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The geochemistry of hydraulic fracturing fluids

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Abstract

The inorganic geochemistry of hydraulic fracturing fluids is reviewed with new insights on the role of entrapped formation waters in unconventional shale gas and tight sand formations on the quality of flowback and produced waters that are extracted with hydrocarbons. The rapid increase of the salinity of flowback fluids during production, combined with geochemical and isotopic changes, indicate mixing of the highly saline formation water with the injected water. The salinity increase suggests that the volume of the injected water that is returned to the surface with the flowback water is much smaller than previous estimates, and thus the majority of the injected water is retained within the shale formations. The high salinity of the flowback and produced water is associated with high concentrations of halides, ammonium, metals, metalloids, and radium nuclides that pose environmental and human health risks upon the release of the hydraulic fracturing fluids to the environment.

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1. Introduction

During the last decade the rapid rise of unconventional shale gas and tight sand oil development through horizontal drilling and high volume hydraulic fracturing has expanded the extraction of hydrocarbon resources in the U.S., Canada, South America, and soon more broadly in China and other parts of the world. Nonetheless, the rapid development of unconventional energy extraction has triggered an intense public debate regarding the potential environmental and human health impacts from hydraulic fracturing, resulting

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in banning of hydraulic fracturing in some U.S. states (e.g., New York) and several countries in Europe (e.g., France). The environmental concerns include fugitive emissions of methane to the atmosphere and contamination of water resources[1,2]. Previous research has shown that hydraulic fracturing can impact water resources through: (1) contamination of shallow aquifers by fugitive hydrocarbon gases (i.e., stray gas contamination)[3-5], (2) contamination of surface water and shallow groundwater from spills, leaks, and/or the disposal of inadequately treated oil and gas wastewater[6,7], (3)accumulation of toxic and radioactive elements in soil or stream sediments near disposal or spill sites[6,7], and (4) the over-extraction of water resources for high-volume hydraulic fracturing, particularly in water-scarce areas[8,9].

2. Results and Discussion

2.1. Volume of flowback and produced waters

The production volume of flowback fluids vary among the different unconventional plays, and typically follow the oil and gas production rates. In the U.S., the typical volume of flowback and produced waters vary from 5.2×10^6 to 25.9×10^6 L per shale gas well and 8×10^6 to 22.8×10^6 L for unconventional oil well[8]. Parallel to the sharp decrease in the production rates in time of hydrocarbons from unconventional oil and gas wells, the flow rates of flowback and produced waters decrease by 2- to 10-fold[2,8]. Over the lifetime of production an unconventional oil or gas well (up to ~10 years to date), the accumulated volume of produced water will become much more significant relative to the short-term and high production rates of flowback water[8]. Based on available data, we estimate that flowback water constitutes only 5 to 10% of the total wastewater that is generated from a shale-gas well during the well’s decade-long production of hydrocarbon extraction. By comparison to flowback water, produced water dominates the volume of water accumulated at the surface and based on the chemical constituents of these fluids, produced waters may have a higher potential to affect the environment over the lifetime of unconventional oil and gas wells.

2.2. Sources of flowback and produced water from unconventional oil and gas wells

Most of the injected water that is used for hydraulic fracturing is retained within the shale or the tight sand formations and thus the volume of the returned (flowback) water is significantly lower than the volume of the injected water[8,10-14]. Data from the Marcellus Shale indicate that only 25% of the injected hydraulic fracturing fluids are returned to the surface as flowback water over 90 days following hydraulic fracturing[10]. The flowback water is characterized by a rapid change in chemistry, with a typically fast increase in salinity during the first few days (Figure 1A)[10,14-16]. The cause for the rise of the salinity of flowback water is debated and three major explanations have been proposed: (1) dissolution of halite and other salts in the shale formation[17]; (2) imbibition of the injected water to the shale formation and diffusional osmosis of ions from the shale to the flowback water[11,13]; and (3) imbibition of the injected water to the shale rocks and exchange with evaporated paleoseawater entrapped in the shale formation complex[14,16,18,19]. This evidence includes (1) the similarity of the chemical composition of Marcellus brines to the composition of evaporated seawater (e.g., high Br/Cl, low Na/Cl)[15,16], (2) the similarity of the Marcellus brines to the composition of formation waters from other geological formations in the Appalachian Basin, (3) the increase of $\delta^{18}O$ with salinity[14] (Figure 1B), which suggests mixing between injected water with low $\delta^{18}O$ and saline end-member with high $\delta^{18}O$, and (4) the difference in the $^{87}Sr/^{86}Sr$ ratios of exchangeable Sr from “dry” shale relative to the Marcellus produced water[18,19]. Consequently, the rise in salinity reflects mixing between the injected water (fresh water or recycled oil and gas wastewater) and the formation water entrapped in the shale.

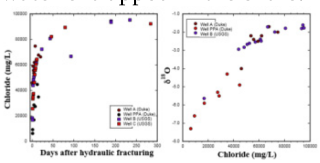

Fig. 1. (a) Changes in salinity (chloride content) of flowback water following hydraulic fracturing in 4 wells from the Marcellus Formation; (b) Correlation between stable isotopes of oxygen and chloride contents in flowback waters from the Marcellus Formation. Data from Duke University and USGS[14].