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NEW THREE PHASE EQUILIBRIUM MODEL (OIL-GAS-BRINE) USED TO INTERPRET PRODUCTION OF LIQUID HYDROCARBONS FROM A GEOPRESSURED GAS WELL

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New three phase equilibrium model (oil-gas-brine) used to interpret production of liquid hydrocarbons from a geopressured gas well

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ABSTRACT

Several of the design geopressured gas wells developed by the U.S. Department of Energy have produced small amounts of liquid hydrocarbons. At all wells, an unusual, aromatic gas condensate has been collected. This condensate differs dramatically from oil, containing predominantly light aromatic hydrocarbons, with subordinate cycloalkanes, branched alkanes, and normal alkanes. Two of the wells have also produced a paraffinic oil. We have analyzed hydrocarbon liquids produced from the Gladys McCall No. 1 well (Cameron Parish, Louisiana).

We have developed a computer program that models detailed phase relations in the system gas-oil-brine, and have used it to interpret the production of hydrocarbon liquids from Gladys McCall No. 1. We conclude that a single dispersed hydrocarbon phase was present within the producing formation, initially at some distance from the wellbore. The aromatic condensate represents the relatively water soluble hydrocarbons which dissolved in the brine. Prolonged production of brine from the well caused the hydrocarbon phase to move toward the well, ultimately leading to production of oil. Adsorption of of less volatile hydrocarbons on minerals and organic matter retarded their transport in the formation, producing some chromatographic separation of the hydrocarbons in the oil.

INTRODUCTION

The U.S. Department of Energy has developed several geopressured gas wells in the Gulf Coast area. These wells are all deep (4-5 km) and abnormally pressured, with initial producing zone pressure approaching lithostatic pressure at that depth. The fluid produced is a hot brine (90°C or greater) which contains dissolved natural gas, 3-6 L gas (STP)/ L brine. While these brines were initially considered a geothermal resource, the gas is now considered to be the major product.

The gas produced from most of these wells contains a small amount of predominantly aromatic liquid hydrocarbons which may be condensed from the gas in a dry ice - acetone trap. A comparable amount of aromatic hydrocarbons remain dissolved in the brine after the gas is separated from it. Two of these wells have also produced a paraffinic oil. The production of aromatic condensate and oil from the L.R. Sweezy No. 1 well has been described by Hamilton and Stanley (1), and Weres *et al.* (2). The production of liquid hydrocarbons from Gladys McCall No. 1 has been described by Keeley and Meriwether (3). Osif (4) has summarized operating data from Gladys McCall No. 1 and several other geopressured wells, and concluded that there is no free gas present in the producing formations of these wells.

The quantity of liquid hydrocarbons produced from these wells is economically insignificant. The production rate of aromatic condensate from Gladys McCall was 17μL/L brine when first measured in October 1983, rising to 31μL/L in June and 38μL/L in December 1984 (3). When oil was produced from this well January to June, 1985, the production rate averaged about 30μL/L. However, production of hydrocarbon liquids was unexpected, as the temperature of the producing formation, 145°C, suggests that only dry gas should be present. The production of these liquids offers at least the distant hope of identifying hitherto unsuspected deep oil deposits, and may shed light on the processes that produce and accumulate hydrocarbons in nature.

ANALYSIS OF HYDROCARBON LIQUIDS

A number of samples of hydrocarbon liquids produced from Gladys McCall No.1 were provided to us by Dr. D. F. Keeley of the University of Southwestern Louisiana. These samples fall into two distinct classes:

1. A paraffic oil produced January to June 1985. Several samples from February and March were analyzed. These samples are all very similar, and consist mostly of normal and branched alkanes. This oil looks like the higher boiling fraction of a high gravity natural oil (Figure 1). This oil is miscible with pentane in all proportions, suggesting that asphaltenes are absent.
2. Aromatic condensate samples which were condensed from the gas using a dry ice/ acetone trap. Sixteen samples collected monthly (with some gaps) are available for the period October 1983 - July 1985. These liquids consist predominantly of aromatic hydrocarbons, mostly benzene, toluene, and xylenes (Figure 2). Alkanes and cycloalkanes are also present in subordinate amounts.

References and illustrations at end of paper

Aromatic hydrocarbons predominate in all aromatic condensate samples, and the aromatic fraction of this liquid does not change noticeably with time (Fig. 2, Fig. 3). The smaller alkanes and cycloalkanes (2-methylhexane, methylcyclopentane, cyclohexane, methyl cyclohexane) are also fairly water soluble. These compounds are present in all samples of the aromatic condensate, and behave like the aromatic compounds. The middle range alkanes (C-7 to C-12) behave very differently. Only traces of these compounds are present in samples taken through November 26, 1984 (Fig. 2). They are present in abruptly larger concentration in the samples taken December 28, 1984 to April 29, 1985 (Fig. 3). The concentration of alkanes is again lower in samples taken June 5 and July 17, 1985. The concentration of n-alkanes C-7 to C-12 versus time is depicted in Figure 4.

The change in the alkane fraction of the aromatic condensate is clearly related to the production history of oil. The first oil was noted and sampled January 19, 1985. Steady oil production began in early February, and continued until some time in June. Obviously, the alkanes present in the aromatic condensate from January 30 to April 29 represent the more volatile fraction of the oil that was being produced at that time. However, the alkanes first appeared in the condensate sample of December 28, 23 days before the production of oil was noted. In fact, the alkanes probably appeared in the condensate sometime earlier in December, but no mid-month sample is available. Likewise, production of oil probably commenced sometime prior to January 19, but was small until the first days of February. In summary, it appears that the alkanes in the aromatic condensate increased about three weeks before the production of oil began.

Figure 5 contains the data from Figure 4 in a different representation. Here the concentration of each compound is divided by the concentration in the sample of February 19 which contained the largest total amount of alkanes. During the period November - February the concentration of the lighter alkanes increased more rapidly than the heavier alkanes. This trend is very regular and all six compounds obey it. During the period February - July, the concentration of C-8, C-9 and C-10 decreased more rapidly than the concentration of C-11 and C-12. Heptane disobeys this rule, probably because it is much more water soluble than the others. These patterns suggest chromatographic separation within the reservoir, involving the partitioning of these compounds between two phases, perhaps oil and gas. That the alkanes probably appeared in the condensate before the oil was produced is another expression of the same phenomenon - the lighter fraction of the oil appeared in the condensate before the heavier fraction appeared in the separator.

We performed a Soxhlet extraction on core material taken from the producing horizon of the well. Only contaminants obviously derived from the wax that had been used to seal the core were detected in the extract - plasticizers and some kerosene.

COMPUTER PROGRAM "RELAX"

Because the production of hydrocarbon liquids from geopressured wells certainly involves the complex phase relations in the system oil - gas - brine at high pressure, we decided to write a computer program which allows us to numerically model these phase relations.

This program, which is called RELAX, calculates phase equilibria involving up to four fluid phases for a single point in space. It is not a full reservoir simulator; rather, it is equivalent to that portion of a reservoir simulator which calculates phase equilibrium within a single element of the grid. RELAX is quite fast and requires little memory. In principal, it could be built into a reservoir simulator. RELAX is among the few programs able to model detailed phase equilibria involving brine as well as oil and gas.

The ability to model three phases is indispensible in the geopressured context, where most of the hydrocarbon gases are initially dissolved in the brine, some oil is present in the formation, and free gas may be released during production. While it is unnecessary in this application, RELAX is also able to model a second liquid hydrocarbon phase, which may be encountered in tertiary recovery operations utilizing miscible displacement with CO_2.

The brine phase is modelled as a solution of sodium chloride in water, in which the various gases and relatively soluble hydrocarbons may dissolve. The solubility of methane in brine is calculated using the empirical formulas presented by Price et al. (5). The solubility of other hydrocarbons is represented in terms of Henry's Law Coefficients which are included in the data base as functions of temperature. Most other solubility data were taken from the API Petroleum Refining Technical Data Book (6). These values are approximately corrected for the effects of salinity and pressure as needed. The properties of the gas and oil phases are calculated using the Soave equation with interaction coefficients estimated following Graboski and Daubert (7,8). The algorithm of RELAX is described in the Appendix.

COMPUTER MODELLING WORK

The computer modelling work is still underway at the time of writing, and the computed results will only be summarized here.

Most production from Gladys McCall No. 1 has been from Sand No. 8, located at 4,620 to 4,721 meters BSL. The initial pressure at 4,602 meters was 881 bar, and the initial temperature was 145°C (4). Following extensive brine production, the downhole pressure dropped to about 800 bar. Total gas/water ratio (including gas that remains dissolved in the brine) is about 5.4 L (STP)/ L brine. The gas consists mostly of methane (79v%) and carbon dioxide (18v%) with small amounts of other hydrocarbons and nitrogen. With no liquid hydrocarbons present, the bubble point estimated using RELAX is about 500 bar. Clearly, no free gas phase in the conventional sense can be present in the formation. Osif (4) came to the same conclusion.

Further calculations quickly demonstrated that only one hydrocarbon phase is present in the formation; there are no distinct oil and gas phases. We estimated the composition of the hydrocarbon phase in the formation from the composition of the aromatic condensate and oil produced from the well. If the brine extracted from the formation initially was in chemical equilibrium with the hydrocarbon phase, the concentration of, say, benzene present in the separated gas will be proportional to the mole fraction of benzene in the hydrocarbon phase. Knowing the temperature and pressure of the formation and the separator, the salinity of the brine, and the gas/brine ratio, we are able to calculate the ratio benzene in the gas/ mole fraction benzene in the hydrocarbon phase using RELAX. From the production rate and analyzed composition of the aromatic condensate we are then able to estimate the mole fraction in the hydrocarbon phase of any compound that is determined in the aromatic condensate. The mole fraction of each gas in the hydrocarbon phase is likewise estimated from the composition of the produced gas. These calculations gives us most hydrocarbons C-1 to C-10. The produced oil probably is the same as the C-11+ fraction of the hydrocarbon phase.

The total mole fraction of gases in the hydrocarbon phase is large - about 75 mole % - and its specific gravity is low - about 0.48. The large gas content is consistent with the large concentration of gas dissolved in the brine downhole, and consequently high fugacity of methane and other gases in the system. The C-5 to C-10 fraction includes 40 mole % aromatics, but only 13 mole % cycloalkanes. Apparently, cycloalkanes have been largely converted to aromatics by the high temperature in the formation.

Otherwise identical calculations were performed at a series of pressures. Separation of the hydrocarbon phase into distinct gas and oil phases did not occur until pressure was reduced to

below 400 bar. Increasing the proportion of nonvolatile hydrocarbons in the hydrocarbon phase did not change this result. While the exact pressure for phase separation will depend on the exact composition and amount of hydrocarbon phase present, it is clear that distinct oil and gas phases cannot coexist in the producing formation. Rather, any liquid hydrocarbons present will pull some amount of gas out of the brine, to form a single gas-rich hydrocarbon phase. The volume and gas fraction of this phase increase with decreasing pressure, until a distinct oil phase separates below 400 bar. In place of a well defined bubble point, we have a gradually expanding gas-rich phase which eventually gives rise to distinct oil and gas phases.

DISCUSSION

Zarrella et al. (9) have analyzed brines from several oil wells for benzene, and concluded that benzene in the brine indicates the presence of oil in the same horizon. The concentration of benzene decreases with increasing distance from the oil deposit. There is no doubt that a gas-rich hydrocarbon phase is present in or near Sand No. 8 of Gladys-McCall No. 1, and that the aromatic condensate is derived from relatively water soluble compounds in the hydrocarbon phase which dissolved in the brine. That the production of aromatic condensate increased during the first months of brine production indicates that this hydrocarbon phase was initially some distance from the well, but gradually migrated toward the well as production continued until oil production finally commenced. The increase in aromatic condensate production preceded oil production by several months, indicating that brine moves more rapidly than the hydrocarbon phase, by a factor of 2 or 3. The absence of extractable hydrocarbons in the core material from the well is consistent with this interpretation.

While the conventional view is that formations above 100°C should not contain liquid hydrocarbons, all of the design geopressured gas wells have produced aromatic condensate, and two have produced oil. Furthermore, Price et al. (10,11,12) have extracted liquid hydrocarbons from core and cuttings obtained from wells are deeper and hotter yet, with downhole temperature as high as 300°C.

Because well head pressure is approximately 270 bar, the separation of oil and gas must occur within the wellbore, near the top of the well. Phase partitioning involving coexisting oil and gas phases within the producing formation cannot explain the chromatographic separation of compounds that is evident in Figure 5, nor that production of oil was first noted three weeks after its more volatile components first appeared in the the aromatic condensate. While distinct oil and gas phases are present in the upper portion of the wellbore between 400 and 270 bar pressure, it is unlikely that the alkanes were separated there. The oil at the top of the wellbore contains a large amount of gas, and is well above its pour point. The viscosity of this oil probably is small. Given the high shear and high turbulence of the fluid flow near the top of the wellbore, it is unlikely that a stationary oil phase could accumulate and persist there, allowing chromatographic separation of compounds to occur. Nor can the data in Figure 5 be explained in terms of processes occurring within the separator.

We believe that the chromatographic separation in Figure 5 and the delayed onset of oil production are due to some other partitioning process that occurs within the producing formation. Most probably, this process involves partial adsorption of the less volatile compounds present in the hydrocarbon phase onto mineral surfaces or clay, or onto organic material in the rock, such as kerogen or bitumen. Bitumen would not be soluble in the methane-rich hydrocarbon phase, and would remain in the formation. This conclusion is consistent with the absence of asphaltenes from the produced oil. Adsorption onto clay or kerogen in turn suggests that the hydrocarbon phase is initially dispersed. If the hydrocarbon phase flowed toward the wellbore as a compact, pore filling phase, the amount of oil present would overwhelm the adsorption capacity of the rock, and separation effects would be small.

We will review the whole story. A gas-rich hydrocarbon phase is initially dispersed within the formation at a moderate distance from the wellbore. Relatively water soluble hydrocarbons are partitioned between the hydrocarbon phase and the brine, with the concentration of aromatics in the brine decreasing with distance from the hydrocarbon phase. Initially brine containing dissolved gas and a small amount of aromatic hydrocarbons is produced. Over a period of months, more brine initially in contact with the hydrocarbon phase is produced, and the production of aromatic hydrocarbons increases. The brine flowing toward the wellbore also pulls some of the hydrocarbon phase with it, migrating at perhaps one-half the velocity of the brine. Adsorption onto minerals and organic materials in the rock retards the less volatile compounds in the hydrocarbon phase, but ultimately does not block their transport. After about a year the hydrocarbon phase reaches the wellbore, and production of oil commences. After a few more months, the hydrocarbon phase is depleted within the brine flushed volume, and production of oil ceases.

Apparently, we have encountered a situation where liquid hydrocarbons are present, but have not migrated to form actual petroleum deposits. Most of the gas remains dissolved in the brine, while the liquid hydrocarbons are dispersed throughout the pore space of a large volume of rock. Brine production dislodges the dispersed hydrocarbon phase, and initiates migration, ultimately causing the production of oil.

These data and our interpretation lead to several practical conclusions:

1. The suggestion made by Zarrella et al. (9) that aromatic hydrocarbons dissolved in the brine indicate an oil-rich hydrocarbon phase is present nearby has been supported. The concentration of aromatic hydrocarbons in the brine will depend on the proximity of the hydrocarbon phase, the temperature, and the composition of hydrocarbon phase.
2. Increasing production of aromatic condensate suggests that the hydrocarbon phase is migrating toward the wellbore.
3. The appearance of substantial amounts of alkanes C-7 and above in the aromatic condensate indicates that production of oil is imminent.
4. Small amounts of a dispersed hydrocarbon phase may be mobile in a geopressured reservoir; that is, conventional ideas regarding minimum phase saturation required for a phase to become mobile may not apply in this case.

That the D.O.E geopressured design wells have produced economically insignificant amounts of oil reflects the small amount of oil present, not the production characteristics of these reservoirs. If more oil were present in the formation, more oil would be produced. The possibility remains that other geopressured formations may yield commercial quantities of oil.

NOMENCLATURE

T_c = critical temperature of a particular compound

ACKNOWLEDGEMENTS

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We wish to thank Dr. D.F. Keeley for providing us with samples of aromatic condensate and oil collected from Gladys-McCall No. 1, and for providing us with a copy of Ref. 3 before it was published.

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APPENDIX - THE ALGORITHM OF RELAX

The input to RELAX includes temperature, pressure, and the number of moles of each chemical component present. The components used by RELAX are actual chemical compounds, not pseudocomponents. About one hundred components are included in the data base. They include sodium chloride, water, carbon dioxide, hydrogen sulfide, nitrogen, and a large number of hydrocarbon liquids and gases. Only the components water, carbon dioxide, hydrogen sulfide, nitrogen, and methane are included in all calculations. Other components are included only if specified in the input. RELAX could easily be converted to pseudocomponents simply by adding needed pseudocomponents to the data base, and specifying the input in terms of pseusocomponents. The hydrocarbons included in the data base were selected based on the composition of the "representative petroleum" analyzed under the auspices of API Project 6 (13).

At the start of the calculation, the various components are distributed among the various phases. All water and *NaCl* are assigned to the brine phase. Other components are assigned to the liquid or gas phases, according to critical temperature. Components with $T_c > T$ are assigned to the oil phase, while components with $T_c < T$ are assigned to the gas phase. If CO_2 is present, and if the temperature is less than the critical temperature of CO_2, all of the CO_2 is assigned to the second liquid hydrocarbon phase; otherwise, no second hydrocarbon phase is "created", and CO_2 is assigned to the gas phase.

This initial distribution will usually be far from equilibrium; the calculation that follows takes it to equilibrium. Essentially, we combine gas, oil and brine in a separatory funnel, and shake the funnel until phase equilibrium is obtained. The fugacity of each component in each phase is calculated using the Soave Equation or Henry's Law, as appropriate. In a real system each component will migrate from the phase where fugacity is higher to the phase where fugacity is lower. Therefore, the direction of migration between phases is immediately known, but the extent of migration is not. A separate calculation estimates the extent of migration. The system is temporarily decomposed into pairs of phases; for example, the system brine - gas - oil is decomposed into the three subsystems brine & gas, brine & oil, and gas & oil. For each subsystem distribution coefficients (K-values) relating the two phases are calculated, and the equilibrium distribution involving just those two phases is calculated with distribution coefficients held constant. This calculation involves solving a single equation using the Newton - Raphson algorithm, and is very fast and stable. In going from the initial state to the two phase equilibrium state, a certain amount of each component is transferred from one phase to the other. These mass transfer vectors are determined for each pair of phases.

The net migration of components among the three phases is taken to be a superposition of the three mass transfer vectors calculated for the three subsystems. When three or more phases are present the mass transfer vectors are multiplied by the "step size" - usually 0.5 or 0.8 - to prevent overshooting the desired solution. New estimates for the composition of each phase are obtained in this way, and new values of mole fraction and fugacity are calculated for each phase. This cycle is repeated until the fugacity of each component is the same in each phase.

One or more phases may disappear in the course of the calculation; for example, the gas phase may dissolve completely in the brine. The program will recognize a very small and steadily shrinking phase as one that is disappearing, and will remove it from the calculation. In other cases two phase may become identical; for example, gas and oil above the critical point of the mixture. The program will recognize this situation as well, and combine the two degenerate phases.

This algorithm is fast and stable. No large matrices are used. Because the basic concept of the algorithm is quite simple, the program is easy to work with and debug. The program is highly structured and easy to modify. It would be a very simple matter to, say, substitute the Peng-Robinson Equation for the Soave Equation.

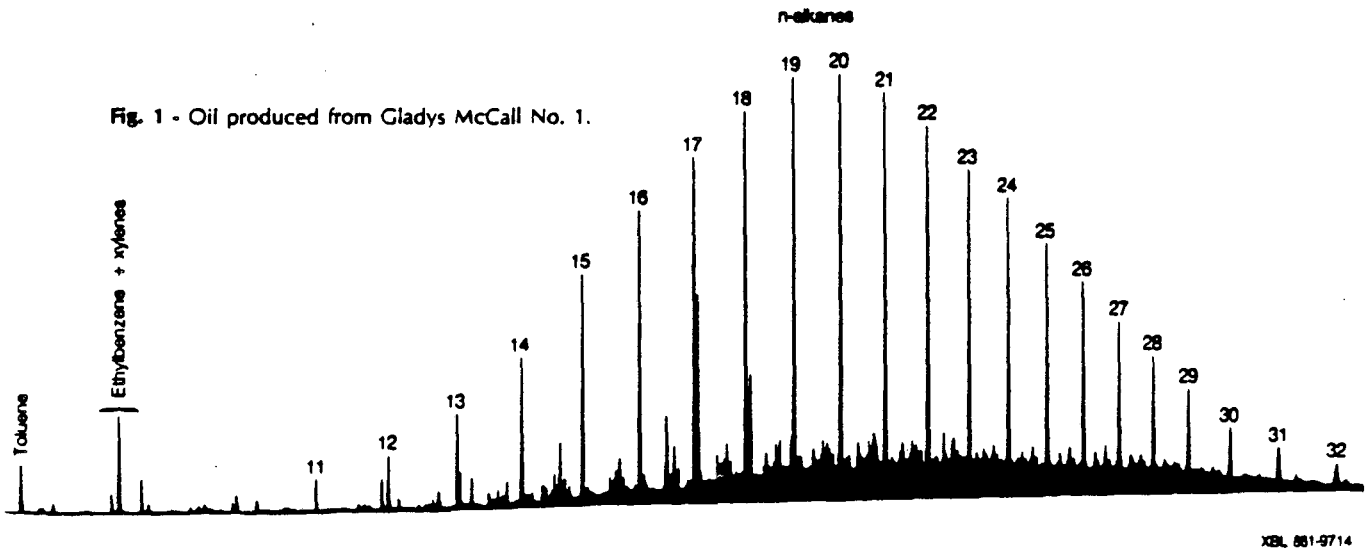

Fig. 1 - Oil produced from Gladys McCall No. 1.

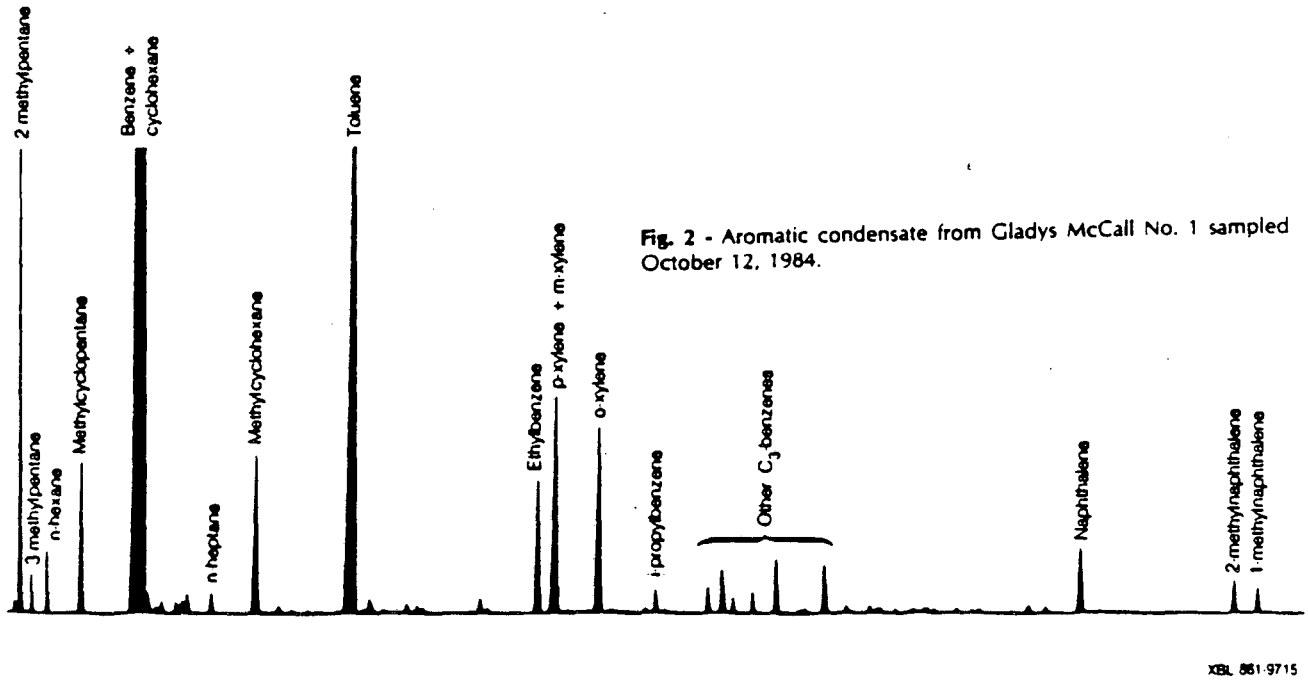

Fig. 2 - Aromatic condensate from Gladys McCall No. 1 sampled October 12, 1984.

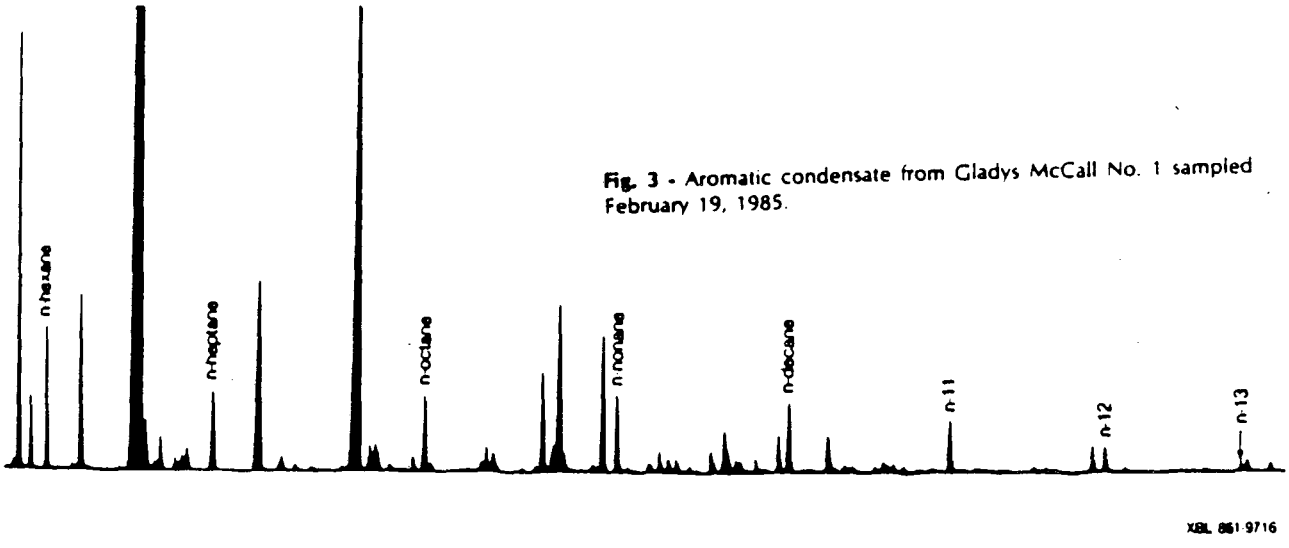

Fig. 3 - Aromatic condensate from Gladys McCall No. 1 sampled February 19, 1985.

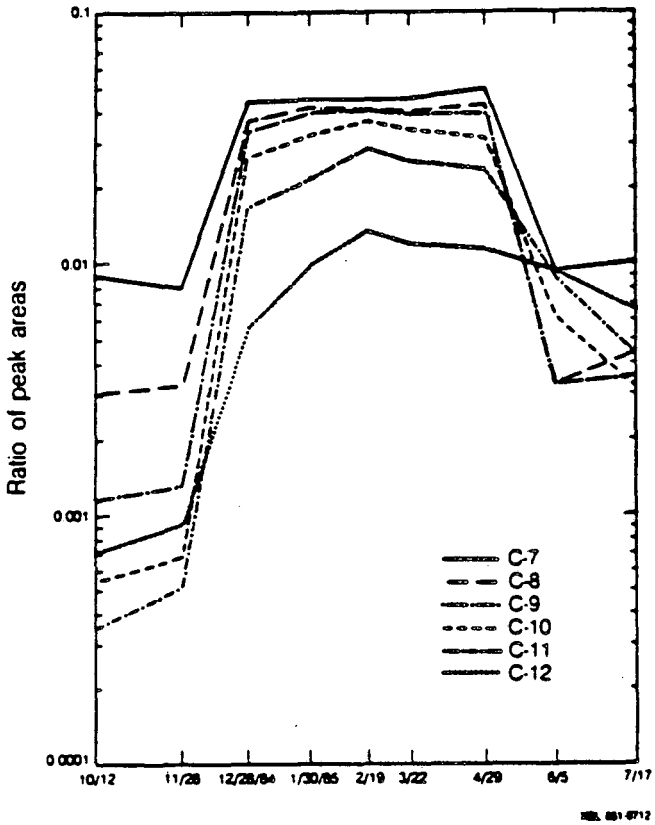

Fig. 4 - Concentration of n-alkanes C-7 to C-12 in aromatic condensate from Gladys McCall No. 1, October 1984 - July 1985. Each point represents the peak area of the given compound in the given sample, divided by the sum of peak areas in that sample of toluene, all C-2 benzenes, all C-3 benzenes, naphthalene, and both methylnaphthalenes.

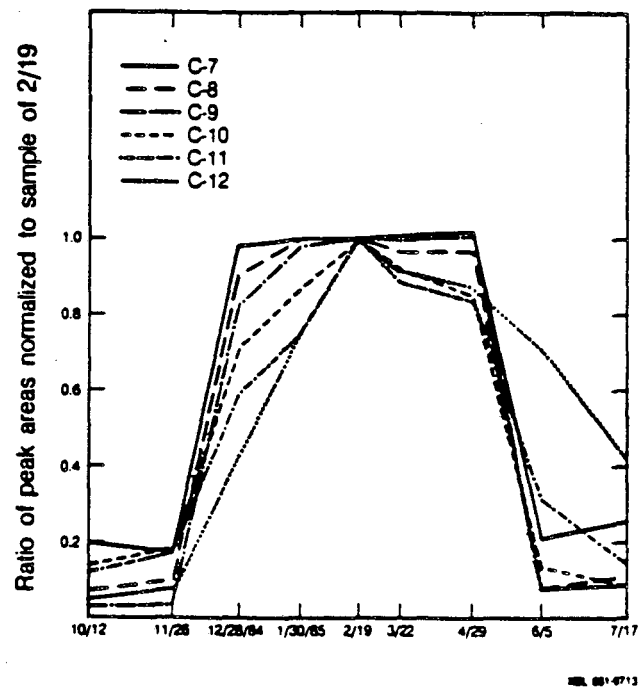

Fig. 5 - Concentration of n-alkanes C-7 to C-12 in aromatic condensate from Gladys McCall No. 1, October 1984 - July 1985. These data are replotted from Fig. 4. Each concentration value was divided by the concentration of the given compound in the sample collected February 19, which contained the largest total concentration of alkanes.

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