The effect of particle size on the supercritical CO_2 extraction of hydrocarbons from the Marcellus Shale

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Introduction

Supercritical gas extractions with carbon dioxide (CO_2) have successfully recovered a wide range of nonpolar polycyclic aromatic hydrocarbons (PAHs) and aliphatic hydrocarbons from different source rock matrices (i.e. coal and shale) under varying extraction conditions. The primary goal of this study is to examine CO_2-based extraction of n-aliphatic hydrocarbons from crushed samples of Marcellus Shale cores, and to evaluate how the efficiency of extraction varies as a function of particle size. The samples are from cores drilled at depths ranging from 1,924 to 2,278 meters, in Pennsylvania, USA.

Results

Reflectance measurements on solid organic matter of the shale samples confirms their high level of thermal maturation, ranging from 2.2-2.6% R_o, and suggests that solid pyrobitumen constitutes the bulk of the total organic carbon (TOC) present in the analyzed samples. Supercritical CO_2-extractions of the crushed shale samples (1000-500μm, 250-125μm, and 63-25μm particle size fractions) achieved successful extraction of diesel-range n-aliphatic hydrocarbons from n-C_{11} through n-$C_{21.}$ Total quantity of recovered n-aliphatic hydrocarbons ranges from approximately 0.01 to 0.6 parts per thousand by weight (milligrams of hydrocarbon per 1 gram of bulk shale sample), or approximately 0.03-1.2% TOC. The distribution and quantity recovered is consistent with the high maturity of the sampled shale. Notably, the recovery of total hydrocarbons increases approximately 15-60% from the 250-125μm particle size in comparison to that extracted from the 1000-500μm fraction for the cores sampled. However, recovery is maximized for the 250-125μm particle size, with lower yields for the 63-25μm particle size. Understanding the chemical and physical interactions of high-density CO_2 with organic and inorganic shale matrices will help further evaluate controls on hydrocarbon recovery within the Marcellus and other unconventional source rocks.