plagioclase has altered to albite and K-feldspar is only rarely preserved (fig. 3). Albitization of plagioclase is probably initiated at about 100°C. It is an important source of calcium; however, total albitization of detrital plagioclase would probably release calcium sufficient to account for no more than 40 percent of the calcite cement postulated to be present prior to leaching. Clearly, additional sources of calcium must be sought. Paragenetically, albitization follows precipitation of quartz overgrowths and, based on microprobe data, regional geothermal gradients, and isotopic data, occurs nearly contemporaneous with kaolinite precipitation.

RESERVOIR QUALITY

Authigenic minerals are the key to reservoir quality because they occlude porosity and reduce permeability. Predicting their behavior should eventually lead to prediction of reservoir quality. The presence or absence of these minerals is controlled by bulk composition, temperature, and pore-fluid composition.

Bulk Composition

According to Loucks et al (1979a, 1980), moderately stable mineral assemblages, such as those of the Frio sandstones on the upper Texas coast and in Brazoria County, are necessary to preserve high-quality reservoirs in the deep subsurface. Because of their susceptibility to dissolution, some optimum content of unstable components (20 to 30 percent) is deemed necessary for development of secondary porosity. However, if the percent of unstable grains is too high, the system becomes highly reactive and porosity is commonly occluded by abundant cements.

Chemical and mechanical instability of framework grains is enhanced by higher temperatures. Higher-than-normal geothermal gradients cause specific diagenetic events to occur earlier in the consolidation sequence and specific reactions to be more intense. Matrix, especially that of volcanic origin, is more reactive. Furthermore, abundant mechanically unstable rock fragments are commonly deformed producing pseudomatrix, which reduces porosity and permeability. Thus, normal thermal gradients in Brazoria County (0.75°C/30.5 m above and 1.38°C/30.5 m within hard geopressure) contribute to its superior reservoir quality.

Water-Rock Interaction

Thermodynamic Data Key minerals are taken from the work of Loucks et al (1979b, 1980). Ferroan calcite and plagioclase compositions are based on their work. Lacking analyses for layer silicates, formulas for chlorite and Ca-montmorillonite were chosen, based on the work of Carroll (1970) and Galloway and Kaiser (1980) (table 1). Because of the fragmentary nature and lack of availability of thermodynamic data, especially for the layer silicates, thermodynamic functions were estimated. Methods differ depending on mineral class and availability of published data. The calculation of change in free energies of formation (ΔG_f) for ferroan calcite and plagioclase is essentially the estimation of ΔG_f for solid solutions (Kaiser et al, 1981). Calculating values of thermodynamic functions for layer silicates presents special problems. Few thermodynamic data are available because of compositional and structural complexity. Free energies can be computed at elevated temperatures if average heat capacity, enthalpy of formation at 25°C, and log K of formation at 25°C are known. Heat capacity was calculated using a structural algorithm (Helgeson et al, 1978). Enthalpy was estimated using a graphical method of Helgeson (Kaiser et al, 1981). Free energy of formation was estimated using the method of Tardy and Garrels (1974). Activities of ionic species were calculated by computer using an updated version of SOLMNEQ (solution-mineral equilibrium computations) (Kharaka and Barnes, 1973).

Water Composition Analyses of 17 Brazoria County waters, 13 Chocolate Bayou, two Halls Bayou, and two Pleasant Bayou No. 2 waters (fig. 1) were selected from the literature (Kharaka et al, 1977, 1979) and tested thermodynamically. Waters come from a depth range of 2,624 to 4,740 m and have a temperature range of 94° to 150°C. They are NaCl waters with a total dissolved solids content ranging from 32,000 to 132,000 mg/L. The two Pleasant Bayou waters are 132,000 and 108,000 mg/L from 4,462 m and 4,740 m, respectively. Four mole ratios of particular interest in evaluating the effects of geologic membranes are compared graphically with seawater in figure 4.

Stability Relations Brazoria County waters were tested for relative stability against six reaction pairs (table 2) by plotting log activity ratios and products for each water on the appropriate stability diagram. All waters are plotted assuming a water activity of 1. Stability relations are shown at 100° and 150°C at 1 bar representing hydropressured conditions and at 600 bars representing geopressured conditions (figs. 5, 6, 7, 8, 9, 10, 11). Figures 8 and 11 especially illustrate the role of pressure in mineral stability, reflecting the number of waters involved in the reaction and water's more positive free energy at elevated pressure (Fisher and Zen, 1971).

Calcite equilibrium largely governs reservoir quality in Brazoria County. Shallow iron-poor calcite and ferroan calcite cements following one another can be explained geochemically. In general, calcite becomes more insoluble at higher temperature, ferroan calcite more so than pure calcite. Waters with a $[Ca^{2+}]/[Fe^{2+}]$ ratio of less than about 10,000 will first yield ferroan calcite. Therefore, to form pure calcite the virtual absence of ferrous iron is required. Pure calcite is stable in surface seawater (fig. 5) and probably would be favored before initiation of the smectite/illite transition in sulfide-rich waters. Thereafter, ferroan calcite might be expected in sulfide-poor geothermal waters at intermediate and deeper burial depths. However, ferroan calcite can also form at shallow depths, perhaps to the depth of circulation of meteoric ground water, where sulfate reduction is inhibited.

All Brazoria County waters at subsurface temperatures plot in the ferroan calcite field (fig. 5). Waters from the shallowest depths cluster closest to the calcite stability field, consistent with the occurrence of shallow iron-poor calcite. The deepest waters cluster well into the ferroan calcite field, yet late-stage ferroan calcite is absent in the Pleasant Bayou wells. Here the critical parameter for carbonate equilibrium is pH. The precipitation of calcite is difficult, if not impossible, at low pH. Using a mass-balance approach, Kharaka et al (1979) calculated an in situ pH of 4.1 for Pleasant Bayou waters. At an equilibrium pH of 4, concentrations of total dissolved carbonate species of approximately 60,000 mg/L are required for calcite precipitation --concentrations that seem unrealistically high. The point is that high pH favors calcite precipitation and has a direct bearing on the timing of calcite precipitation and the depth at which it occurs. Because pH generally decreases with depth, calcite precipitation should be expected early and at shallow depths. The absence of late-stage, porosity-occluding ferroan calcite in the Pleasant Bayou wells is thus attributed to low pH. Indeed, shallow waters (numbers 1, 2, 3, 4, 5, 13, 14, 19, and 20) plotting less deeply into the ferroan calcite field may be an indicator of superior reservoir quality at depth.

Brazoria County waters plot in the chlorite stability field (fig. 6) contradictory to petrographic evidence. Chlorite is essentially absent in Brazoria County sandstones. Factors controlling chlorite stability relative to kaolinite are pH and Mg-Fe log activity product. Note that hydrogen ion activity is to the 9.2 power (fig. 6). At a pH of 4 and water activity of 0.87 Pleasant Bayou geopressured waters are shifted into the kaolinite field (fig. 7). At pH's of 4 to 6, not unreasonable in situ values for subsurface waters, all tested waters would plot in the kaolinite stability field, consistent with petrographic evidence. By implication then, calcite leaching must have preceded and/or occurred contemporaneous with precipitation of kaolinite cement. The conspicuous absence of chlorite is explained by temperature--it is too low in Brazoria County for chlorite stability. However, Pleasant Bayou waters would fall in the chlorite field as temperatures rise above 150°C to about 175°C (fig. 7).

Insofar as kaolinite is an indicator of acidic waters and undersaturation with respect to calcite, then shallow waters clustering toward the kaolinite stability field (small Mg-Fe log activity products) might be an indicator of deep secondary porosity. Incidently, these waters have an equilibrium pH of approximately 6 under hydrostatic conditions. It is not unreasonable to expect shallower waters in a subsiding, compacting basin like the Gulf Basin to bear some imprint of those at depth and presumably reservoir quality.

Two albitization reactions were considered (equations 3 and 4, table 2). Under hydropressured conditions all Brazoria waters plot well into the albite field at 100°C, implying that albitization is initiated in the hydropressured interval at less than 100°C (fig. 8). Under geopressured conditions the plagioclase stability field expands and suggests that plagioclase surviving albitization may be preserved at depth (fig. 3C). In reality, albitization becomes more difficult with depth. Figure 9 shows that sodium metasomatism of microcline clearly is possible in these sodium- rich waters. The obvious clustering of waters reflects a depth controlled increase in the $[K^+]/[Na^+]$ ratio (fig. 4) and in the Pleasant Bayou wells a loss of K-feldspar below 3,720 m. The shallowest waters have the smallest log ratio and lower silica activity.

Thermodynamically, K-feldspar is stable relative to albite, but less so at increased temperatures whereas albite is stable relative to plagioclase. This means that plagioclase will be albitized before K-feldspar as is the case in the Pleasant Bayou wells. Furthermore, albite and microcline cements will be favored in low-temperature hydropressured waters, with albite favored in waters similar to seawater ($\log [K^+]/[Na^+] \sim -1.75$) and microcline in fresher waters at even lower temperatures. Figure 9 shows that albite cement can form in the full range of waters tested from Brazoria County at less than 75°C, suggesting that albite cements formed before quartz overgrowths.

The relative stability of feldspar versus kaolinite is examined in equations 5 and 6. In shallower, hydropressured waters plagioclase is stable at temperatures of less than 100°C (fig. 10). On the other hand, kaolinite is stable relative to plagioclase under geopressured conditions. Thus, kaolinite probably begins forming as a grain replacement of plagioclase, eventually consuming the whole grain as diagenesis proceeds. Based on figure 11, the prediction is made that albitization began somewhat earlier than major precipitation of kaolinite. Under hydropressured conditions, that is, relatively early in the diagenetic history Brazoria waters plot in the albite field. Later, at increased pressure, the kaolinite field expands dramatically to encompass all waters at presumed lower in situ pH's. Furthermore, the reaction plagioclase = albite is thermodynamically favored over plagioclase = kaolinite. In short, albite is stable before kaolinite, thus the inference that albitization began earlier.

Finally, we speculate on the fate of the clay cutans, assuming such delicate features survive a long and complex diagenetic history. Prediction is that Ca-montmorillonite would persist in hydropressured waters at pH's greater than 6. In tested waters Na-montmorillonite and kaolinite are stable. In geothermal waters kaolinite would likely coexist with Na-montmorillonite.

CONCLUSIONS

1. Sandstones composed of chemically and mechanically stable grains are a prerequisite for preserving good reservoir quality in the Frio Formation.

2. The most important controls on reservoir quality are bulk composition, temperature, pH, pressure, and ionic activities.

3. Water data complement rock data adding new insight on paragenetic sequence, in situ pH, and relative mineral stabilities. Most diagenesis occurs in the hydropressured interval. Carbonate equilibrium requires early precipitation of calcite in diagenesis. Low temperature and pH explain the general absence of chlorite in Brazoria County.

4. Predictions based on equilibrium thermodynamics are consistent with petrographic data suggesting that estimated equilibrium constants (log K_r's) are correct to one or two orders of magnitude.

5. The use of solution-mineral equlibria as a predictor of reservoir quality is inconclusive and cannot be decisively tested until comparison between areas of good and poor reservoir quality is made. The well-defined clustering of log activity products and ratios demonstrated here indicates that comparisons between regions will probably be possible. The next phase of this work will evaluate the South Texas Frio of Kenedy County, an area noted for deep, low-permeability reservoirs. The key to predicting reservoir quality at depth may be the shallow waters.

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ACKNOWLEDGMENT

This research was funded by the U.S. Department of Energy, Division of Geothermal Energy, under contract No. DE-AC08-79ET27111.

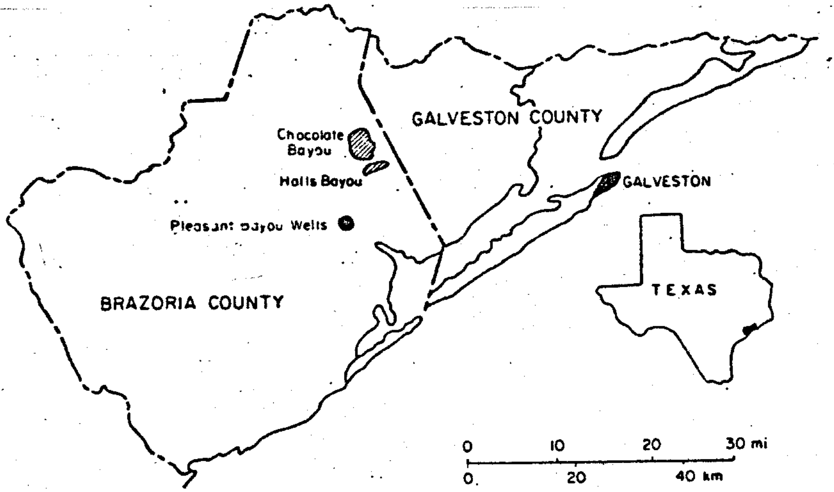

Fig. 1. Location of Chocolate Bayou Field, Halls Bayou Field, and Pleasant Bayou test wells.

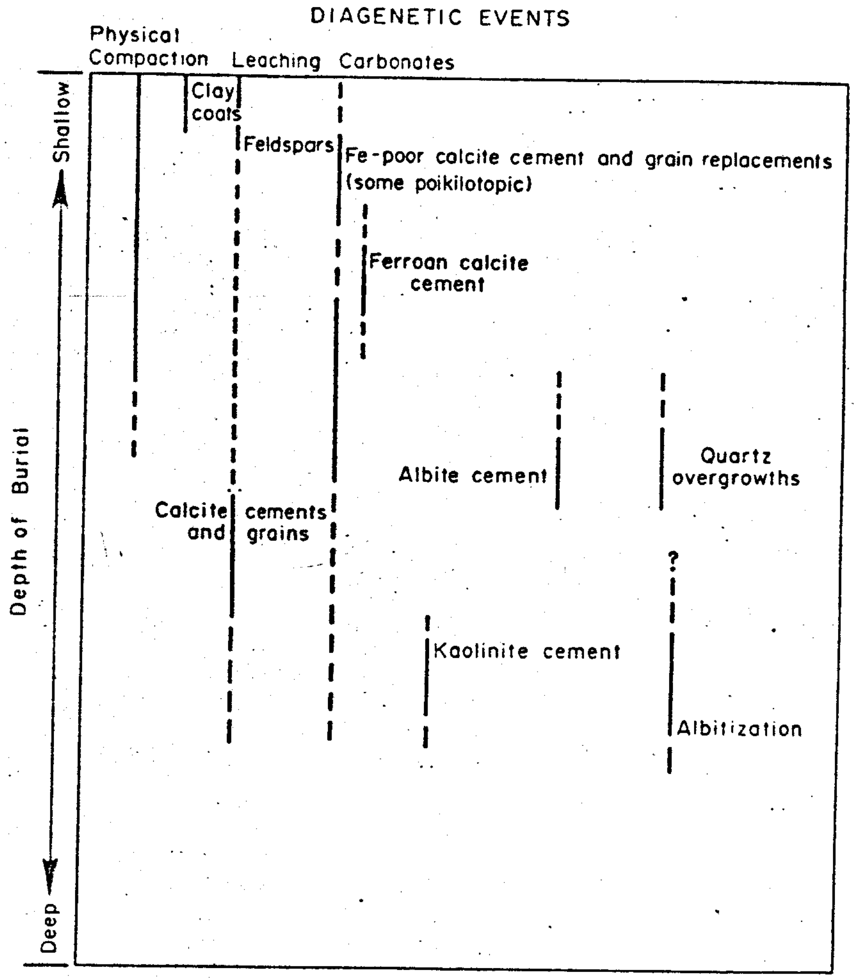

Fig. 2. Diagenetic history, Chocolate Bayou-Pleasant Bayou area. Modified from Loucks et al (1980).

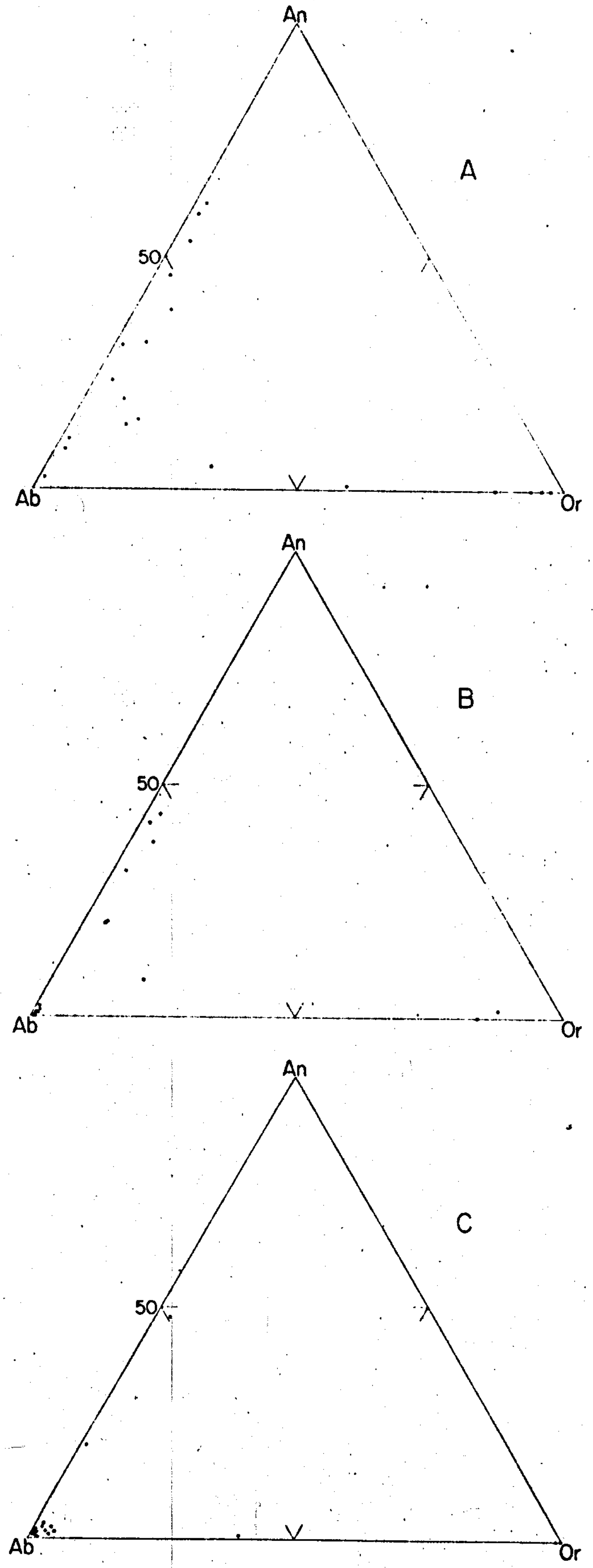

Fig. 3. Albitization of detrital feldspars in the Pleasant Bayou No. 1 and No. 2 wells. Albite-anorthite-orthoclase plots for 20 feldspars. A. Sample depth 3,130 m, well No. 1, average plagioclase (An 32). B. Depth 3,668-3,674 m, well No. 2. C. Depth 3,918-3,921 m, well No. 2.

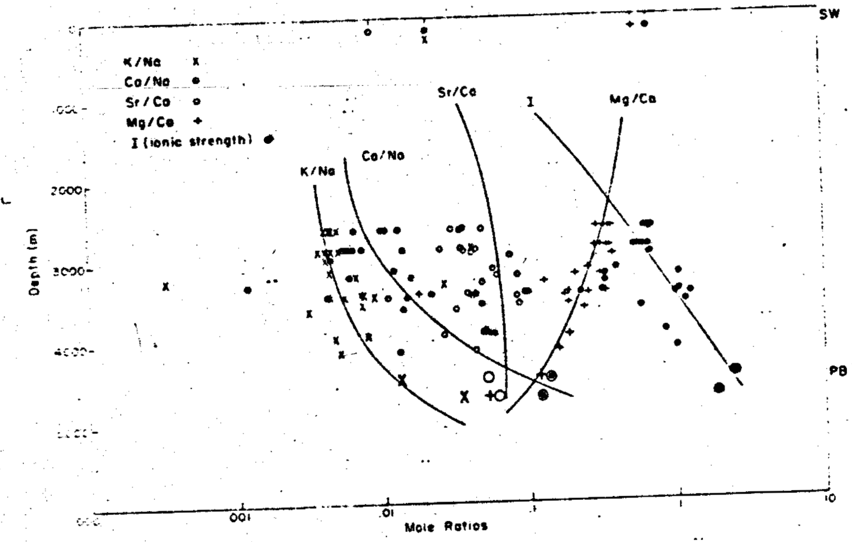

Fig. 4. Mole ratios of Brazoria County waters versus depth and seawater (SW). Pleasant Bayou (PB) waters at 4,462 m and 4,740 m shown in larger symbols.

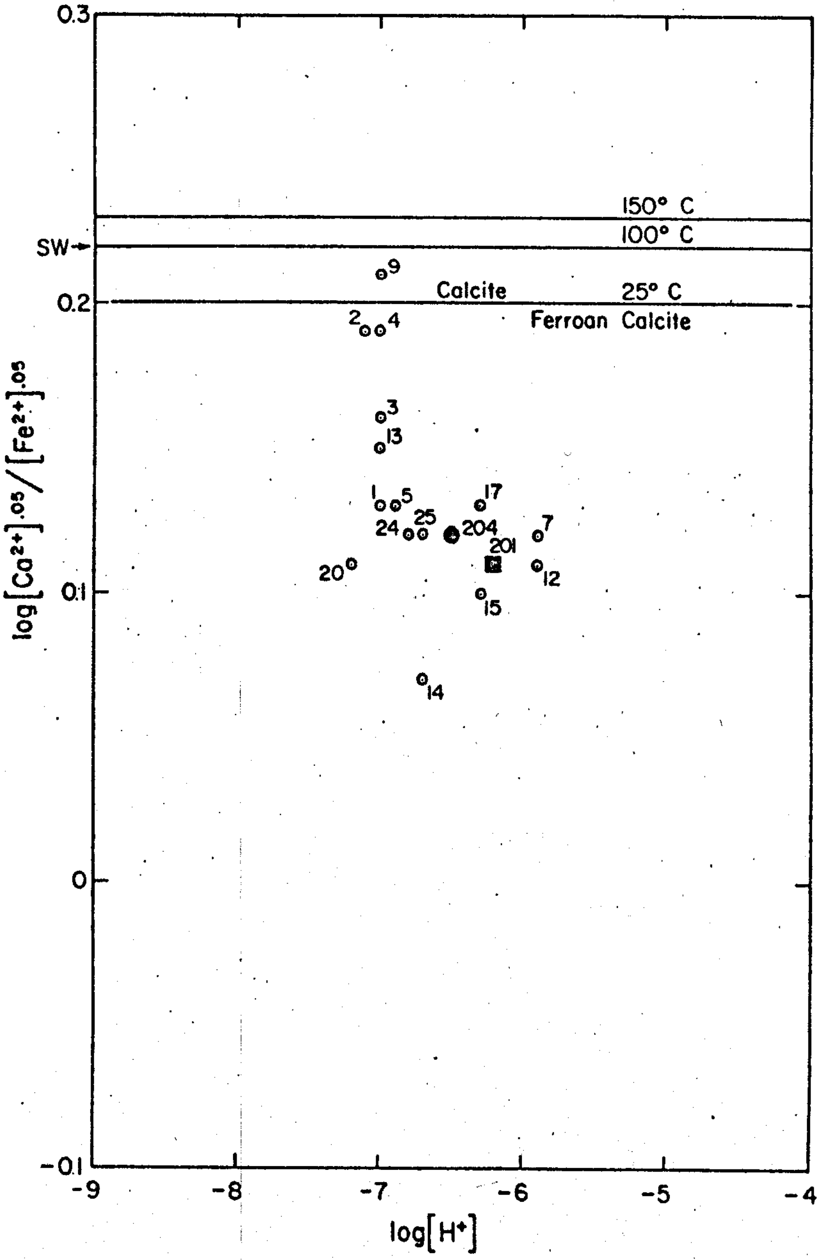

Fig. 5. Stability diagram for the reaction calcite = ferroan calcite (equation 1, table 2). Pleasant Bayou waters solid symbols, circle (4,462 m) and square (4,740 m). Numbers are the Kharaka et al (1977, 1979) sample number.

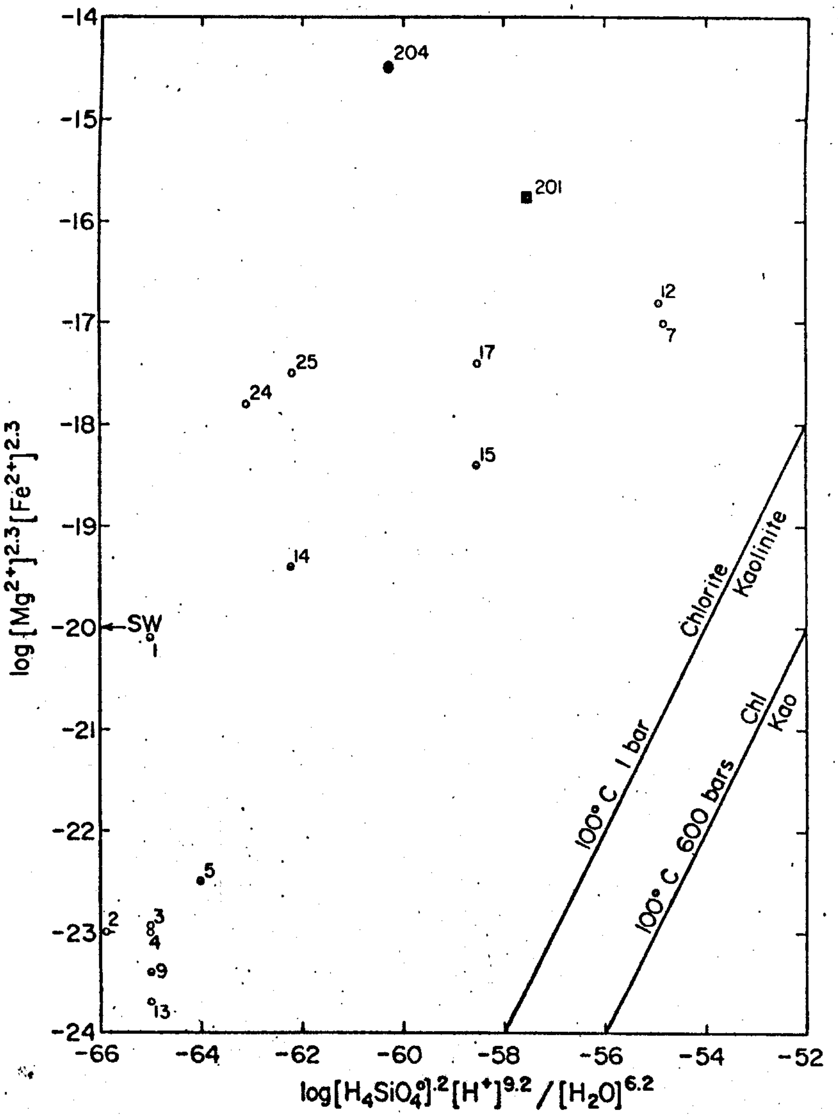

Fig. 6. Stability diagram for the reaction kaolinite = chlorite (eq. 2).

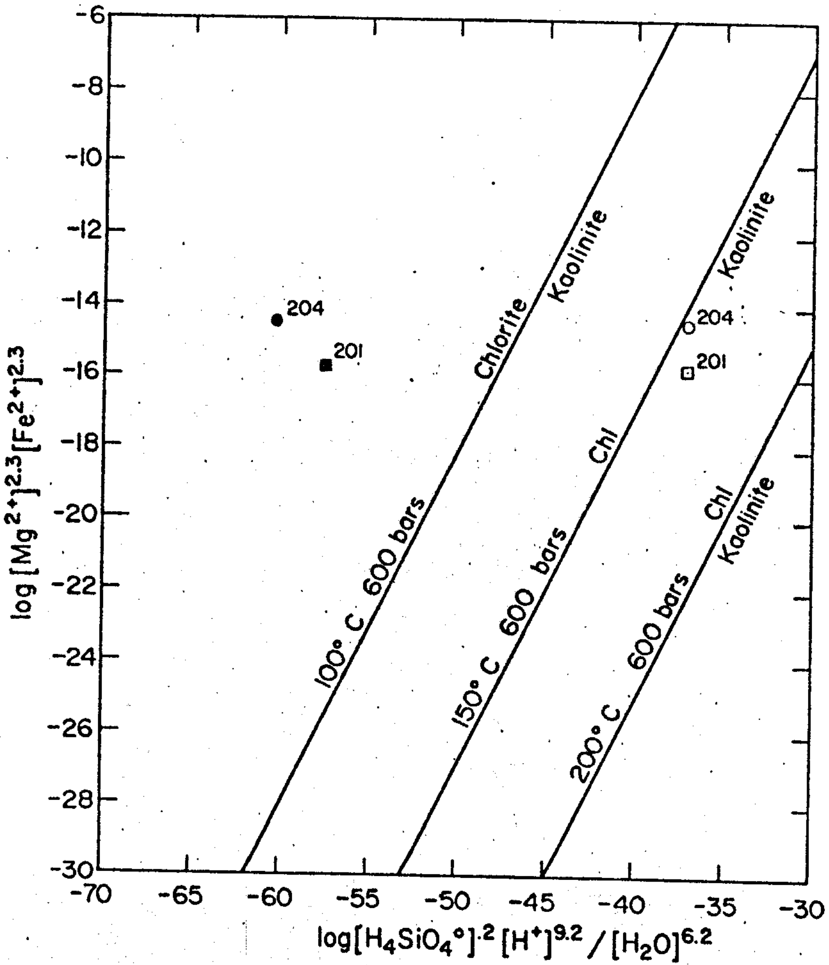

Fig. 7. Stability diagram for the reaction kaolinite = chlorite. Pleasant Bayou waters at wellhead pH (solid) and in situ pH (open).

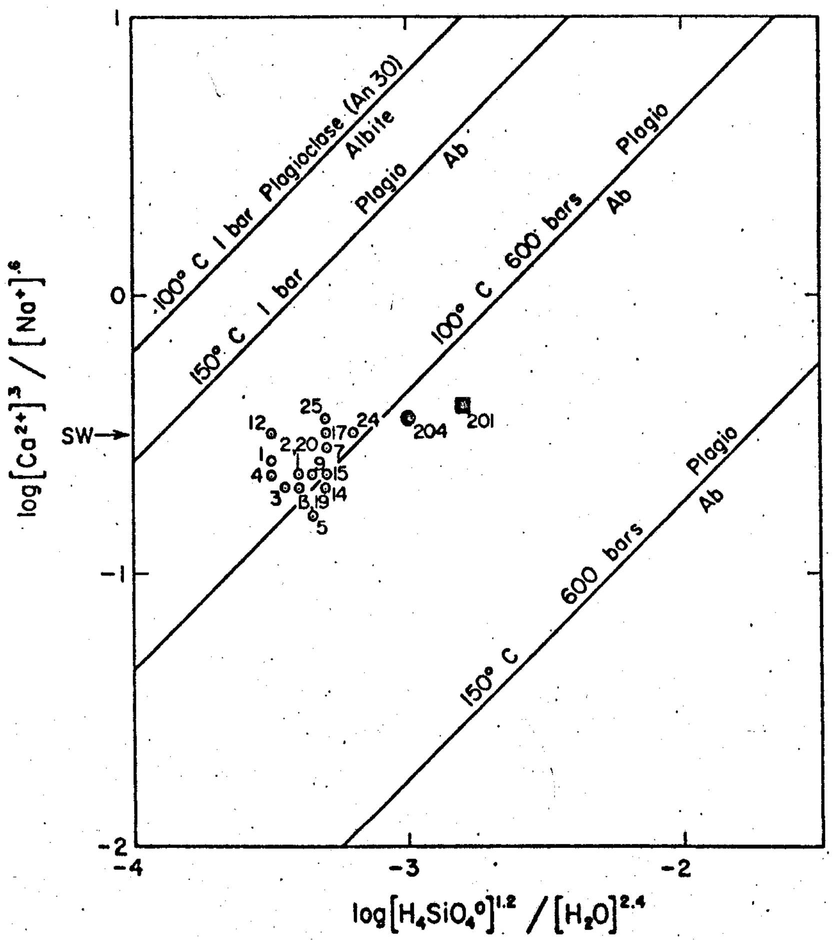

Fig. 8. Stability diagram for the reaction plagioclase (An 30) = albite (eq. 3).

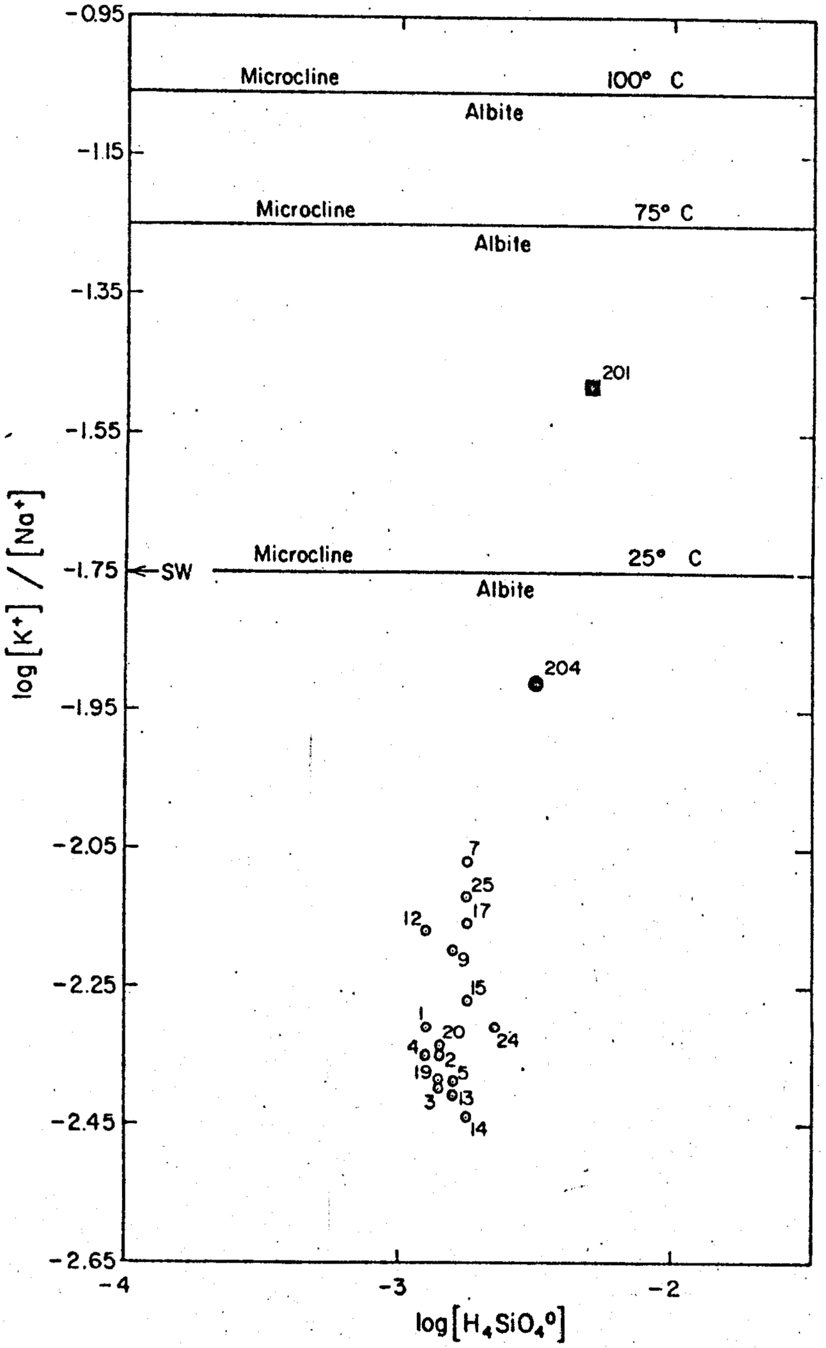

Fig. 9. Stability diagram for the reaction microcline = albite (eq. 4).

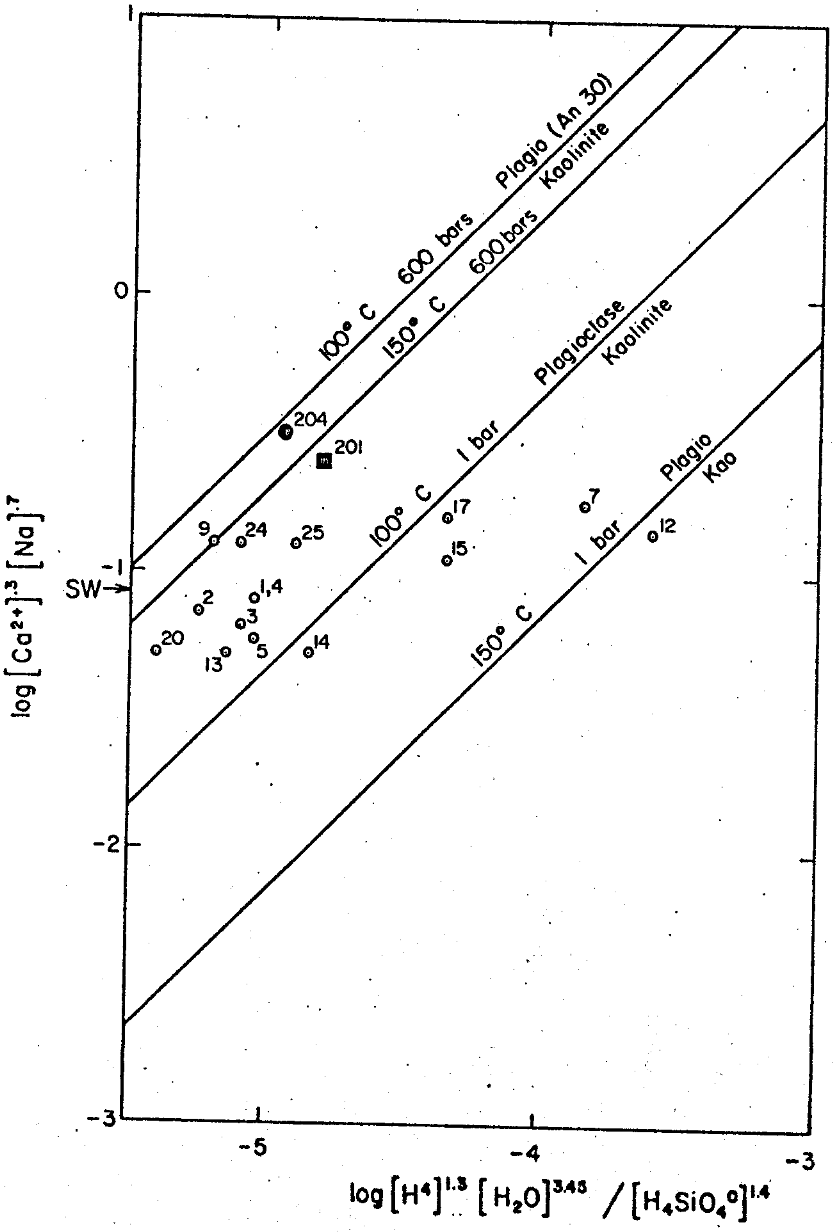

Fig. 10. Stability diagram for the reaction plagioclase = kaolinite (eq. 5).

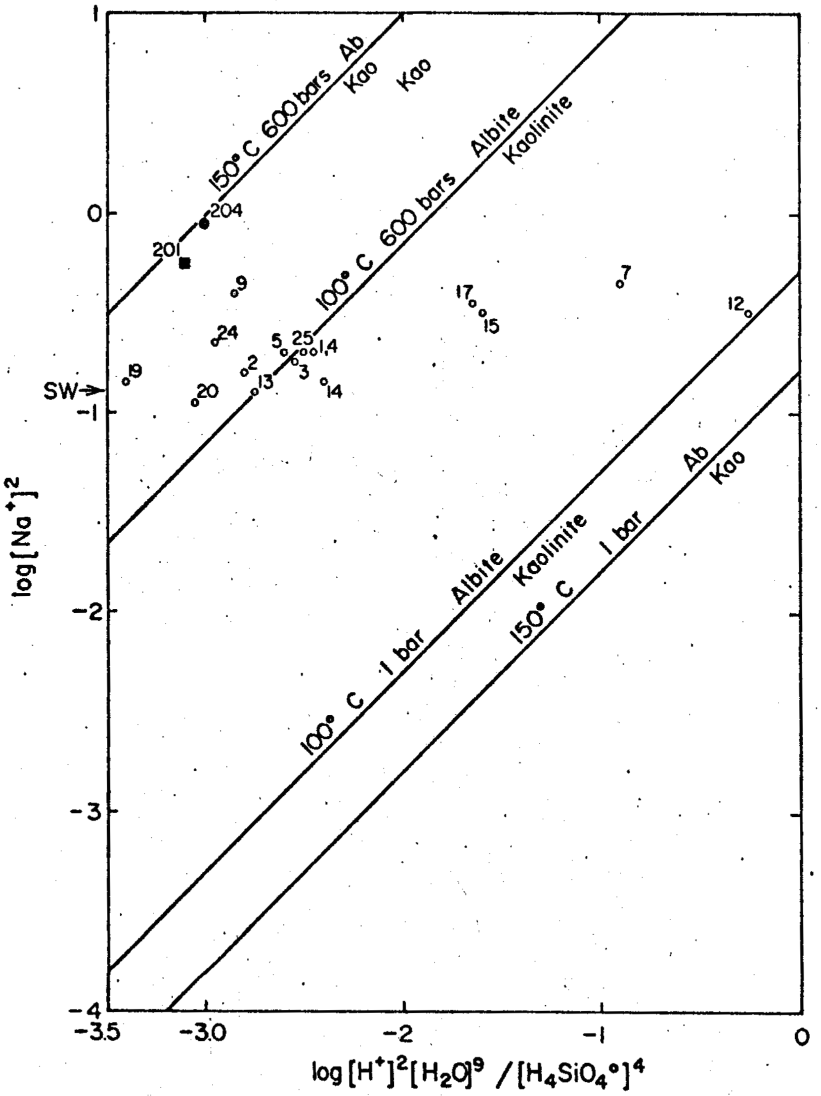

Fig. 11. Stability diagram for the reaction albite = kaolinite (eq. 6).

Table 1. Mineral composition.

Authigenic minerals		Detrital minerals	
Quartz	SiO_2	Plagioclase	$Na_{.7}Ca_{.3}Al_{1.3}Si_{2.7}O_8$
Calcite	$CaCO_3$	(An 30)	
Ferroan calcite	$Ca_{.95}Fe_{.05}CO_3$	Microcline	$KAlSi_3O_8$
Kaolinite	$Al_2Si_2O_5(OH)_4$		
Albite	$NaAlSi_3O_8$	Clay coats	
Chlorite	$Mg_{2.3}Fe_{2.3}Al_{2.8}Si_{2.6}O_{10}(OH)_8$	Calcium-montmorillonite	$Ca_{.16}(Al_{1.56}Mg_{.25}Fe_{.25})Si_4O_{10}(OH)_2$

Table 2. Reaction pairs[a].

1. Calcite + .05 Fe^{2+} = Ferroan calcite + .05 Ca^{2+}
2. 1.4 Kaolinite + 2.3 Mg^{2+} + 2.3 Fe^{2+} + 6.2 H_2O = Chlorite + .2 $H_4SiO_4^{\circ}$ + 9.2 H^+
3. Plagioclase + .6 Na^+ + 1.2 $H_4SiO_4^{\circ}$ = 1.3 Albite + .3 Ca^{2+} + 2.4 H_2O
4. Microcline + Na^+ = Albite + K^+
5. Plagioclase + 1.3 H^+ + 3.45 H_2O = .65 Kaolinite + .3 Ca^{2+} + .7 Na^+ + 1.4 $H_4SiO_4^{\circ}$
6. 2 Albite + 2 H^+ + 9 H_2O = Kaolinite + 2 Na^+ + 4 $H_4SiO_4^{\circ}$

[a] Reactions written with Al immobile.

FIGURE CAPTIONS

Fig. 1. Location of Chocolate Bayou Field, Halls Bayou Field, and Pleasant Bayou test wells.

Fig. 2. Diagenetic history, Chocolate Bayou-Pleasant Bayou area. Modified from Loucks et al (1980).

Fig. 3. Albitization of detrital feldspars in the Pleasant Bayou No. 1 and No. 2 wells. Albite-anorthite-orthoclase plots for 20 feldspars. A. Sample depth 3,130 m, well No. 1, average plagioclase (An 32). B. Depth 3,668-3,674 m, well No. 2. C. Depth 3,918-3,921 m, well No. 2.

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Fig. 5. Stability diagram for the reaction calcite = ferroan calcite (equation 1, table 2). Pleasant Bayou waters solid symbols, circle (4,462 m) and square (4,740 m). Numbers are the Kharaka et al (1977, 1979) sample number.

Fig. 6. Stability diagram for the reaction kaolinite = chlorite (eq. 2).

Fig. 7. Stability diagram for the reaction kaolinite = chlorite. Pleasant Bayou waters at wellhead pH (solid) and in situ pH (open).

Fig. 8. Stability diagram for the reaction plagioclase (An 30) = albite (eq. 3).

Fig. 9. Stability diagram for the reaction microcline = albite (eq. 4).

Fig. 10. Stability diagram for the reaction plagioclase = kaolinite (eq. 5).

Fig. 11. Stability diagram for the reaction albite = kaolinite (eq. 6).

LEGIBILITY NOTICE

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