

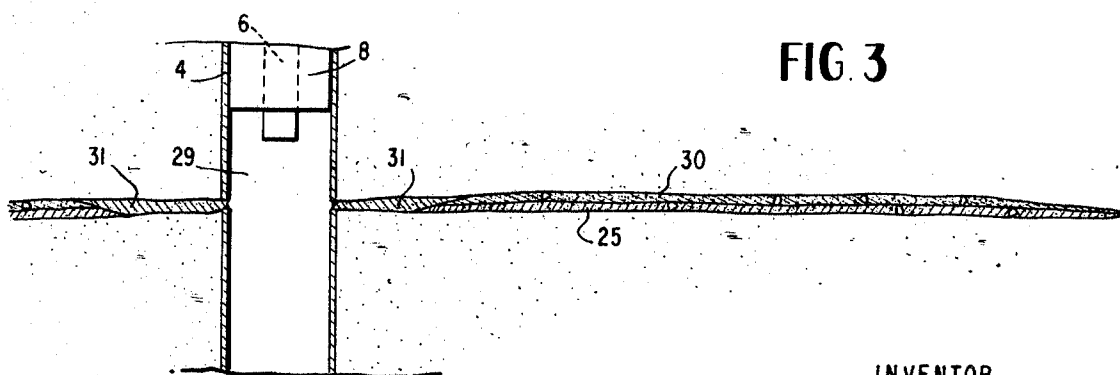
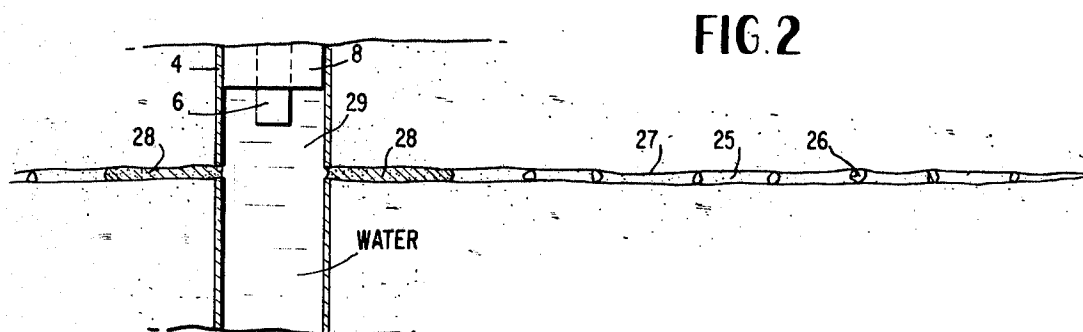
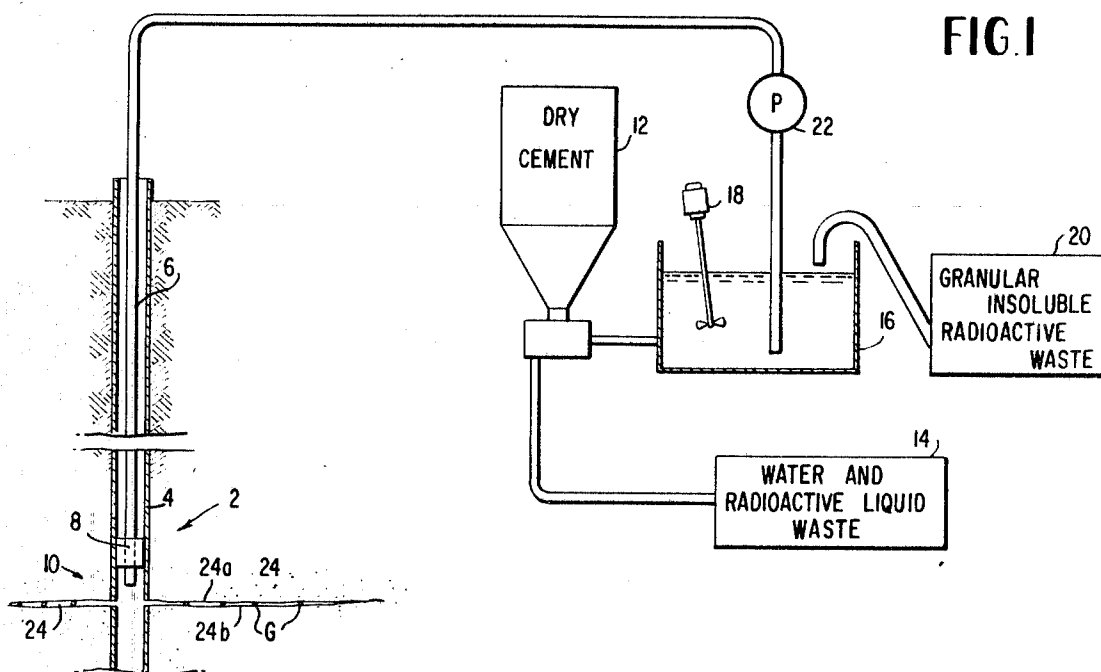
May 19, 1970

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3,513,100

METHOD FOR SUBSURFACE DISPOSAL OF RADIOACTIVE WASTE

Filed Sept. 25, 1967



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3,513,100 METHOD FOR SUBSURFACE DISPOSAL OF RADIOACTIVE WASTE

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Filed Sept. 25, 1967, Ser. No. 670,034

Int. Cl. G21c 19/44

U.S. Cl. 252—301.1

7 Claims

ABSTRACT OF THE DISCLOSURE

A method for disposing of high level, solid, radioactive waste material by delivering such material in a continuous, water-phase cement to a subterranean formation. The solid waste is used as a propping agent to hold formation fractures open as a slurry containing the solid, radioactive waste material is pumped thereinto.

BACKGROUND OF THE INVENTION

This invention relates to the disposal of radioactive waste and more particularly, to the disposal of radioactive waste by storage in underground formations.

The utilization of atomic energy for producing power or other purposes results in an accumulation of radioactive waste material which must be disposed of in such a way that it is not harmful to public safety. The rate at which the radioactivity of the material decays is such that a great deal of time is required before the waste material is rendered sufficiently harmless that it may be disposed of by dilution or other conventional techniques. While the radioactive waste is undergoing decay, it must be shielded. The shielding must be substantially impermeable to avoid the danger of leakage of the radioactive waste through or around the shield.

It has been popular recently to inject low and intermediate level wastes into subterranean chambers that are sealed with cement or other means. Low level wastes typically comprise liquid solutions, in which relatively small amounts of radioactive substances are dissolved. These solutions require relatively large volumes of storage space, in relation to the weight of radioactive material contained in the solution. As the high rate of production of radioactive wastes increases, disposal of low level wastes in this manner becomes unsatisfactory because the available subterranean formations are of limited capacity.

High level wastes consist of solid, insoluble particles which produce radiant energy at a high rate. These particles have been disposed of by casting large concrete blocks containing the particles and then sinking the blocks in the ocean in deep water. The high rate of radiation emanating from these solid, high level wastes requires that this waste be adequately shielded. Such shielding incurs expense. Further, there is always the possibility of shield rupturing and the conveying of released waste by ocean currents.

In a U.S. Nelson Pat. 3,262,274, a technique is disclosed for disposing of radioactive waste materials. FIG. 1 of this patent discloses an oil-base cement containing solid radioactive material which is disposed of within a subterranean cavern. While Nelson proposes the disposal of solid radioactive waste as a constituent of cement, pumped into a subterranean formation, it was not obvious to Nelson to utilize solid radioactive waste materials as fracture propping agents. When these solid radioactive waste particles are thus employed, in practicing the present invention, they are not only positively locked into a formation, even while the cement is fluid, but serve as vehicles for providing an enlarged subterranean cavity to receive the waste radioactive substance.

Significantly, the oil-base cement featured in the Nelson disclosure, after hardening, may be permeable or porous so as to be vulnerable to subsequent leaching. Such permeability may result, for example, from the subterranean crushing of hardened cement having high porosity attributable to the initial oil suspending media or from the mere setting of the oil-base cement.

Further, patents such as Nelson wholly fail to recognize or propose techniques for mixing cement and solid radioactive waste so as to simultaneously minimize mixing time and control the amount of radioactive materials incorporated in a cement slurry at a well head.

In recognition of problems such as those described above, it is an object of the present invention to provide a method for disposing of solid radioactive waste material wherein this solid material is pumped into formation fractures and employed to prop these fractures open.

It is a further object of the invention to transport such solid radioactive waste into a well bore, while the solid radioactive waste is suspended in and generally uniformly distributed through, a continuous water-phase cement.

It is, likewise an object of the invention to provide a technique for effecting the rapid and controlled mixing of cement constituents and radioactive materials at a well head so as to speed the disposal operation and promote overall safety in the handling of radioactive materials.

SUMMARY OF THE INVENTION

In accomplishing the objects of the invention, a method of disposing of high level solid radioactive waste materials is presented. In practicing this method, dry cementitious material is mixed with fluid radioactive waste, including water. The mixed dry cement and the fluid radioactive waste is agitated. Particulate, high level, solid, radioactive waste material is injected into the dry cement and fluid radioactive waste while agitating the overall mixture so as to substantially uniformly disperse the particles of solid high level radioactive waste. The injection of the solid radioactive waste material into the mixture is metered so as to limit and control the solids content of the mixture. The mixture is conveyed to a well bore and pumped thereinto while retarding the setting of the cement components of the mixture and maintaining a continuous water phase in the mixture. Sufficient pressure is applied to the mixture, adjacent a formation, to effect formation fracturing and force the mixture into formation of the solid radioactive waste material into the mixture fracture means. The formation fracture means is propped open with the solid radioactive waste material. The mixture is then caused to solidify within the propped-open formation fracture means. An annular seal of cement, free of radioactive material, is interposed radially between the mixture and the well bore.

DESCRIPTION OF THE DRAWINGS

This preferred embodiment is illustrated in the accompanying drawings showing schematically the apparatus for carrying out the method of this invention. In these drawings:

FIG. 1 illustrates a well bore and a controlled mixing apparatus for quickly mixing dry cement, fluid, and solid radioactive material for injection into a formation through the well bore;

FIG. 2 illustrates the uniquely effective technique for injecting radioactive waste into an annular subterranean zone and sealing the radioactive material using an annular, non-radioactive, cementitious seal encircling a well bore; and

FIG. 3 illustrates a modification of the FIG. 2 disposal technique wherein a single non-radioactive annular cement seal is employed to isolate a plurality of superposed radioactive lenses from a well bore.

RADIOACTIVE WASTES

Radioactive liquid waste is generally divided into three categories: low level, intermediate level, and high level. Low level radioactive waste is usually in liquid form with the radioactive constituent being dissolved in a liquid solvent. The solvent may be water or another liquid. For example, the low level waste may be a solution containing radioactive isotopes such as strontium-90 and cesium-137. The specific gravity of the low level waste solution may be greater than 1, and the radioactivity of the waste is less than 10^{-6} curies per gallon.

Intermediate level liquid waste has a radioactivity level in the range of .001 to 1.0 curie per gallon. This waste is produced primarily in the chemical reprocessing of irradiated fuel elements containing fissionable material.

High level liquid waste has a radioactivity greater than 100 curies per gallon. These highly radioactive liquid wastes are a product of the reprocessing of fuel elements. High level, radioactive liquids and sludges are usually aqueous solutions or dilute suspensions of fission products. Generally, the radioactivity of these liquid wastes ranges between 500 and 10,000 curies per gallon.

In addition to the various liquid wastes described above, it is also necessary to dispose of solid waste that is highly radioactive. High level solid waste has a radioactivity of between 500 and 100,000 curies per gallon. Solid wastes may include spent fissionable material which may be sintered or granular products from a fluidized bed that was used to convert high level liquid waste into granular solids. If these solids are stored underground, the storage sites must be sealed from the surrounding formation because of the danger of leaching by subterranean water.

PREFERRED DISPOSAL TECHNIQUES

Referring to FIG. 1, a bore hole is drilled through a formation 2 and a casing 4 is cemented in the bore hole. A cementing string 6 is run in the casing 4 in a conventional manner, with a packer 8, shown schematically on the cementing string 6.

At the appropriate depth, the packer 8 is expanded to isolate a formation zone 10, which is to be fractured in accordance with the method of this invention. In accordance with conventional practice, the casing 4 is perforated at the zone 10 to establish communication between the formation and the interior of the casing 4. Although the bore hole is shown as having a casing 4 extending below the zone 10, the bore hole may be uncased or the lower end of the casing may be positioned above the zone 10, in which case it may be unnecessary to perforate prior to fracturing the formation.

Unique batch cement mixing apparatus utilized for mixing the dry pulverulent cement with dry granular, water insoluble, high level radioactive waste is shown in FIG. 1. The cement is stored in a tank 12 and is mixed as a slurry with fluid, low, intermediate or high level liquid radioactive waste, including water. This waste is supplied from a storage tank 14. Apparatus for effecting the mixing of a dry cement and liquid to form such a slurring is described, for example, in chapter 14 of *Petroleum Engineering*, by Carl Gatlin (Prentiss-Hall, Inc., 1960). The slurry of cement and water is transferred to a mixing tank 16. The tank 16 has a power stirrer 18 to provide continual agitation of the liquid in the tank. Dry, granular, high level, solid radioactive waste is dispensed from a tank 20 into the mixing tank 16 at a predetermined rate, corresponding to the rate of supply of cement from the tank 12. The injection of the solid radioactive material from the tank or receptacle 20 is positively controlled or metered as described, for example, in the U.S. Hathorn et al. Pat. 3,161,203 or the U.S. Copland et al. Pat. 3,090,439. Through such metering, the ratio of radioactive solid material to the overall mass of the mixture may be positively controlled. This will enable a mixing operator to insure that adequate solid materials are available for propping purposes, while avoiding too high a

solid content which would impede the basic mixing and fracturing operations.

Friction reducers and cement setting retarding agents are mixed with the slurry in the tank 16. Representative retarding agents to slow the setting of cement in a well bore are set forth on page 273 of the treatise *Petroleum Engineering*, by Carl Gatlin (Prentiss-Hall, Inc., 1960).

When the slurry has been thoroughly mixed so as to uniformly disperse the solid radioactive waste, it is injected into the cementing string 6, under pressure by means of the pump 22. The pressure of the cementing fluid is sufficiently great to cause lateral fractures 24 in the zone 10. The cement slurry flows into the fractures. Since the solid radioactive waste granules G are contained in the slurry, the waste material is embedded in the fractures 24. The pumping is continued until substantially all of the high level radioactive waste is deposited in the fractures.

In the fracture 24, the solid radioactive waste granules G function as a propping agent, holding the fracture open. The walls 24a and 24b of the fracture, in engaging the solid waste, secure it within the fracture and prevent it from discharging into the well bore while the slurry sets.

Additional cement slurry that does not contain radioactive material is mixed in the tank 16 and follows the radioactive slurry through the cementing string 6 to seal the casing 4 below the packer 8.

It is desirable to mix setting retardants with the slurry to allow sufficient time for the slurry to be pumped into the fractures before the slurry becomes unpumpable.

The solid, high level radioactive waste is usually in the form of sand or gravel-like granules. The usually sharp, angular configuration of this material is particularly desirable in carrying out the fracturing steps in accordance with the method of this invention. The sharp corners of the particles resist displacement of the particles relative to each other and relative to the walls of the fracture, and thus the particles act as locked-in propping agents to maintain the fractures in an open condition as the cement is being pumped into the formation, and while the cement hardens.

It is in order to supplement the disposal rate of radioactive wastes, that liquid radioactive waste is mixed with the water prior to mixing this resulting fluid and cement. The total radioactive content from the slurry should be maintained at a sufficiently low level, however, so as not to endanger the health of the operating personnel.

It is particularly important for the resulting concrete structure to be impermeable to liquids. Since water may flow through or around the formation in which the radioactive waste has been placed, there is a danger that radioactive materials will be leached from the concrete, if the concrete is sufficiently porous to allow the water to flow through the interior of the concrete. It has been found that the slurry of this invention forms a non-porous concrete structure, since it does not contain emulsifiers or substantial quantities of oil or other additives which might cause the resulting concrete to be porous. Specifically, the slurry must contain less than 5% of oil or other liquid that does not react with the hydraulic cement. If higher amounts of oil or other liquids are present in the slurry, they may tend to generate gas or cause the formation of pockets, or to contribute generally to a permeable concrete formation, which is undesirable.

FIG. 2 illustrates a technique for uniquely isolating a mass of radioactive material, dispersed in cement, from a well bore.

As there shown, a lens of cement material 25 containing solid radioactive material 26 has been injected into a formation fracture 27 as previously described. The slurry containing the radioactive material is followed into the well bore by a non-contaminated mass of cement slurry, i.e., a water-base cement or slug, free of radioactive material. This mass of non-contaminated cement is pumped back into the formation.

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The mass or "slug" of non-contaminated cement is followed by water. This water will occupy the well bore 29 so as to insure the formation of an annular seal 28 contained substantially within the fracture 27. The seal 28 may be allowed to harden while pressure is maintained on water within the well bore. The hardened cement seal 28 provides an effective annular, seal isolating a well bore 29 from the radioactive lens 25, thereby minimizing the danger of leakage or leaching of radioactive material from the lens 26 into the well bore.

FIG. 3 illustrates a significant extension of the concept shown in FIG. 2.

As shown in FIG. 3, after the lens of radioactive material 25 has been formed, it is allowed to harden in place without interposing the uncontaminated seal 28. After the lens 25 is formed, a subsequent lens 30 of radioactive containing material is formed, as described generally in connection with the FIG. 1 embodiment. This process of forming superposed contiguous lenses of radioactive cementitious material may be continued until a series of several stacked lenses is formed.

As will be appreciated, the fracturing of each lens, after a prior lens has been formed, is facilitated by the generally planar discontinuity between the initially formed lenses and the adjacent formation. Such discontinuity provides a zone uniquely vulnerable to fracturing.

After the desired number of superposed lenses have been formed, a seal 31 of radioactive free cement may be installed, substantially as described in connection with the seal 28 of the FIG. 2 embodiment.

The method of this invention permits the disposal of high level solid radioactive waste in a convenient, safe manner without danger of contamination to personnel. Since the solid waste is disposed of in solid form, the disposal volume that is required is considerably less than that required by other methods wherein the solids are diluted or otherwise transformed to liquids which are subsequently disposed of. Furthermore, the method of this invention utilizes existing cementing apparatus and therefore investment in additional or new equipment is not required in order to carry out this process.

MAJOR ADVANTAGES AND SCOPE OF THE INVENTION

One principal advantage of the invention resides in the disclosed technique for minimizing the time involved in mixing cementitious material and solid radioactive wastes. Time is conserved, so as to reduce radiation exposure hazards, by directly mixing dry cement with fluid radioactive wastes and then immediately blending in the solid radioactive waste materials.

The metering of the solid radioactive waste materials during the mixing operation provides effective control. The solids content of the resulting mixture may be accurately regulated so as to provide a mixture which may be readily pumped through a well bore. In regulating the solids content of the mixture, the overall content of radioactive material of the mixture will be controlled. The dispersion of the solids in the slurry, coupled with their becoming locked into fractures while functioning as propping agents, effectively disperses the radioactive material within the formation. This minimizes the deleterious effects of radioactive material generated heat.

The use of the solid radioactive materials as a fracture propping agent serves two functions. The material, through a "boot-strap" technique, holds formation fractures open so as to allow the fractures to accommodate greater quantities of radioactive waste. The solid radioactive materials which function as propping agents are effectively locked into the fracture by the formation itself. Thus, the chances of solid high level radioactive waste materials migrating back into a well bore before the cement has set are substantially minimized or altogether eliminated.

The annular non-radioactive seal interposed radially between the well bore and a fracture formed with radio-

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active material provides a uniquely simple but highly effective technique for avoiding well bore contamination. With this seal, the well bore may be used again and again for the purpose of injecting radioactive materials without the danger that would result from the presence of radioactive materials in the well bore. Significantly, this safety advantage is achieved without having to block or obstruct the well bore itself.

The annular seal is most advantageously employed to isolate a plurality of superposed lenses of injected radioactive materials. This utilization of the annular seal economizes on the time, material and equipment required to effect the sealing operation.

In describing the invention, reference has been made to preferred embodiments. However, those skilled in the art and familiar with the disclosures of the invention may well recognize additions, deletions, substitutions or other modifications which would fall within the purview of the appended claims.

I claim:

1. A method of disposing of solid, radioactive, particulate waste material, said method comprising:

mixing dry cementitious material with fluid comprising water;

agitating the said mixed dry cement and said fluid; injecting particulate, solid, radioactive waste material into said dry cement and fluid while agitating the mixture thereof to substantially uniformly disperse the particles of said solid, radioactive waste;

conveying said mixture to a well bore and pumping said mixture into a well bore while maintaining a continuous water phase in said mixture;

applying sufficient pressure to said mixture adjacent a formation to effect formation fracturing and force said mixture into formation fracture means;

propping said formation fracture means open with said solid radioactive waste material; causing said mixture to solidify within said propped formation fracture means.

2. A method of disposing of high-level, solid, radioactive, particulate waste material, said method comprising:

mixing dry cementitious material with fluid radioactive waste, including water;

agitating the said mixed dry cement and said fluid radioactive waste;

injecting particulate, high-level, solid, radioactive waste material into said dry cement and fluid radioactive waste while agitating the mixture thereof to substantially uniformly disperse the particles of said solid, high-level, radioactive waste;

metering the injection of said solid radioactive material to limit the solids content of said mixture; conveying said mixture to a well bore and pumping said mixture into a well bore while

retarding the setting of the cement component of said mixture, and maintaining a continuous water phase in said mixture;

applying sufficient pressure to said mixture adjacent a formation to effect formation fracturing and force said mixture into formation fracture means;

Propping said formation fracture means open with said solid radioactive waste material; causing said mixture to harden within said propped formation fracture means.

3. A method as described in claim 2

wherein said cement and solid radioactive waste is mixed with fluid comprising at least 95% water by volume;

wherein a cement slurry substantially free of radioactive material is pumped through said well bore and allowed to harden therein, thereby forming a seal for said mixture.

4. A method as described in claim 3:

wherein said cementitious material free of radioactive

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material is pumped into said formation fracture means radially between said well bore and the mixture of cement slurry and radioactive material; and wherein water is circulated through said well bore and past said formation fracture means to remove cementitious material from said well bore and leave an annular formation of cement free of radioactive material interposed radially between said well bore and the said radioactive material.

5. A method as described in claim 4:

wherein a first mixture of cement and radioactive material is injected into one formation fracture and allowed to there harden; and

wherein a second mixture of radioactive material and cement is injected into a second fracture vertically adjacent said first mixture and allowed to there harden; and

wherein said mixture of cement slurry free of radioactive material is posed radially between said well bore and each of said vertically aligned and independently hardened mixture of cement and radioactive materials.

6. A method of disposing of radioactive waste material comprising:

dispersing solid radioactive material in a cement slurry; pumping said cement slurry into a well bore;

forcing said cement slurry and radioactive material into a formation fracture communicating with said well bore;

injecting cementitious material, free of radioactive material, into said fracture radially between said well bore and the mixture of cement slurry and radioactive material; and

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circulating water through said well bore and past said fracture to remove cementitious material from said well bore and leave an annular formation of cement, free of radioactive material, interposed radially between said well bore and the said radioactive material.

7. A method as described in claim 4:

wherein a first mixture of cement slurry and radioactive material is injected into one formation fracture and allowed to there harden; and

wherein a second mixture of radioactive material and cement slurry is injected into a second fracture vertically adjacent said first hardened mixture of radioactive material and allowed to there harden, and

wherein said cementitious material, free of radioactive material, is interposed radially between said well bore and each of said vertically aligned and independently hardened mixtures of cement slurry and radioactive materials.

References Cited

UNITED STATES PATENTS

3,108,439	10/1963	Reynolds et al.	252—301.1 X
3,262,274	7/1966	Nelson	252—301.1 X
3,272,756	9/1966	Kaser	252—301.1

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