Chapter 4
Hydraulic Fracturing Fluids

This chapter summarizes the information EPA collected on the types and volumes of fracturing fluids and additives that may be used for hydraulic fracturing of coalbed methane wells. This chapter also provides EPA's evaluation of the fate and transport of fracturing fluids that are injected into targeted coal layers during the hydraulic fracturing process. This evaluation was conducted to provide the Agency with information on whether a Phase II study is warranted. Captioned photographs in this chapter show the use of fracturing fluids at a coalbed methane well (Figures 4-1 through 4-11 at the end of this chapter).

4.1   Introduction

The types and use of fracturing fluids have evolved greatly over the past 60 years and continue to evolve. The U.S. oil and gas industry has used fluids for fracturing geologic formations since the early 1940s (Ely, 1985). *The Handbook of Stimulation Engineering* (Ely, 1985), a comprehensive history of the evolution of hydraulic fracturing fluids in the oil and gas industry, was used as a source of information for this chapter. In addition, EPA identified fluids and fluid additives commonly used in hydraulic fracturing through literature searches, reviews of relevant MSDSs provided by service companies, and discussions with field engineers, service company chemists, and state and federal employees.

Available scientific literature indicates that hydraulic fracturing fluid performance became a prevalent research topic in the late 1980s and the 1990s. Most of the literature pertaining to fracturing fluids relates to the fluids' operational efficiency rather than their potential environmental or human health impacts. There is very little documented research on the environmental impacts that result from the injection and migration of these fluids into subsurface formations, soils, and USDWs. Some of the existing literature does offer information regarding the basic chemical components present in most of these fluids. The composition of fracturing fluids and additives is discussed in detail in the next section.

The main goal of coalbed hydraulic fracturing is to create a highly conductive fracture system that will allow flow through the methane-bearing coal zone to the production well used to extract methane (and groundwater). Hydraulic fracturing fluids are used to initiate and/or expand fractures, as well as to transport proppant into fractures in coalbed formations. Proppants are sand or other granular substances injected into the formation to hold or "prop" open coal formation fractures created by hydraulic fracturing. The viscosity of fracturing fluids is considered when they are formulated, to provide for efficient transport and placement of proppant into a fracture. Most of the fracturing

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fluids injected into the formation are pumped back out of the well along with groundwater and methane gas (see section 3.3 in Chapter 3 for a more detailed discussion of fracturing fluid recovery).

4.2   Types of Fracturing Fluids and Additives

Service companies have developed a number of different oil- and water-based fluids and treatments to more efficiently induce and maintain permeable and productive fractures. The composition of these fluids varies significantly, from simple water and sand to complex polymeric substances with a multitude of additives. Each type of fracturing fluid has unique characteristics, and each possesses its own positive and negative performance traits. For ideal performance, fracturing fluids should possess the following four qualities (adapted from Powell et al., 1999):

- Be viscous enough to create a fracture of adequate width.

- Maximize fluid travel distance to extend fracture length.

- Be able to transport large amounts of proppant into the fracture.

- Require minimal gelling agent to allow for easier degradation or "breaking" and reduced cost.

Water-based fracturing fluids have become the predominant type of coalbed methane fracturing fluid (Appendix A: DOE, Hydraulic Fracturing). However, fracturing fluids can also be based on oil, methanol, or a combination of water and methanol. Methanol is used in lieu of, or in conjunction with, water to minimize fracturing fluid leakoff and enhance fluid recovery (Thompson et al., 1991). Polymer-based fracturing fluids made with methanol usually improve fracturing results, but require 50 to 100 times the amount of breaker (e.g., acids used to degrade the fracturing fluid viscosity, which helps to enhance post-fracturing fluid recovery) (Ely, 1985). In some cases, nitrogen or carbon dioxide gas is combined with the fracturing fluids to form foam as the base fluid. Foams require substantially lower volumes to transport an equivalent amount of proppant. Diesel fuel is another component of some fracturing fluids although it is not used as an additive in all hydraulic fracturing operations. A variety of other fluid additives (in addition to the proppants) may be included in the fracturing fluid mixture to perform essential tasks such as formation clean up, foam stabilization, leakoff inhibition, or surface tension reduction. These additives include biocides, fluid-loss agents, enzyme breakers, acid breakers, oxidizing breakers, friction reducers, and surfactants such as emulsifiers and non-emulsifiers. Several products may exist in each of these categories. On any one fracturing job, different fluids may be used in combination or alone at different stages in the fracturing process. Experienced service company engineers will devise the most effective fracturing scheme, based on formation characteristics, using the fracturing fluid combination they deem most effective.

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The main fluid categories are:

- Gelled fluids, including linear or cross-linked gels.

- Foamed gels.

- Plain water and potassium chloride (KCl) water.

- Acids.

- Combination treatments (any combination of 2 or more of the aforementioned fluids).

Some of the fluids and fluid additives may contain constituents of potential concern. Table 4-1, at the end of section 4.2.6, lists examples of chemicals found in hydraulic fracturing fluids according to the MSDSs provided by service companies, and potential human health effects associated with the product. It is important to note that information presented in MSDSs is for pure product. Each of the products listed in Table 4-1 is significantly diluted prior to injection.

EPA also obtained two environmental impact statements that were prepared by the Bureau of Land Management (BLM). In these impact statements, BLM identified additional chemical compounds that may be in fracturing fluids including methyl tert butyl ether (MTBE) (U.S. Department of the Interior, CO State BLM, 1998). However, EPA was unable to find any indications in the literature, on MSDSs, or in interviews with service companies that MTBE is used in fracturing fluids to stimulate coalbed methane wells.

4.2.1   Gelled Fluids

Water alone is not always adequate for fracturing certain formations because its low viscosity limits its ability to transport proppant. In response to this problem, the industry developed linear and cross-linked fluids, which are higher viscosity fracturing fluids. Water gellants or thickeners are used to create these gelled fluids. Gellant selection is based on formation characteristics such as pressure, temperature, permeability, porosity, and zone thickness. These gelled fluids are described in more detail below.

Linear Gels

A substantial number of fracturing treatments are completed using thickened, water-based linear gels. The gelling agents used in these fracturing fluids are typically guar gum, guar derivatives such as hydroxypropylguar (HPG) and carboxymethylhydroxypropylguar (CMHPG), or cellulose derivatives such as carboxymethylguar or hydroxyethylcellulose (HEC). In general, these products are biodegradable. Guar is a polymeric substance derived from the seed of the guar plant

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(Ely, 1985). Guar gum, on its own, is non-toxic and, in fact, is a food-grade product commonly used to increase the viscosity and elasticity of foods such as ice cream.

To formulate a viscous fracturing gel, guar powder or concentrate is dissolved in a carrier fluid such as water or diesel fuel. Increased viscosity improves the ability of the fracturing fluid to transport proppant and decreases the need for more turbulent flow. Concentrations of guar gelling agents within fracturing fluids have decreased over the past several years. It was determined that reduced concentrations provide better and more complete fractures (Powell et al., 1999).

Diesel fuel has been frequently used in lieu of water to dissolve the guar powder because its carrying capacity per unit volume is much higher (Halliburton, Inc., 2002). "Diesel is a common solvent additive, especially in liquid gel concentrates, used by many service companies for continuous delivery of gelling agents in fracturing treatments" (GRI, 1996). Diesel does not enhance the efficiency of the fracturing fluid; it is merely a component of the delivery system (Halliburton, Inc., 2002). Using diesel instead of water minimizes the number of transport vehicles needed to carry the liquid gel to the site (Halliburton, Inc., 2002).

The percentage of diesel fuel in the slurried thickener can range between 30 percent and almost 100 percent, based on the MSDSs summarized in Table 4-1. Diesel fuel is a petroleum distillate and may contain known carcinogens. One such component of diesel fuel is benzene, which, according to literature sources, can make up anywhere between 0.003 percent and 0.1 percent by weight of diesel fuel (Clark and Brown, 1977; R. Morrison & Associates, Inc., 2001). Slurried diesel and gel are diluted with water prior to injection into the subsurface. The dilution is approximately 4 to 10 gallons of concentrated liquid gel (guar slurried in diesel) per 1,000 gallons of make-up water to produce an adequate polymer slurry (Halliburton, Inc., Virginia Site Visit, 2001; Schlumberger, Ltd., 2001; Consolidated Industrial Services, Inc., Virginia Site Visit, 2001; BJ Services, 2001).

Cross-linked Gels

One major advance in fracturing fluid technology was the development of cross-linked gels. The first cross-linked gels were developed in 1968 (Ely, 1985). When cross-linking agents are added to linear gels, the result is a complex, high-viscosity fracturing fluid that provides higher proppant transport performance than do linear gels (Messina, Inc. Web site, 2001; Ely, 1985; Halliburton Inc., Virginia Site Visit, 2001). Cross-linking reduces the need for fluid thickener and extends the viscous life of the fluid indefinitely. The fracturing fluid remains viscous until a breaking agent is introduced to break the cross-linker and, eventually, the polymer. Although cross-linkers make the fluid more expensive, they can considerably improve hydraulic fracturing performance, hence increasing coalbed methane well production rates.

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Cross-linked gels are typically metal ion-cross-linked guar (Ely, 1985). Service companies have used metal ions such as chromium, aluminum, titanium, and other metal ions to achieve cross-linking (Ely, 1985). In 1973, low-residue (cleaner) forms of cross-linked gels, such as cross-linked hydroxypropylguar, were developed (Ely, 1985).

According to MSDSs summarized in Table 4-1, cross-linked gels may contain boric acid, sodium tetraborate decahydrate, ethylene glycol, and monoethylamine. These constituents are hazardous in their undiluted form and can cause kidney, liver, heart, blood, and brain damage through prolonged or repeated exposure. According to a BLM environmental impact statement, cross-linkers may contain hazardous constituents such as ammonium chloride, potassium hydroxide, zirconium nitrate, and zirconium sulfate (U.S. Department of the Interior, CO State BLM, 1998). Concentrations of these compounds in the fracturing fluids were not reported in the impact statement. The final concentration of cross-linkers is typically 1 to 2 gallons of cross-linker per 1,000 gallons of gel (Halliburton, Inc., Virginia Site Visit, 2001; Schlumberger, Ltd., 2001).

4.2.2   Foamed Gels

Foam fracturing technology uses foam bubbles to transport and place proppant into fractures. The most widely used foam fracturing fluids employ nitrogen or carbon dioxide as their base gas. Incorporating inert gases with foaming agents and water reduces the amount of fracturing liquid required. Foamed gels use fracturing fluids with higher proppant concentrations to achieve highly effective fracturing. The gas bubbles in the foam fill voids that would otherwise be filled by fracturing fluid. The high concentrations of proppant allow for an approximately 75-percent reduction in the overall amount of fluid that would be necessary using a conventional linear or cross-linked gel (Ely, 1985; Halliburton, Inc., Virginia Site Visit, 2001). Foaming agents can be used in conjunction with gelled fluids to achieve an extremely effective fracturing fluid (Halliburton, Inc., Virginia Site Visit, 2001).

Foam emulsions experience high leakoff; therefore, typical protocol involves the addition of fluid-loss agents, such as fine sands (Ely, 1985; Halliburton, Virginia Site Visit, 2001). Foaming agents suspend air, nitrogen, or carbon dioxide within the aqueous phase of a fracturing treatment. The gas/liquid ratio determines if a fluid will be true foam or simply a gas-energized liquid (Ely, 1985). Carbon dioxide can be injected as a liquid, whereas nitrogen must be injected as a gas to prevent freezing (Halliburton, Inc., Virginia Site Visit, 2001).

According to the MSDSs summarized in Table 4-1, foaming agents can contain diethanolamine and alcohols such as isopropanol, ethanol, and 2-butoxyethanol. They can also contain hazardous substances including glycol ethers (U.S. Department of the Interior, CO State BLM, 1998). One of the foaming agent products listed in Table 4-1 can cause negative liver and kidney effects, although the actual component causing these effects is not specified on the MSDS. The final concentration is typically 3 gallons of

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foamer per 1,000 gallons of gel (Halliburton, Inc., Virginia Site Visit, 2001;
Schlumberger, Ltd., 2001).

4.2.3   Water & Potassium Chloride Water Treatments

Many service companies use groundwater pumped directly from the formation or treated
water for their fracturing jobs. In some coalbed methane well stimulations, proppants are
not needed to prop fractures open, so simple water or slightly thickened water can be a
cost-effective substitute for an expensive polymer or foam-based fracturing fluid with
proppant (Ely, 1985). Hydraulic fracturing performance is not exceptional with plain
water, but, in some cases, the production rates achieved are adequate. Plain water has a
lower viscosity than gelled water, which reduces proppant transport capacity.

Similar to plain water, another fracturing fluid uses water with potassium chloride (KCl)
in addition to small quantities of gelling agents, polymers, and/or surfactants (Ely, 1985).
Potassium chloride is harmless if ingested at low concentrations.

4.2.4   Acids

Acids are used in limestone formations that overlay or are interbedded within coals to
dissolve the rock and create a conduit through which formation water and coalbed
methane can travel (Ely, 1985). Typically, the acidic stimulation fluid is hydrochloric
acid or a combination of hydrochloric and acetic or formic acid. For acid fracturing to be
successful, thousands of gallons of acid must be pumped far into the formation to etch the
face of the fracture (Ely, 1985). Some of the cellulose derivatives used as gelling agents
in water and water/methanol fluids can be used in acidic fluids to increase treatment
distance (Ely, 1985). As discussed in section 4.2.5, acids may also be used as a
component of breaker fluids.

In addition, acid can be used to clean up perforations of the cement surrounding the well
casing prior to fracturing fluid injection (Halliburton, Inc., Virginia Site Visit, 2001;
Halliburton, Inc., 2002). The cement is perforated at the desired zone of injection to ease
fracturing fluid flow into the formation (Halliburton, Inc., Virginia Site Visit, 2001;
Halliburton, Inc., 2002).

Table 4-1 provides information on formic and hydrochloric acids. Acids are corrosive,
and can be extremely hazardous in concentrated form. Acids are substantially diluted
with water-based or water-and-gas-based fluids prior to injection into the subsurface.
The injected concentration is typically 1,000 times weaker than the concentrated versions
presented in the product MSDSs (Halliburton, Inc., Virginia Site Visit, 2001;
Schlumberger, Ltd., 2001).

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4.2.5   Fluid Additives

Several fluid additives have been developed to enhance the efficiency and increase the success of fracturing fluid treatments. The major categories of these additives are defined and briefly described in the following sections.

Breakers

Breaker fluids are used to degrade the fracturing fluid viscosity, which helps to enhance post-fracturing fluid recovery, or flowback. Breakers can be mixed with the fracturing fluid during pumping, or they can be introduced later as an independent fluid. There are a variety of breaker types including time-release and temperature-dependent types. Most breakers are typically acids, oxidizers, or enzymes (Messina, Inc. Web site, 2001). According to a BLM environmental impact statement, breakers may contain hazardous constituents, including ammonium persulfate, ammonium sulphate, copper compounds, ethylene glycol, and glycol ethers (U.S. Department of the Interior, CO State BLM, 1998). Concentrations of these compounds in the fracturing fluids were not presented in the environmental impact statement.

Biocides

One hydraulic fracturing design problem that arises when using organic polymers in fracturing fluids is the incidence of bacterial growth within the fluids. Due to the presence of organic constituents, the fracturing fluids provide a medium for bacterial growth. As the bacteria grow, they secrete enzymes that break down the gelling agent, which reduces the viscosity of the fracturing fluid. Reduced viscosity translates into poor proppant placement and poor fracturing performance. To alleviate this degradation in performance, biocides, bactericides, or microbicides are added to the mixing tanks with the polymeric gelling agents to kill any existing microorganisms (e.g., sulfate-reducing bacteria, slime-forming bacteria, algae), and to inhibit bacterial growth and deleterious enzyme production. Bactericides are typically hazardous by nature (Messina, Inc. Web site, 2001). They may contain hazardous constituents, including polycyclic organic matter (POM) and polynuclear aromatic hydrocarbons (PAHs) (U.S. Department of the Interior, CO State BLM, 1998).

Information from MSDSs for a biocide and a microbicide is summarized in Table 4-1. These concentrated products are substantially diluted prior to injection into the subsurface. Typical dilution in the make-up water is 0.1 to 0.2 gallons of microbicide in 1,000 gallons of water (Halliburton, Inc., Virginia Site Visit, 2001; Schlumberger, Ltd., 2001).

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Fluid-Loss Additives

Fluid-loss additives restrict leakoff of the fracturing fluid into the exposed rock at the fracture face. Because the additives prevent excessive leakoff, fracturing fluid effectiveness and integrity are maintained. Fluid-loss additives of the past and present include bridging materials such as 100 mesh sand, 100 mesh soluble resin, and silica flour, or plastering materials such as starch blends, talc silica flour, and clay (Ely, 1985).

Friction Reducers

To optimize the fracturing process, water-based fluids must be pumped at maximum rates and fluids must be injected at maximum pressures. Increasing flow velocities and pressures in this manner can lead to undesirable levels of friction within the injection well and the fracture itself. In order to minimize friction, friction reducers are added to water-based fracturing fluids. The friction reducers are typically latex polymers or copolymers of acrylamides. They are added to slick water treatments (water with solvent) at concentrations of 0.25 to 2.0 pounds per 1,000 gallons (Ely, 1985). Some examples of friction reducers are oil-soluble anionic liquid, cationic polyacrilate liquid, and cationic friction reducer (Messina, Inc. Web site, 2001).

Acid Corrosion Inhibitors

Corrosion inhibitors are required in acid fluid mixtures because acids will corrode steel tubing, well casings, tools, and tanks. The solvent acetone is a common additive in corrosion inhibitors (GRI, 1996). Information from MSDSs for acid inhibitors is summarized in Table 4-1. These products can affect the liver, kidney, heart, central nervous system, and lungs. They are quite hazardous in their undiluted form. These products are diluted to a concentration of 1 gallon per 1,000 gallons of make-up water and acid mixture (Halliburton, Inc., Virginia Site Visit, 2001; Schlumberger, Ltd., 2001). Acids and acid corrosion inhibitors are used in very small quantities in coalbed methane fracturing operations (500 to 2,000 gallons per treatment).

4.2.6   Proppants

The purpose of a proppant is to prop open a hydraulic fracture. An ideal proppant should produce maximum permeability in a fracture. Fracture permeability is a function of proppant grain roundness, proppant purity, and crush strength. Larger proppant volumes allow for wider fractures, which facilitate more rapid flowback to the production well. Over a period of 30 minutes, 4,500 to 15,000 gallons of fracturing fluid will typically transport and place approximately 11,000 to 25,000 pounds of proppant into the fracture (Powell et al., 1999).

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Table 4-1: Characteristics of Undiluted Chemicals Found in Hydraulic Fracturing Fluids (Based on MSDSs)

Product	Chemical Composition Information[1]	Hazards Information	Toxicological Information[2]	Ecological Information
Linear gel delivery system	1) 30-60% by wt. Guar gum derivative 2) 60-100% by wt. Diesel	• Harmful if swallowed • Combustible	• Chronic effects/Carcinogenicity – contains diesel, a petroleum distillate and known carcinogen • Causes eye, skin, respiratory irritation • Can cause skin disorders • Can be fatal if ingested	Slowly biodegradable
Water gelling agent	1) 60-100% by wt. Guar gum 2) 5-10% by wt. Water 3) 0.5-1.5% by wt. Fumaric acid	None	May be mildly irritating to eyes	Biodegradable
Linear gel polymer	1) <2% by wt. Fumaric acid 2) <2% by wt. Adipic acid	Flammable vapors	Can cause eye, skin and respiratory tract irritation	Not determined
Linear gel polymer slurry	1) 30-60% by wt. Diesel oil #2	• Causes irritation if swallowed • Flammable	• Carcinogenicity – Possible cancer hazard based on animal data; diesel is listed as a category 3 carcinogen in EC Annex I • May cause pain, redness, dermatitis	Partially biodegradable
Crosslinker	1) 10-30% by wt. Boric Acid 2) 10-30% by wt. Ethylene Glycol 3) 10-30% by wt. Monoethanolamine	• Harmful if swallowed • Combustible	• Chronic effects/Carcinogenicity D5 may cause liver, heart, brain reproductive system and kidney damage, birth defects (embryo and fetus toxicity) • Causes eye, skin, respiratory irritation • Can cause skin disorders and eye ailments	Not determined
Crosslinker	1) 10-30% by wt. Sodium tetraborate decahydrate	May be mildly irritating: • to eyes and skin • if swallowed	May be mildly irritating	• Partially biodegradable • Low fish toxicity
Foaming agent	1) 10-30% by wt. Isopropanol 2) 10-30% by wt. Salt of alkyl amines 3) 1-5% by wt. Diethanolamine	• Harmful if swallowed • Highly flammable	• Chronic effects/Carcinogenicity – may cause liver and kidney effects • Causes eye, skin, respiratory irritation • Can cause skin disorders and eye ailments	Not determined
Foaming agent	1) 10-30% by wt. Ethanol 2) 10-30% by wt. 2-Butoxyethanol 3) 25-55% by wt. Ester salt 4) 0.1-1% by wt. Polyglycol ether 5) 10-30% by wt. Water	Harmful if swallowed or absorbed through skin	• May cause nausea, headache, narcosis • May be mildly irritating	Harmful to aquatic organisms
Acid treatment - hydrochloric acid	1) 30-60% by wt. Hydrochloric acid	• May cause eye, skin and respiratory burns • Harmful if swallowed	• Chronic effects/Carcinogenicity – prolonged exposure can cause erosion of teeth • Causes severe burns, and skin disorders	Not determined
Acid treatment - formic acid	1) 85% by wt. Formic acid	• May cause mouth, throat, stomach, skin and respiratory tract burns • May cause genetic changes	• May cause heritable genetic damage in humans • Causes severe burns • Causes tissue damage	Not determined
Breaker Fluid	1) 60-100% by wt. Diammonium peroxidisulphate	• May cause respiratory tract, eye or skin irritation • Harmful if swallowed	May cause redness, discomfort, pain, coughing, dermatitis	Not determined

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Table 4-1: Characteristics of Undiluted Chemicals Found in Hydraulic Fracturing Fluids (Based on MSDSs)

Product	Chemical Composition Information[1]	Hazards Information	Toxicological Information[2]	Ecological Information
Microbicide	1) 60-100% by wt. 2-Bromo-2 nitro1,3-propanedol	• May cause eye and skin irritation	• Chronic effects/Carcinogenicity – not determined • Can cause permanent eye damage, skin disorders, abdominal pain, nausea, and diarrhea if ingested	Not determined
Biocide	1) 60-100% by wt. 2,2-Dibromo-3-nitrilopropionamide 2) 1-5% by wt. 2-Bromo-3-nitrilopropionamide	• Causes severe burns • Harmful if swallowed • May cause skin irritation; may cause allergic reaction upon repeated skin exposure	• Harmful if swallowed; large amounts may cause illness • Irritant; may cause pain or discomfort to mouth, throat, stomach; may cause pain, redness, dermatitis	Not determined
Acid corrosion inhibitor	1) 30-60% by wt. Methanol 2) 5-10% by wt. Propargyl alcohol	• May cause eye and skin irritation, headache, dizziness, blindness and central nervous system effects • May be fatal if swallowed • Flammable	• Chronic effects/Carcinogenicity – may cause eye, blood, lung, liver, kidney, heart, central nervous system and spleen damage • Causes severe eye, skin, respiratory irritation • Can cause skin disorders	Not determined
Acid corrosion inhibitor	1) 30-60% by wt. Pyridinium, 1-(Phenylmethyl)-, Ethyl methyl derivatives, Chlorides 2) 15% by wt. Thiourea 3) 5-10% Propan-2-ol 4) 1-5% Poly(oxy-1,2-ethanediyl)-nonylphenyl-hydroxy 5) 10-30% Water	• Cancer hazard (risk depends on duration and level of exposure) • Causes severe burns to respiratory tract, eyes, skin • Harmful if swallowed or absorbed through skin	• Carcinogenicity – Thiourea is known to cause cancer in animals, and possibly causes cancer in humans • Corrosive - short exposure can injure lungs, throat, and mucous membranes; can cause burns, pain, redness swelling and tissue damage	• Toxic to aquatic organisms • Partially biodegradeable

[1] Information presented is for the pure product, which is significantly diluted prior to injection. MSDS chemical composition percentages may total more than 100%.
[2] Toxicity is concentration dependant.

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4.3   The Fate and Transport of Stimulation Fluids Injected into Coal and Surrounding Rock During Hydraulic Fracturing of Coalbed Methane Reservoirs (with a Special Focus on Diesel Fuel)

Diesel fuel is sometimes a component of gelled fluids. Diesel fuel contains constituents of potential concern regulated under SDWA – benzene, toluene, ethylbenzene, and xylenes (i.e., BTEX compounds). The use of diesel fuel in fracturing fluids poses the greatest threat to USDWs because BTEX compounds in diesel fuel exceed the MCL at the point-of-injection (i.e. the subsurface location where fracturing fluids are initially injected).

The remainder of this section presents EPA's qualitative evaluation of the fate and transport of fracturing fluids injected into targeted coal layers in the subsurface during hydraulic fracturing. Although EPA's MOA with the three major service companies has largely eliminated diesel fuel from fracturing fluids injected directly into any USDWs, there may still be rare instances in which diesel fuel is used by other service companies or operators (USEPA, 2003). Therefore an evaluation of the use of diesel fuel in fracturing fluid, which also provides follow-up on the draft of this report published in August, 2002, is included in this chapter.

EPA revised its procedure for assessing the potential effects of fracturing fluid constituents on USDWs from the procedure presented in the August 2002 draft of this report as follows:

- EPA has revised the fraction of BTEX compounds in diesel used to estimate the point-of-injection concentrations from a single value to a documented broader range of values for the fraction of BTEX in diesel fuel. For example, the fraction of benzene in diesel was revised from $0.00006 g_{benzene}/g_{diesel}$ to a range with a minimum value of $0.000026\ g_{benzene}/g_{diesel}$ and a maximum value of $0.001\ g_{benzene}/g_{diesel}$. If the maximum value for benzene in diesel is used to estimate the concentration of benzene at the point-of-injection, the resulting estimate is 17 times higher than that presented in the Draft Report.

- In this report, EPA used more current values for two of the parameters used to estimate the point-of-injection concentrations of BTEX compounds. Specifically, the estimates in this report use a density of the diesel fuel-gel mixture of 0.87 g/mL compared to 0.84 g/mL in the Draft Report, and a fraction of diesel fuel in gel of $0.60\ g_{diesel}/g_{gel}$ compared to $0.52\ g_{diesel}/g_{gel}$ in the Draft Report. The use of these more current values does not affect the order of magnitude of the revised point-of-injection calculations.

- The August 2002 Draft Report included estimates of the concentration of benzene at an idealized, hypothetical edge of the fracture zone located 100 feet from the point-of-injection. Based on new information and stakeholder input, EPA concluded that the edge of fracture zone calculation is not an appropriate model for reasons including:

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- Mined-through studies reviewed by EPA indicated that hydraulic fracturing injection fluids had traveled several hundred feet beyond the point-of-injection.

- The assumption of well-mixed concentrations within the idealized fracture zone is insufficient. One mined-through study indicated an observed concentration of gel in a fracture that was 15 times the injected concentration, with gel found to be hanging in stringy clumps in many fractures. The variability in gel distribution in hydraulic fractures indicates that the gel constituents are unlikely to be well mixed in groundwater.

- Based on more extensive review of the literature, the width of a typical fracture was estimated to be much thinner than that used in the Draft Report (0.1 inch versus 2 inches). The impact of the reduced width of a typical fracture is that the calculated volume of fluid that can fit within a fracture is less. After an initial volume calculation using the new width, EPA found that the volume of the space within the fracture area may not hold the volume of fluid pumped into the ground during a typical fracturing event. Therefore, EPA assumes that a greater volume of fracturing fluid must "leakoff" to intersecting smaller fractures than what was assumed in the Draft Report, or that fluid may move beyond the idealized, hypothetical "edge of fracture zone." This assumption is supported by field observations in mined-through studies, which indicate that fracturing fluids often take a stair-step transport path through the natural fracture system.

- In the Draft Report, EPA approximated the edge of fracture zone concentrations considering only dilution. Based on new information and stakeholder input on the Draft Report, EPA does not provide estimates of concentrations beyond the point-of-injection in the final report. Developing such concentration values with the precision required to compare them to MCLs would require the collection of significant amounts of site-specific data. This data in turn would be used to perform a formal risk assessment, considering numerous fate and transport scenarios. These activities are beyond the scope of this Phase I study.

The remainder of this section includes a discussion of the following components of EPA's analysis:

- The concentrations of BTEX at the point-of-injection.

- The percentage of fracturing fluids recovered during the recovery process.

- The influence of the capture zone.

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- Factors that would increase or decrease the concentrations of BTEX remaining in the subsurface.

The first step in EPA's analysis of the potential threat to USDWs from the injection of fracturing fluids was calculating the point-of-injection concentrations of BTEX introduced from diesel fuel in the gelling agent. In Step 2, EPA considered factors that affect the degree to which hydraulic fracturing fluids are recovered. Steps 3, 4, and 5 provide analyses of physical/chemical, hydrogeological, and biological processes that could affect the fate and transport of hazardous chemicals introduced into coal seams. These steps are summarized in Table 4-2.

4.3.1   Point-of-Injection Calculation

The formulations or "recipes" for fracturing fluids differ among service companies and among sites; the amount of fracturing fluid used will also vary. Thus, a range of point-of-injection concentrations likely exists. According to field paperwork obtained during EPA's site visits (Consolidated Industrial Services, Inc., 2001; Halliburton, 2001) and information provided by three service company scientists (BJ Services, 2001; Halliburton, 2001; Schlumberger, Ltd., 2001), between 4 and 10 gallons of diesel-containing gelling agent are added to each 1,000 gallons of water used in hydraulic fracturing, when diesel is used. In addition, the fraction of BTEX in diesel may range by up to two orders of magnitude (Potter and Simmons, 1998). The lower and upper ranges of the values presented in Potter and Simmons (1998), as well as the three different values cited for gelling agent, were used to estimate point-of-injection concentrations for each of three fracturing fluid recipes (i.e., the ratio of fracturing gel to water). The resulting 24 point-of-injection calculations are provided in Table 4-2. These estimates provide the basis for a qualitative assessment regarding whether a Phase II study is warranted.

The following example illustrates how EPA estimated the concentrations of BTEX at the point-of-injection. Due to the variations in the recipe used by service companies, EPA's analysis begins with three different possible scenarios, as follows:

- Low ratio: 4 gallons of gel per 1,000 gallons of water

- Medium ratio: 6 gallons of gel per 1,000 gallons of water

- High ratio: 10 gallons of gel per 1,000 gallons of water

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The concentration of benzene in fracturing fluid at the point-of-injection ([benzene]$_{inj}$) can be calculated using the following equation:

$$[benzene]_{inj} = (r_{gw}) \times (\rho_{dg}) \times (f_{dg}) \times (f_{bd}) \times (3{,}785 \text{ mL}_{gel}/\text{gal}_{gel}) \times (1 \text{ gal}_{water}/3.785 \text{ L}_{water}) \times (10^6 \ \mu g/g)$$

Where:

$r_{gw} =$ the ratio of diesel fuel-gel mixture to injection water (gal$_{gel}$/1,000 gal$_{water}$)
(4 gal$_{gel}$/1,000gal$_{water}$, 6gal$_{gel}$/1,000 gal$_{water}$, and 10 gal$_{gel}$/1,000 gal$_{water}$ represent the low, medium, and high ratios, respectively)

$\rho_{dg} =$ the density of the diesel fuel-gel mixture (g$_{gel}$/mL$_{gel}$) = 0.84 g$_{gel}$/mL$_{gel}$ (Halliburton, 2002)

$f_{dg} =$ the fraction of diesel fuel in the gel (g$_{diesel}$/g$_{gel}$) = 0.52 g$_{diesel}$/g$_{gel}$ (Halliburton, 2002)

$f_{bd} =$ the fraction of benzene in diesel fuel (g$_{benzene}$/g$_{diesel}$) = 0.000026 to 0.001 g$_{benzene}$/g$_{diesel}$ (Potter and Simmons, 1998)

3,785 mL$_{gel}$/gal$_{gel}$ =   volume conversion factor

1 gal$_{water}$/3.785 L$_{water}$ =   volume conversion factor

$10^6 \ \mu g/g$ =   mass conversion factor

The concentration of benzene at the point-of-injection is calculated for the three gel/water ratios and the minimum and maximum concentrations of benzene in diesel fuel.

Using r_{gw} = 4 gal$_{gel}$/1,000gal$_{water}$ and f_{bd} = 0.000026 g$_{benzene}$/g$_{diesel}$ as an example, [benzene]$_{inj}$ is calculated as follows:

$$[benzene]_{inj} = (4 \text{ gal}_{gel}/1{,}000\text{gal}_{water}) \times (0.84 \text{ g}_{gel}/\text{mL}_{gel}) \times (0.52 \text{ g}_{diesel}/\text{g}_{gel}) \times$$
$$(0.000026 \text{ g}_{benzene}/\text{g}_{diesel}) \times (3{,}785 \text{ mL}_{gel}/\text{gal}_{gel}) \times (1 \text{ gal}_{water}/3.785 \text{ L}_{water}) \times (1{,}000 \text{ mL/L}) \times (10^6 \ \mu g/g) = 45 \ \mu g/L$$

Table 4-2 summarizes the estimated injection concentrations of each BTEX constituent for the three assumed gel/water ratios and the minimum and maximum concentrations of BTEX in diesel fuel. It also presents the MCL for each compound. Many of the estimated concentrations of BTEX exceed the MCL at the point-of-injection.

Table 4-2 and the remainder of this section provide a qualitative assessment of the fate and transport processes that could attenuate the concentrations of BTEX in groundwater. Factors that would influence the availability of constituents of potential concern in fracturing fluids and decrease their concentrations include:

- Fluid Recovery - much of the fluid is eventually pumped back to the surface.

- Adsorption and entrapment - some of these constituents will undergo adsorption to the coal or become entrapped in the formation.

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- Biodegradation - some fracturing fluid constituents, such as benzene, may undergo partial biodegradation.

4.3.2   Fracturing Fluid Recovery

Following the injection of fracturing fluids into the subsurface through coalbed methane wells (i.e., production wells), considerable amounts of the fracturing fluids are removed. During the recovery process, the injected fluids and ambient groundwater are pumped out of the formation through the production well to reduce formation pressure, enabling methane desorption and extraction. Palmer et al. (1991a) found that 61 percent of fracturing fluids were recovered based on samples collected from coalbed methane wells over a 19-day period. Their study predicted total recovery to be between 68 and 82 percent.

Palmer et al. (1991a) also discussed the possibility that a "check-valve effect" could trap some of the fracturing fluid on one side (i.e., upgradient, during production) of a collapsed or narrowed fracture, preventing the fluid from flowing back to the production well. This check-valve effect can occur in both natural and induced fractures when the fractures narrow again after the injection of fracturing fluid ceases, formation pressure decreases, and extraction of methane and groundwater begins.

Another factor preventing full recovery of injected fluids is the high injection pressure used during hydraulic fracturing operations. Fracturing fluids are forced into the subsurface under high pressure to enlarge and propagate existing fractures. The hydraulic gradients that cause fluids to flow away from the well during injection are much greater than the hydraulic gradients that occur during fluid recovery. As a result, some of the fracturing fluids will travel beyond the capture zone of the production well. The capture zone of a production well is the portion of the aquifer that contributes water to the well. The size of this zone will be affected by regional groundwater gradients, and by the drawdown caused by the well (USEPA, 1987). Fluids that flow beyond the capture zone of the production well generally are not recovered during the flowback process.

Gel contained in fracturing fluids may be unrecovered because its properties differ from that of water and highly soluble constituents of fracturing fluids. One mined-through study reviewed by EPA described evidence of gel clumps within many fractures (Steidl, 1993). One observed concentration of gel in a fracture was 15 times the injected concentration. When the fluids exist as undissolved gel, they may remain attached to the sides of the fractures or be trapped within smaller fractures or pores present in formations that surround the coalbed. The mined-through studies suggest that such fluids are unlikely to flow with groundwater during production, but they may present a source of gel constituents to flowing groundwater subsequent to fluid recovery. Fate and transport processes discussed later in this section can serve to reduce gel constituent concentrations that may result from trapped fluids. Mechanisms that may affect the recovery of fracturing fluids are discussed in section 3.3.2 of Chapter 3.

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4.3.3   The Influence of the Capture Zone

The recovery process typically lasts approximately 10-20 years. During that time, groundwater within the production well's capture zone flows toward the production well. Assuming complete mixing, the predicted recovery of injected BTEX is between 68 and 82 percent (Palmer et al., 1991a). Thus, between 20 and 30 percent of the BTEX injected is expected to remain in the formation. It is reasonable to expect that most of the unrecovered fluid lies outside the capture zone and that the residual concentrations of BTEX within the capture zone are substantially less than the injection concentrations. Chemicals such as BTEX that are not recovered from within the capture zone during groundwater production may be diluted by groundwater that flows into the formation to replace production water. Additional attenuation from sorption and biodegradation may occur. Subsequent to production, dispersion and diffusion may serve to reduce residual BTEX concentrations. The fracturing fluids that flow beyond the capture zone are affected by regional groundwater flow and may be diluted by groundwater.

4.3.4   Fate and Transport Considerations

BTEX that has moved beyond the production well's capture zone is of the greatest concern. The fate and transport mechanisms that may affect BTEX concentrations beyond the capture zone are evaluated in this section. Factors that would likely decrease exposure concentrations and/or availability of BTEX include attenuation through groundwater flow dynamics, biological processes, and adsorption.

BTEX outside of the capture zone will likely be transported by groundwater flowing according to regional hydraulic gradients. This flow and transport are not influenced by production pumping. Nevertheless, mechanical dispersion will cause BTEX to spread horizontally and vertically in the aquifer, thereby reducing the concentrations. The degree of mechanical dispersion depends in part on the velocity of flow and increases with increased travel distance. Dilution can have a significant effect on the BTEX concentrations that could migrate to drinking water wells, especially if these wells are hundreds to thousands of feet from a hydraulically induced fracture. The process of molecular diffusion (i.e., the movement of BTEX from areas of higher to lower concentration due to the concentration differences) will further reduce BTEX concentrations. Collectively, mechanical dispersion and molecular diffusion are referred to as hydrodynamic dispersion (Fetter, 1994).

The biodegradation of diesel fuel constituents, including BTEX, has been studied in other geologic settings and laboratory studies and may lead to reductions in concentrations in coalbeds given the appropriate site conditions. No information was found about the occurrence of biodegradation or biodegradation rates of BTEX in coalbeds or surrounding rock. In order for biodegradation to occur, organisms capable of using BTEX as a food source must be present and conditions such as favorable pH, salinity, and sometimes the availability of oxygen, nitrogen, and phosphorous must be met to ensure bacterial survival. Generally, substantial benzene degradation occurs in aerobic environments. The levels of oxygen in a particular formation vary widely depending primarily on the depth of coalbeds

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from the surface. Data regarding biodegradation of benzene in an anaerobic environment indicates a range from no degradation to relatively slow degradation (USEPA, 1999).

As groundwater flows through a formation, chemicals such as BTEX may be retarded by adsorption. Although adsorption in coalbeds is likely, quantification of adsorption is difficult in the absence of laboratory or site-specific studies (due to competition for adsorption between BTEX and more lipophilic and less soluble constituents of diesel fuel and coal, and fracture thickness). Other processes, such as desorption of BTEX from the coal surface, and dissolution of BTEX from the gel phase may play a role in BTEX transport. Entrapment of gel in pore spaces and fractures may also influence the degree to which BTEX is available to groundwater. In some cases, the gel may be entrapped in such a way that it is neither available to flow back towards the production well nor flow towards a USDW in response to regional groundwater gradients.

According to the information listed on MSDSs provided to EPA, several of the constituents of potential concern listed in Table 4-1 can have toxic effects when people are exposed to sufficiently high concentrations through the susceptible route(s) of exposure (i.e., inhalation, ingestion, skin contact). However, only the BTEX compounds originating from diesel fuel are regulated under SDWA. None of the other constituents in Table 4-1 appear on the Agency's draft Contaminant Candidate List (CCL). The drinking water CCL is the primary source of priority contaminants for evaluation by EPA's drinking water program. Contaminants on the CCL are known or anticipated to occur in public water systems and may require regulations under SDWA. Information on the GSA study is available at http://www.epa.gov/fedrgstr/EPA-WATER/2004/April/Day-02/w7416.htm**.**

Further, EPA does not believe that the other Table 4-1 constituents potentially contained in fracturing fluids are introduced through coalbed methane fracturing in concentrations high enough to pose a significant threat to USDWs. First, it is EPA's understanding, based on conversations with field engineers and on witnessing three separate fracturing events, that fracturing fluids used for coalbed methane fracturing do not contain most of the constituents listed in Table 4-1. Second, if the Table 4-1 constituents were used, EPA believes some of the same hydrodynamic phenomena listed in steps 2 and 4 (flowback, dilution and dispersion), step 3 (adsorption and entrapment), and potentially step 5 (biodegradation) would minimize the possibility that chemicals included in the fracturing fluids would adversely affect USDWs.

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Table 4-2: Estimated Concentrations of Diesel Contaminants in Fracturing Fluids at the Point-of-Injection and Factors Affecting Their Concentration and Movement in Groundwater

| | | Point-of-Injection (POI) Calculation[1] | | | | Factors Affecting the Concentration and Movement of Fluids in Groundwater | | | |
| | | Step 1 | | | | Step 2 | Step 3 | Step 4 | Step 5 |
Recipe (gallons of gel per 1,000 gallons of water)	Chemical (MCL µg/L)	Fraction of Chemical in Diesel — Min.	Fraction of Chemical in Diesel — Max.	Point-of-Injection Concentration (µg/L)[2] — Min.	Point-of-Injection Concentration (µg/L)[2] — Max.	Influence of Pumping Well Within the Capture Zone	Adsorption and Entrapment	Dilution and Dispersion	Biological Degradation
4	Benzene 5	0.000026	0.001	45	1,700	Movement of groundwater will be toward production well during its operation (about 10 to 20 years); assumes fluid recovered during pumping to be between 68% and 82% (Palmer et al., 1991a).	Adsorption likely to occur in coalbeds; dependent on width of fractures and flow or diffusion into coal matrix. Desorption may act as a continuing source of BTEX to groundwater. Adsorption less likely in surrounding strata such as shale and sandstone. Entrapment of gel may reduce the availability of BTEX to the surrounding groundwater.	Unrecovered BTEX that may be in pore spaces of formation(s) can be diluted by flow of clean groundwater through pore spaces or it may diffuse from pore spaces. Dilution may significantly reduce BTEX concentrations available to drinking water wells, especially when they are great distances from the hydraulic fracture.	If indigenous microorganisms at the site are capable of anaerobic degradation of BTEX, partial biodegradation may be a relevant attenuation mechanism (Haener, et al., 1995; Hess et al., 1997; Aronson and Howard, 1997; Tan et al., 2002).
6				68	2,600				
10				110	4,400				
4	Ethyl benzene 700	0.00007	0.002	120	3,500				
6				180	5,200				
10				310	8,700				
4	Toluene 1,000	0.000069	0.007	120	12,000				
6				180	18,000				
10				300	31,000				
4	Xylenes 10,000	0.00019[3]	0.006[3]	330	10,000				
6				500	15,000				
10				830	26,000				

[1] Using benzene as an example: $[\text{benzene}]_{poi} = (r_{gw}) \times (r_{dg}) \times (f_{dg}) \times (f_{bd}) \times (3{,}785\ mL_{gel}/gal_{gel}) \times (1\ gal_{water}/3.785\ L_{water}) \times (10^6\ ng/g)$, where:
r_{gw} = the ratio of gel to injection water $(gal_{gel}/1{,}000\ gal_{water})$ = 4 $gal_{gel}/1{,}000gal_{water}$, 6$gal_{gel}/1{,}000\ gal_{water}$ and 10 $gal_{gel}/1{,}000\ gal_{water}$
r_{dg} = the density of the diesel fuel-gel mixture (g_{gel}/mL_{gel}) = 0.84 g_{gel}/mL_{gel} (Halliburton, 2002)
f_{dg} = the fraction of diesel fuel in the gel (g_{diesel}/g_{gel}) = 0.52 g_{diesel}/g_{gel} (Halliburton, 2002)
f_{bd} = the fraction of benzene in diesel fuel $(g_{benzene}/g_{diesel})$ = 0.000026 to 0.001 $g_{benzene}/g_{diesel}$

Conversion factors: 3,785 mL_{gel}/gal_{gel} = volume conversion factor; 1 $gal_{water}/3.785\ L_{water}$ = volume conversion factor; $10^6\ \mu g/g$ = mass conversion factor.
[2] Values are rounded to two significant digits.
[3] Represents sum of meta-/para- and ortho-isomer analyses, since only one result was reported by Potter and Simmons for total xylenes.

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4.4   Summary

Fracture engineers select fracturing fluids based on site-specific characteristics including formation geology, field production characteristics, and economics. Hydraulic fracturing operations vary widely in the types of fracturing fluids used, the volumes of fluid required, and the pump rates at which they are injected. Based on the information EPA collected, water or nitrogen foam frequently constitutes the solute in fracturing fluids used for coalbed methane stimulation. Other components of fracturing fluids used to stimulate coalbed methane wells may contain only benign ingredients, but in some cases, they contain constituents such as diesel fuel that can be hazardous in their undiluted forms. Fracturing fluids are significantly diluted prior to injection.

Water with a simple sand proppant can be adequate to achieve a desired fracture at some sites. In some cases, water must be thickened to achieve higher proppant transport capabilities. Thickening can be achieved by using linear or cross-linked gelling agents. Cross-linkers are costly additives compared to simple linear gels, but a fluid's fracturing efficiency can be greatly improved using cross-linkers. Foam fracturing fluids can be used to considerably reduce the amount of injected fluid required. The reduced water volume requirement translates into a space and cost savings at the treatment site because fewer water tanks are needed. Foam fracturing fluids also promote rapid flowback and reduced volumes of flowback water requiring disposal.

The use of diesel fuel in fracturing fluids poses the greatest potential threat to USDWs because the BTEX constituents in diesel fuel exceed the MCL at the point-of-injection. Given the concerns with the use of diesel fuel, EPA recently entered into agreements with three major service companies to eliminate diesel fuel from hydraulic fracturing fluids injected directly into USDWs to stimulate coalbed methane production. Industry representatives estimate that these three companies perform approximately 95 percent of the hydraulic fracturing projects in the United States.

In situations when diesel fuel is used in fracturing fluids, a number of factors would decrease the concentration and/or availability of BTEX. These factors include fluid recovery during flowback, adsorption, dilution and dispersion, and potentially biodegradation of constituents. For example, Palmer et al. (1991a) documented that only about one-third of fracturing fluid that is injected is expected to remain in the formation. EPA expects fate and transport considerations would minimize the possibility that chemicals included in fracturing fluids would adversely affect USDWs.

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Figures 4-1 and 4-2. Liquid nitrogen tanker trucks transport gas to the site for nitrogen foam fracturing. Nitrogen will travel through pipes to be mixed with water and a foaming agent at the wellhead prior to injection. The foam is used to create and propagate the fracture deep within the targeted coal seam.

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Figures 4-3 and 4-4.
Chemicals are stored on site in a support truck. Fracturing fluid additives such as the foaming agent can be pumped directly from storage containers to mix tanks.

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Figure 4-5.
The fracturing fluid (water with additives) is stored on site in large, upright storage tanks. Each tank contains mix water imported from off-site, or formation water extracted directly from the gas well.

Figure 4-6.
Gelled water is pre-mixed in a truck-mounted mixing tank. Photograph shows a batch of linear, guar-based gel. This gel is used to transport the sand proppant into the fracture propagated by the nitrogen foam treatment.

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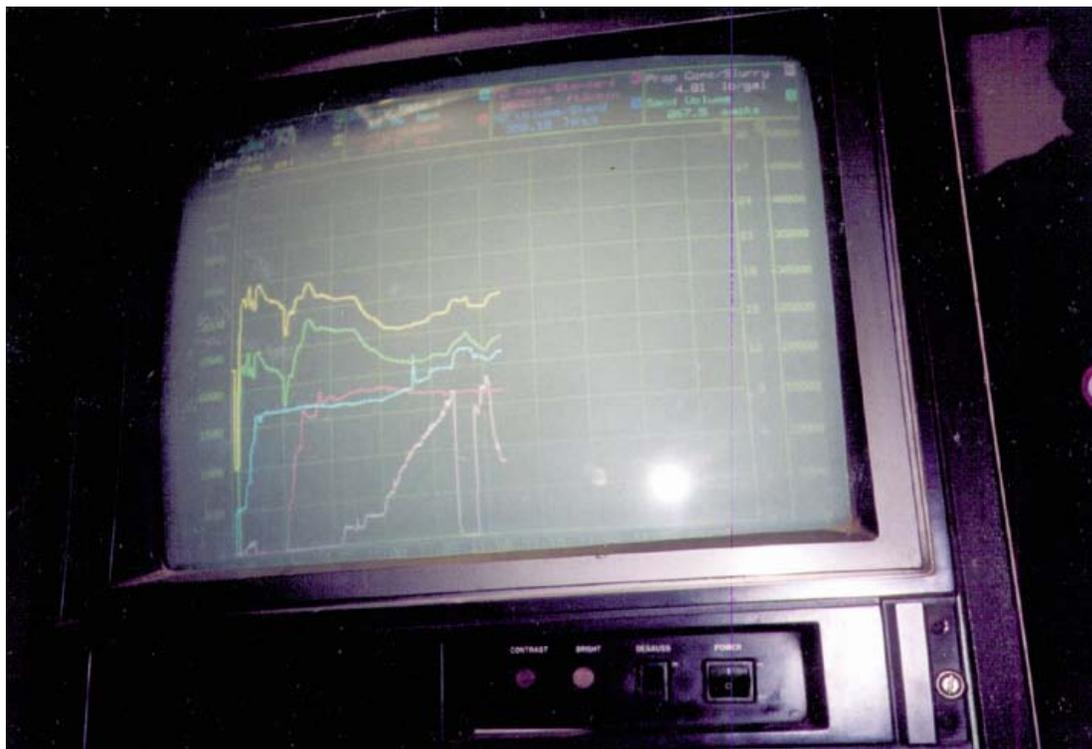Coalbed Methane Reservoirs

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Figure 4-7.
The fracturing fluids, additives, and proppant are pumped to the wellhead and mixed just prior to injection. The flow rate of each injected component is monitored carefully from an on-site control center.

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Figures 4-8 and 4-9.
Electronic monitoring systems provide constant feedback to the service company's operators.
Fluid flow rates and pressure buildup within the formation are monitored to ensure that fracture
growth is safe and controlled.

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Figures 4-10 and 4-11. Fluid that is extracted from the well is sprayed through a diffuser and stored in a lined trench until it is disposed of off-site or discharged.

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