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Characterizing the variability in chemical composition of flowback water – results from laboratory studies

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Abstract

The large volumes and unknown composition of flowback and produced waters cause public concerns about the environmental and social compatibility of hydraulic fracturing and the exploitation of unconventional gas. Flowback and produced waters contain not only residues of fracking additives but also chemical species that are dissolved from the shales. Interactions of different shales with an artificial fracturing fluid were studied in lab experiments under ambient and elevated temperature and pressure conditions. Fluid-rock interactions change the chemical composition of the fracturing fluid and this indicates that geochemistry of the fractured shale needs to be considered to understand flowback water composition.

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1. Introduction

The growing importance of exploring gas and oil from unconventional reservoirs by hydraulic fracturing (HF) raises public concerns about the potential impacts on human health and the environment. Beside others, concerns are related to the high water demand for this technique as well as the application of tons of chemicals in the applied

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fracturing fluids and the composition of the resulting flowback and produced waters. On the one hand, provision of these huge amounts of water may provide problems to natural environments and drinking water supplies but on the other hand fracturing fluid and flowback water with unknown composition are feared to be contaminants and handling and proper treatment of these fluids are still under debate.

HF is a stimulation technique used to increase production of oil and gas and involves the injection of fluids under pressures great enough to fracture the oil- and gas-containing formations [1]. With HF, permeability in shales, tight sands, coal-beds, and other gas and oil-bearing strata is increased [2]. HF is used in conventional oil and gas reservoirs, but also to develop geothermal energy resources and unconventional oil and gas reservoirs. HF has been used since the late 1940s and wells stimulated by HF may be vertical, deviated, or horizontal in orientation, and they may be newly drilled or older at the time the fracturing is done [1]. The fracturing fluids generally consist of three parts: 1) the base fluid (i.e. water), 2) the additives and 3) the proppant. Each additive is a mixture of various chemicals with the main ingredient serving a specific purpose during HF (e.g. friction reducer, gelling agent, crosslinker, breakers, biocide, stabilizer) [3,4]. The particular composition of the fracturing fluid is selected by a design engineer based on empirical experience, the geological setting, reservoir geochemistry, economics, availability of chemicals and preference of the service company or operator [1]. The largest constituent of a typical fracturing fluid is water (>90%), followed by proppants (<10%) and additives (0.5 – 2%). After the formation has been fractured the pressure is released from the well which causes the fluid mixture to flowback to the surface. This fluid is generally classified as either flowback or produced water [2]. Flowback water is commonly defined as the water that is released within the initial two weeks following the completion of the HF process [5]. Produced water, however, is the naturally occurring water within the shale formation [2].

Composition of flowback is related to the composition of the initial fracturing fluid, the composition of the natural formation water of the shale and the possible interactions between fracturing fluid and shale system over time at the in-situ conditions. Initially this water is mostly fracturing fluid, but with time, it becomes more similar to the natural formation water, e.g. increase in salinity, and decrease in dissolved organic carbon (DOC) [6]. Concerning composition of flowback water, inorganic constituents (metals, salts), organic compounds (hydrocarbons, organic acids) and naturally occurring radioactive material (NORM) have to be considered. Therefore, to understand controls on the flowback water composition, composition of the initial fracturing fluids, natural formation waters and possible processes relevant in the reservoir need to be clarified.

To date, most publications on composition of flowback and produced water deal with samples from the Marcellus shale gas exploitation (see for summary [5]), only few publications exist from other shale gas sites in the US, Canada, China or Europe. Until now, the influence of the shale formation on the flowback water composition is quite often mentioned but not evaluated. Here, also experimental studies are necessary to assess the potential of the respective shale to release organic and inorganic constituents into the flowback water. Based on the extraction results of two European shale samples with an artificial fracturing fluid it became obvious that the flowback shows shale-specific characteristics in both, inorganic and organic composition [7]. In addition to this, it was shown that the thermal maturity of the individual shale sample has a strong influence on the extracted organic compounds [8]. Other laboratory experiments showed that the majority of reactions took place in the first 48 to 72 hours. Ca and Sr were released from the shale, indicating carbonate dissolution and the increase of metals such as Al, Cd, Co, Cr and Ni suggests the formation of secondary minerals [9]. Flowback was also reproduced and analyzed by Labus and coworkers [10]. The autoclave experiments implied that the composition of flowback water is more controlled by interactions of fracturing fluids and original pore water than by the properties of the reservoir rock. The fluid-rock interaction was based on carbonate and silicate dissolution and the oxidation of pyrite leading to an increase of sulfate. The initial high increase in the first 2 to 3 days was explained with a release of pore water.

The mobility of elements is strongly dependent on pH values, redox-conditions and only to a lower degree on temperature and solid:water ratios [7,9,11]. This was explored with batch experiments using core samples from the Bakken Formation (US). Similar experiments were done by Wang and coworkers using samples from the Eagle Ford Formation [12]. The pH value varies with the dissolution of carbonate and the oxidation of pyrite, therefore making it dependent on the chemistry of the reservoir rock. The interaction of reservoir rock and glutaraldehyde, a commonly used biocide additive in fracturing fluids, was investigated in lab experiments. Under high temperature and pressure conditions a rapid autopolymerisation of glutaraldehyde forming water-soluble dimers and trimers as well as precipitation at high temperatures were observed [13].

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