Dec. 20, 1966 G. A. HILL ETAL 3,292,693

METHOD OF STORING TOXIC FLUIDS AND THE LIKE

Filed June 18, 1962 2 Sheets-Sheet 1

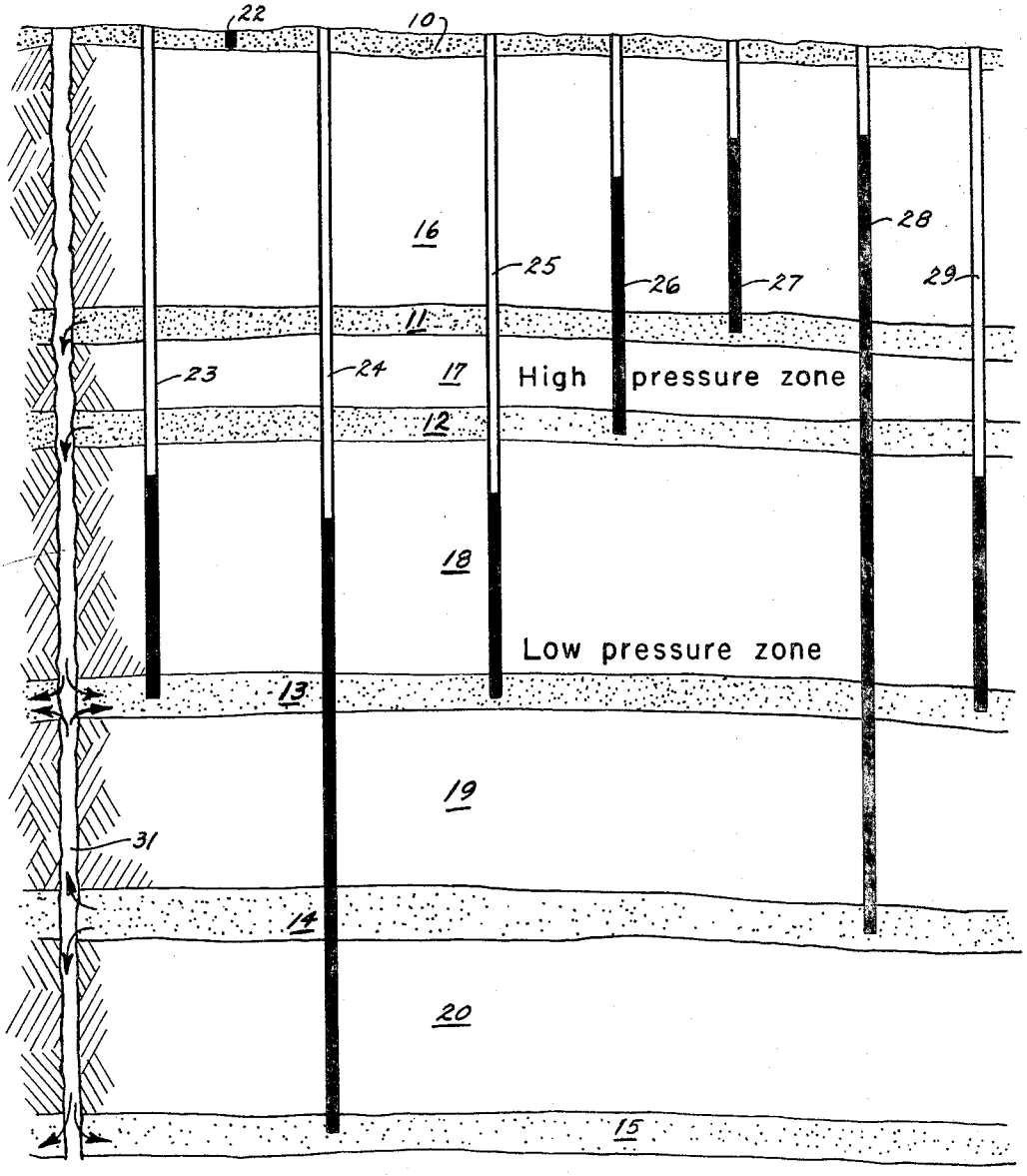


Fig 1

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METHOD OF STORING TOXIC FLUIDS AND THE LIKE

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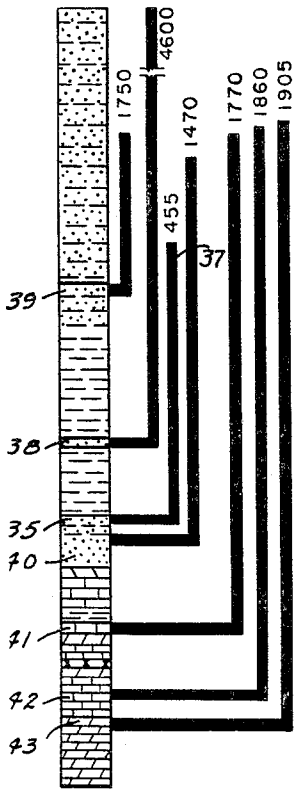


FIG 3

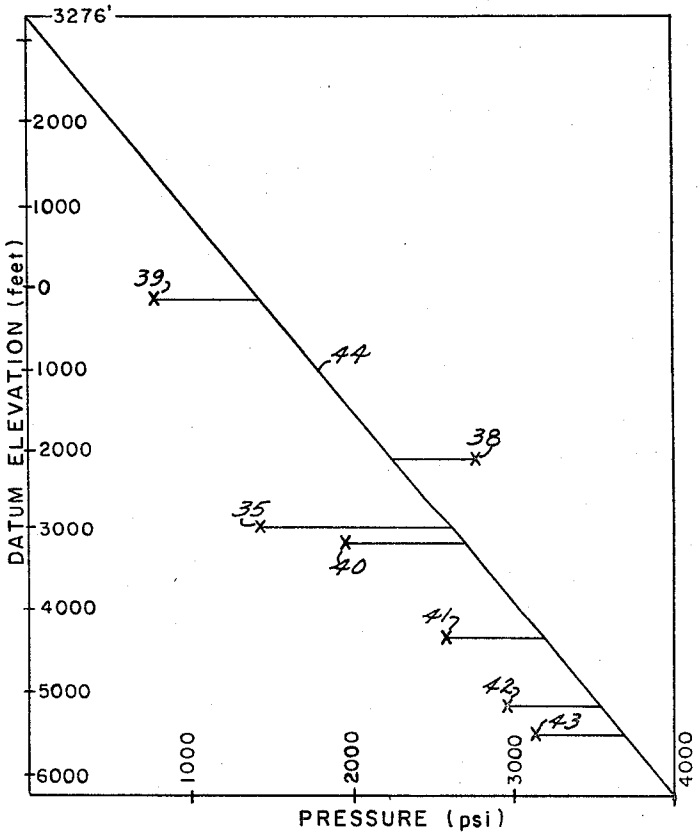


FIG 4

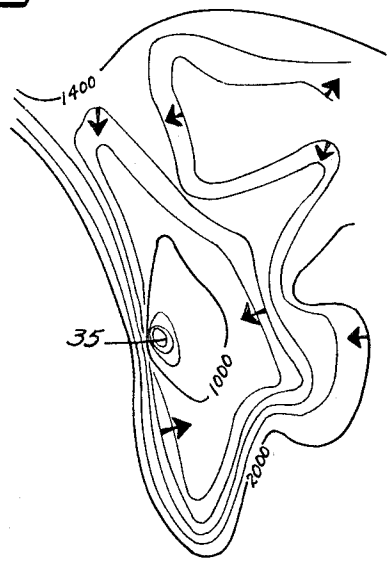


FIG 2

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3,292,693

METHOD OF STORING TOXIC FLUIDS AND THE LIKE

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20 Claims. (Cl. 166—2)

This invention relates to the storage of fluids and particularly to an improved method for effecting safe and permanent storage or disposal of toxic waste products and the like.

One of the most pressing problems facing industry is the safe and inexpensive disposal of toxic waste products. The need for disposal of waste products of chemical processes and particularly of toxic waste products has given rise to serious problems because of the necessity of effecting the disposal without contaminating accessible areas of the land or the sea. These problems have become particularly acute in the disposal of waste products in the atomic industry. Many of the methods proposed have required excessive handling and are expensive, others require continuous monitoring, and still others require careful control of ambient conditions. One method, for example, has been the sealing of the waste products in containers which are then deposited in the ocean in areas of great depth. Such disposal of waste products is expensive and furthermore involves risk due to possible deterioration of the containers with time, and leakage due to imperfections in manufacture or to injury of the containers subsequent to their deposit on the ocean floor. Accordingly, it is an object of this invention to provide an improved and inexpensive method for effecting storage of chemical waste products.

It is another object of this invention to provide an improved and safe method for effecting the disposal of toxic waste products and the like.

It is another object of this invention to provide an improved method for effecting the safe and permanent disposal of toxic waste products such as those produced in the atomic industry.

Briefly, in carrying out the objects of this invention, a geologic formation is utilized wherein a relatively low datum pressure zone lies intermediate higher datum pressure zones. A well is then selected which enters the low pressure zone or such a well is drilled. The waste fluid products to be stored are then injected into the low pressure zone through the well bore whereupon they are held there because of the pressure differences prevailing and which differences prevent the migration of the toxic products to any other zones.

The features of novelty which characterize this invention are pointed out with particularity in the claims annexed to and forming a part of this specification.

For a better understanding of the invention in its various aspects together with further objects and advantages thereof, reference may be had to the following description taken in connection with the accompanying drawings, in which:

FIG. 1 is a diagrammatic view through a geologic section illustrating prevailing conditions suitable for the practice of the method of this invention;

FIG. 2 is a potentiometric surface map of an aquifer suitable for the practice of this invention;

FIG. 3 is a graph indicating the pressure datum fluid heads or potentiometric surface elevations prevailing in a plurality of the permeable formations of a geologic section including the formation of FIG. 2; and

FIG. 4 is a pressure-depth graph indicating the variations in pressure prevailing between the formations of FIG. 3.

During the course of the investigations undertaken in connection with the making of the present invention, it was observed that, in a geologic section comprising a plurality of aquifers or permeable zones separated by layers of shale, the datum pressures (i.e., pressures measured in each zone converted to a common datum elevation) varied widely. Furthermore, it was found that in certain aquifers the datum pressures were lower than the datum pressures in other aquifers both stratigraphically higher and stratigraphically lower in the sedimentary section. Further, it was observed that the pressures in one aquifer varied and that occasionally the water datum pressure in one portion of the aquifer would be found to be lower than the datum pressures in surrounding parts of the same aquifer. It was then established that these locations of low pressure provide reservoirs in which fluids, both gases and liquids, may be safely stored without likelihood of their migration to other portions of the formation.

By way of example, FIG. 1 illustrates a geologic section comprising permeable zones or aquifers **10**, **11**, **12**, **13**, **14**, and **15** lying at successive depths in a formation comprising relatively impermeable layers or shales indicated at **16**, **17**, **18**, **19**, and **20**. Wells have been indicated as drilled in the section, the first, indicated at **22**, being a water well for producing water from the formation **10**. The other wells are deep wells as indicated at **23**, **24**, **25**, **26**, **27**, **28**, and **29**. The wells **23**, **25**, and **29** all are in communication with the same formation **13**. The well **24** is extended to the formation **15**, the wells **26** and **27** to formations **12** and **11**, respectively, and the well **28** is in communication with the formation **14**. The shaded portions of the wells indicate the datum pressures of the aquifers converted to the same datum elevation.

It will now be observed that the pressure of the well **24** is lower than any of the others and that the datum pressures of the wells **23**, **25**, and **29** are somewhat higher than that of the well **24** but are lower than any of the other pressures. Further, it will be observed that the datum pressure of the well **28** entering the aquifer **14** is higher than the pressures in the aquifers **13** and **15**. Further, it will be observed that the pressures within the aquifer **13** as indicated by the shaded portions on the wells **23**, **25**, and **29** vary and that the pressure at the well **25** is lower than at either of the wells **23** and **29**. It will be understood that the flow of water along any one of the aquifers is of the so-called Darcy type and is proportional to the difference in pressure and dependent upon the permeability of the aquifer. Flow between the permeable aquifers or zones may result from both Darcy type flow through fractures, fault zones, and other permeable channels, and from osmotic-like movement of water through the very fine pore spaces of the geologic membrane-like shales which separate the permeable strata.

In order that conditions of the type indicated in FIG. 1 may exist over a long period of time, it is necessary that the water be moved by osmotic-like processes through the shales from the aquifers **13** and **15** to adjacent higher pressure aquifers such as **14** and **12**. Because the permeable aquifer **14** separates the aquifers **13** and **15**, any Darcy type flow from the aquifer **14** through a fault such as indicated at **31** is toward the aquifer **15** or aquifer **13**. However, even though the aquifer **15** is of lower pressure than the aquifer **13**, the interposition of the higher pressure aquifer **14** in fluid contact with the fault prevents a transfer of liquids via the fault from the aquifer **13** to the lower pressure aquifer **15**.

The fault **31** may be any form of permeable path, such as fractures, unpluged drill holes, wells with leaky cement seal around casing, etc., connecting the several aquifers.

Water entering this fault from a higher pressure aquifer will flow toward the lower pressure aquifers such as **13** and **15** and in general this Darcy type flow of water through the geologic section is indicated by the solid arrows.

Various procedures have been suggested for disposing of toxic fluids underground and these methods have raised a primary concern that the fluids so deposited might migrate along natural or man-made leakage paths from the injection zone to another zone or area where the contamination would be harmful. The various leakage paths which may be met include those occurring as faults or fractures in addition to the man-made paths such as occur in faulty cement work, leaky casings, and inadequately plugged drill holes, by way of example. However, toxic fluids injected into aquifer **13** could not leak via any such leakage paths from aquifer **13** to any other aquifer shown in this section. Consequently, aquifer **13** would provide a safe toxic fluid storage zone.

As an example of a particularly favorable storage site for toxic wastes and the like, there is shown in FIG. 2 a potentiometric surface map of an actual formation, the contour lines on the plot being indicated at intervals of 200 feet of potentiometric surface elevation. This plot might be considered generally by way of example as a map of the potentiometric surface elevations of a formation corresponding to that of the aquifer **13** in FIG. 1. The total area of the plot shown in FIG. 2 is of the general order of 150 by 200 miles. The pressure contours on this plot vary from the outer contour of 2000 feet of potentiometric surface elevation to an inner contour indicated at **35** which is the 400-foot contour. It will be observed that the potentiometric surface contours of the map decrease inwardly from the outer indicated 2000-foot contour to the inner 400-foot contour and that there is thus a wide area within which the formation fluid pressures vary in the manner generally indicated as that of the aquifer **13** in FIG. 1.

FIG. 2 clearly indicates the variation of datum pressures or potentiometric surface contours throughout a single permeable formation or aquifer and indicates a formation wherein the water flow is converging from all directions on the lowest pressure area. This inward flow of the Darcy law type is such that liquid introduced or injected at any part of the formation within the convergent flow area will migrate toward the lowest pressure area **35** and cannot escape. This injected fluid cannot escape by any natural processes without a change in the prevailing pressure conditions which are maintained by the geologic environment. The low pressures prevailing in the formation are maintained over long periods of geologic time by the continuous very low velocity osmotic transfer of water from the lower pressure zone to the high pressure zones through the ultrafine pore spaces which exist in the membrane-like shale separating the permeable aquifers.

Evidence secured in the field in addition to laboratory data and theoretical evaluation indicates that the osmotic membrane transfer of water can be caused by conditions such as (1) differences in electrical potential caused by variations in the degree of oxidation and reduction (redox potential) of the minerals, organic matter, and fluids in the rocks, (2) differences in temperature (thermal potential), and (3) differences in water salinity (chemical potential). The osmotic transfer of water through the shale pore spaces is completely harmless because (1) the extremely low velocities of movement of fluids in such fine pore spaces result in movement of only a few inches to a few feet in a thousand years, (2) toxic ions and many nonionized toxic products in the water are filtered out of solution by the membrane-like properties of shales, thereby preventing their movement through the shales, and (3) the very high cation exchange capacities of the clay mineral surfaces in the shale result in adsorbing a large portion of the toxic ions on the clay minerals, thereby fixing their location and preventing their further movement.

It will now be apparent that, if chemical wastes such as the toxic wastes from atomic operations are introduced through a well into a low pressure zone as illustrated, for example, by the zone about the well **25** in the formation **13** of FIG. 1, the wastes will flow into the formation and move outward away from well **25** at a rate determined by the fluid injection rate and injection pressure. The introduced liquid tends to flow into and will remain in the lowest pressure portion of the formation until that pressure has been changed.

As pointed out above, the toxic wastes introduced into a formation such as that shown at **13** in FIG. 1 cannot flow toward the higher pressure aquifers on either side thereof. This will be more readily apparent from FIG. 3 which is a graph of a vertical section through the center of the portion **35** of the formation of FIG. 2 and shows the other formations on either side thereof together with the datum pressures of the several aquifers, which portion **35** has a datum pressure equivalent to 455 feet of water above sea level as indicated by the liquid column **37**. Above the aquifer **35** there are aquifers **38** and **39** having datum pressures of 4600 feet and 1750 feet, respectively. Below the aquifer **35** there are aquifers **40**, **41**, **42**, and **43** which have datum pressures equivalent to water levels of 1470, 1770, 1860, and 1905 feet above sea level, respectively. It will thus be noted that the aquifer **35** has a datum pressure over 1000 feet of water head less than that of either adjacent aquifer.

FIG. 4 is a datum elevation pressure curve to show the relationship of the pressures in the several aquifers indicated in FIG. 3. This graph, which indicates the variation in pressure between the many geologic formations, includes a sloping line **44** which indicates the pressures which would exist in the various formations if fresh hydrostatic conditions prevailed through the entire sedimentary section. It will be noted that the pressures in the formation as indicated by the points marked for each of the formations **35**, **38**, **39**, **40**, **41**, **42**, and **43** vary widely from the conventionally expected hydrostatic pressure head.

In FIG. 4 lines drawn parallel to the line **44** through the pressure points indicated will intersect zero elevation at the datum pressure for the point through which the line is drawn and will intersect the zero pressure axis at the elevation of the potentiometric surface of this point. Thus a line drawn through the point corresponding to the aquifer **35** and parallel to the line **44** will intersect the zero pressure or vertical axis at 455 feet of water above sea level, which is the potentiometric surface elevation indicated in FIG. 3.

It will thus be seen that the example of the formation **35** as indicated in FIGS. 2, 3, and 4 provides a region of convergent flow of water within the selecter aquifer and further provides a formation in which no flow of the deposited wastes can occur outwardly from the selected low-pressure aquifer. The selected aquifer is thus one which has both laterally and vertically convergent flow of substantial magnitude and such environment is safe because the injected toxic fluid cannot escape along any Darcy flow leakage path. The only escape of fluid from the injection zone is by osmotic-type transfer of water through the ultrafine pore spaces of the shale membranes, and as pointed out above this escape path is completely harmless even over long periods of geologic time.

In addition to employing convergent flow aquifers, the method of this invention is practical under other flow conditions. For example, a low datum pressure aquifer such as **15** in FIG. 1 can be a safe zone for toxic waste storage even though flow in **15** may be all in one direction and not converging. If the rate of flow of water in aquifer **15** is sufficiently low so that the time required for the toxic wastes to move to areas where the water is used for other purposes is very much longer than the dangerous life of the waste, then **15** would be a safe disposal zone. For example, representative rates of flow in permeable

geologic formations are from a few inches to a few tens of feet per year. Therefore, if the dangerous life of a particular waste is 200 years and the rate of flow is 2 feet per year, the waste would travel 400 feet during the dangerous life of the waste. Under such flow conditions it would be safe to store wastes of 1000-year-danger life in aquifer 15 if the reservoir water from 15 were not used for other purposes at locations closer than a few miles from the injection site.

The capacity of even a relatively small permeable lens for storing waste material is large. For example, a 3-mile by ½-mile wide 30-foot sand lens with 15 percent porosity would contain approximately 1,500,000,000 gallons or approximately 33,000,000 barrels of formation water. If the waste material is injected in one or more wells and a comparable amount of formation water is withdrawn from other wells in a manner to control properly the fluid flow pattern, approximately one-half of the formation, that is, 16,000,000 barrels, may be replaced before any injection water is produced in the withdrawal wells. If the toxic waste material exists as a dissociated cation, then, due to cationic exchange of the mineral surfaces, from 3 to 10 times this volume of water, that is, from 50,000,000 to 150,000,000 barrels, may be injected before any toxic cations are produced at the withdrawal wells.

The electrochemical membrane flow, that is, osmotic flow, is the important factor in creating the desired low datum pressure environment which safely traps any injected toxic fluid. Although numerous faults, fractures, leaky casings, poor cement jobs, or other leakage paths exist between the zones which must be protected from contamination and the permeable lens selected for waste disposal, no toxic fluid can escape therefrom because all fluid flow along these leakage paths will be toward the injection lens. The only flow out of this injection lens will be by the very slow osmotic membrane movement of water through the shale pore spaces where any toxic cations will be substantially excluded by membrane filtration or absorbed by ion exchange processes. Consequently, an aquifer located in this low datum pressure environment is a safe place to dispose of toxic fluids.

When a formation has been selected for the disposal of chemical wastes or other toxic fluids, a plurality of injection wells may be drilled in a row, the spacing being in the order of 40 acres, and a second row of withdrawal wells may be drilled along a line approximately offsetting the injection wells by 40-acre spacing. When atomic waste is injected in the first row of wells, reservoir water is withdrawn from the withdrawal well to obviate the need for high injection pressures and to control the movement of waste into the reservoir. The fluids from each withdrawal well will be monitored to detect the presence of the waste, for example by the introduction of a Geiger counter in the monitoring well, and as soon as the waste appears at that well, injection at the first well is stopped and a third well drilled beyond the withdrawal well is then employed as a withdrawal well and injection is begun through the first withdrawal well. By this procedure a great area may be covered at a rate dependent upon the actual distribution of the waste materials in the formation.

The method of this invention provides for the storage of vast quantities of chemical wastes and particularly of toxic wastes without the danger of contamination of other zones throughout indefinite periods in a manner that the storage is effectively permanent, safe, and inexpensive. Toxic atomic wastes can be stored in this manner indefinitely without danger of leakage through any fracture or other damage to the reservoir formation.

While the invention has been described in connection with particular environments and specific formations, various other applications and different formation conditions may be employed. Therefore it is not desired that the invention be limited to the specific details illustrated and described and it is intended by the appended claims to cover all modifications which fall within the spirit and scope of the invention.

We claim:

1. The method for storing fluids, which comprises utilizing a geologic section having first and second and third relatively permeable zones spaced from one another and separated by layers of low permeability, the second zone being located intermediate the first and third zones and having a datum pressure lower than those of the first and third zones, providing a well bore extending into said second zone, and injecting fluid to be stored through said bore into said second zone.

2. The method of storing fluids as set forth in claim 1 including the steps of drilling into said second zone a second bore spaced from said first bore, withdrawing reservoir fluid from the second bore and monitoring the fluid withdrawn from the second bore to determine when fluid injected through said first bore reaches said second bore, and thereupon stopping injection into said first bore and thereafter injecting fluid into said second bore.

3. The method of storing fluids as set forth in Claim 2 including the steps of drilling a third bore into said zone and withdrawing reservoir fluid from the third bore.

4. The method of storing fluids as set forth in claim 1 including drilling into said second zone a second bore spaced from said first bore, withdrawing reservoir fluid from the second bore and monitoring the fluid withdrawn from the second bore to determine when fluid injected through said first bore reaches said second bore, and thereupon stopping withdrawal from said second bore.

5. The method for effecting permanent storage of fluids for toxic-waste-product disposal and the like, which comprises utilizing a geologic section having first and second and third relatively permeable zones spaced from one another and separated by layers of low permeability, the second zone being located intermediate the first and third zones and having a datum pressure lower than those of the first and third zones, providing a well bore extending into said second zone, and injecting fluid to be stored through said bore into said second zone.

6. The method for storing fluids and for preventing leakage of the stored fluids, which comprises utilizing a geologic section having a zone in which all natural pressure induced fluid flow is converging by means of only the naturally existing fluid flow characteristics of the geologic section toward one part of the geologic section, providing a well bore extending into said zone, and injecting fluid to be stored through said bore into said zone.

7. The method of storing fluids as set forth in claim 6 including the steps of drilling into said zone a second bore spaced from said first bore, withdrawing reservoir fluid from the second bore and monitoring the fluid withdrawn from the second bore to determine when fluid injected through said first bore reaches said second bore, and thereupon stopping withdrawal from said second bore.

8. The method of storing fluids as set forth in claim 7 including the steps of drilling a third bore into said zone and withdrawing reservoir fluid from the third bore.

9. The method for effecting permanent storage of fluids for toxic waste disposal and the like and for preventing leakage of the stored fluids, which comprises utilizing a geologic section having a zone in which all natural pressure induced fluid flow is converging by means of only the natural existing fluid flow characteristics of the geologic section toward one part of the geologic section, providing a well bore extending into said zone, and injecting fluid to be stored through said bore into said zone.

10. The method of storing fluids as set forth in claim 9 including the steps of drilling into said zone a second bore spaced from said first bore, withdrawing reservoir fluid from the second bore and monitoring the fluid withdrawn from the second bore to determine when fluid injected through said first bore reaches said second bore, and thereupon stopping withdrawal from said second bore.

11. The method of storing fluids as set forth in claim 10 including the steps of drilling a third bore into said zone and withdrawing reservoir fluid from the third bore.

12. The method for effecting permanent storage of fluids for toxic waste disposal and the like which comprises utilizing a geologic section having a zone in which fluid flow is converging toward one part of the geologic section, providing a well bore extending into said zone, injecting fluid to be stored through said bore into said zone, drilling into said zone a second bore spaced from said first bore, withdrawing reservoir fluid from the second bore and monitoring the fluid withdrawn from the second bore to determine when fluid injected through said first bore reaches said second bore, and thereupon stopping withdrawal from said second bore.

13. The method of storing fluids as set forth in claim 12 including the steps of drilling a third bore into said zone and withdrawing reservoir fluid from the third bore.

14. The method of storing fluids which comprises utilizing a geologic section having a zone in which fluid is converging toward one part of the geologic section providing a well bore extending into said zone, injecting fluid to be stored through said bore into said zone, drilling into said zone a second bore spaced from said first bore, withdrawing reservoir fluid from the second bore and monitoring the fluid withdrawn from the second bore to determine when fluid injected through said first bore reaches said second bore, and thereupon stopping withdrawal from said second bore.

15. The method of storing fluids as set forth in claim 14 including the steps of drilling a third bore into said zone and withdrawing reservoir fluid from the third bore.

16. The method for storing fluids and for preventing leakage of the stored fluids, which comprises utilizing a geologic section having a plurality of geologic layers and including a zone in which all natural pressure induced fluid flow produced only by the naturally existing fluid flow characteristics of the geologic section is toward one geologic layer which is at a lower relative natural pressure than the other layers of said zone, providing a well bore into said zone, and injecting fluid to be stored through said well bore into said zone.

17. The method of storing fluids as set forth in claim 16 including the steps of drilling into said zone a second bore spaced from said first bore, withdrawing reservoir fluid from the second bore and monitoring the fluid withdrawn from the second bore to determine when fluid injected through said first bore reaches said second bore, and thereupon stopping withdrawal from said said second bore.

18. The method of storing fluids as set forth in claim 17 including the steps of drilling a third bore into said zone and withdrawing reservoir fluid from the third bore.

19. The method for storing fluids which comprises utilizing a geologic section having a plurality of geologic layers and including a zone in which fluid flow is toward one geologic layer which is at a lower relative pressure than the other layers of said zone, providing a well bore into said zone, injecting fluid to be stored through said well bore into said zone, drilling into said zone a second bore spaced from said first bore, withdrawing reservoir fluid from the second bore and monitoring the fluid withdrawn from the second bore to determine when fluid injected through said first bore reaches said second bore, and thereupon stopping withdrawal from said second bore.

20. The method of storing fluids as set forth in claim 19 including the steps of drilling a third bore into said zone and withdrawing reservoir fluid from the third bore.

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