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Invited review

# Clay mineralogy and unconventional hydrocarbon shale reservoirs in the USA. I. Occurrence and interpretation of mixed-layer R3 ordered illite/smectite

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ABSTRACT

The mineralogy of many of the major unconventional hydrocarbon shale reservoirs in the USA, which span practically the whole spectrum of Phanerozoic time, is reviewed from a survey of relevant published literature. This survey reveals that there is a remarkable uniformity in the mineralogy of these shales, both with regard to non-clay minerals but particularly to the clay minerals. It was found that the clay mineralogy of practically all of the shale reservoirs older than the Upper Cretaceous are dominated by illitic clays, both in discrete form and as illite-dominated, mixed-layer, illite-smectite (I/S). The layer stacking arrangement of the latter is of the long-range type described as R3, such that every smectite layer tends to be preceded and succeeded by three illite layers in a sequence like IIISIIIS. Such material is conventionally interpreted (a) as having formed from a smectite precursor, (b) as existing in MacEwan-type crystallites consisting of about 5 to 15 unit layers in thickness where there is three-dimensional regularity across the smectite interlayers, and (c) as having interlayers of a truly smectitic character. Using evidence from the fundamental particle concept of Nadeau et al. (1984b) this interpretation is rejected. Instead, it is proposed that R3-type I/S (a) forms *de novo*, crystallizing from pore waters of appropriate chemical composition in a particular pressure and temperature stability field, as it does in conventional sandstone reservoirs, (b) consists primarily of thin illite crystallites or crystals <50 Å in thickness, and (c) that the "smectite" interlayers can be accounted for by the ability of such thin illite stacks, which have no three-dimensional register between the fundamental particles when sedimented onto glass slides, to adsorb ethylene glycol between the particles so leading to a false diagnosis of "smectite". This interpretation could have major consequences on the physicochemical properties of the shale, a matter that is examined more closely in the second part of this review.



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## 1. Introduction

Designation of a hydrocarbon reservoir as unconventional emphasizes the fact that the rock acts as a source, store/reservoir and seal of the hydrocarbons it contains. In contrast, the conventional reservoir usually involves the accumulation of hydrocarbons after their generation and migration from a different and often distant source rock, as well as being sealed by an impermeable cap rock belonging to another lithological formation. An unconventional hydrocarbon reservoir may, therefore, be regarded as being self-sourced and of a highly impermeable nature, usually requiring stimulation through induced hydraulic fracturing if it is to be commercially viable.

Unconventional hydrocarbon reservoirs are often described as "shales" but the meaning of this term varies according to the context in which it is used. For the geologist the defining characteristics of shale are that it is a sedimentary rock, fine-grained and fissile or laminated. A fairly comprehensive definition of shale is that of the American Geological Institute (Bates and Jackson, 1980). "A fine-grained detrital sedimentary rock formed by consolidation (especially compression) of clay, silt or mud. It is characterized by a finely laminated structure, which imparts a fissility approximately parallel to the bedding along which the rock breaks readily into thin layers,…., and by an appreciable content of clay minerals and detrital quartz; a thinly laminated or fissile claystone, siltstone or mudstone. It normally contains at least 50% silt with 35% clay or fine mica fraction and 15% chemical or authigenic material." In the petroleum industry, however, the term "shale" is used in a very much broader sense and may refer to sedimentary rocks which are not fissile, laminated or particularly fine-grained, as well as those rocks falling within the above geological definition. For example, the Bowland shale in the UK is a formation of Mississippian age and contains a wide range of lithologies, including calcareous mudstones, siltstones, turbiditic packstones and even sandstones (Clark et al., 2014). Such heterogeneity in nominal shale formations is also typical of gas shale formations in the USA. Thus, the clay content of different lithologies of the well-known Barnett Shale varies from 8 to 48% in siliceous mudstones, 7 to 34% in calcareous mudstones and 8 to 24% in calcareous, turbiditic packstones (Loucks and Ruppel, 2007), and with regard to the Utica Shale play in New York State, according to one geologist it should be more appropriately referred to as the "Utica shale and associated organic-rich calcareous shale and interbedded limestone and shale play." Such lithological heterogeneity must be borne in mind when considering the possible influence of clay mineralogy on hydrocarbon exploitation.

It is generally agreed that it is important to characterize the mineralogy of the bulk rock when attempting to evaluate the potential quality of unconventional hydrocarbon reservoir rocks, irrespective of whether the mineralogical data have been obtained from logs, cores or cuttings. Mineralogy is known to impinge upon a variety of petrophysical parameters including, for example, porosity, permeability, water saturation, as well as attributes related to rock strength such as Young's modulus and Poisson's ratio which are crucial for optimizing the potential of the formation for hydraulic fracture stimulation. A recent contribution to the influence of mineralogy on the quality of unconventional reservoirs developed a classification for organic mudstones, based on ternary plots of the normalized contents of clay minerals, carbonate minerals and silicate minerals, as estimated from geochemical logs (Gamero-Diaz et al., 2013). This classification subdivides organic mudstones into 16 different categories occupying separate areas on the ternary plot and was considered to provide a qualitative means of visualizing the relationship between overall bulk mineralogy of the rock and indicators of reservoir and completion qualities. It was found that there was a strong correlation between bulk mineralogy and completion quality, based on indicators such as minimum closure stress and mineral brittleness index. There was also a good correlation between mineralogy and reservoir quality, based on parameters such as effective porosity, matrix permeability and hydrocarbon saturation, although the correlation was not as strong as that between mineralogy and completion quality.

However, the mineralogy of unconventional hydrocarbon shale reservoirs has been less extensively characterized than conventional sandstone reservoirs, at least by core analyses using standard analytical techniques such as X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM). For the unconventional reservoirs, the usual industry practice is to use the mineralogical analyses of selected samples that are available to calibrate the petrophysical data acquired by downhole logging tools. In this way, quantitative (or semi-quantitative) mineralogical analyses are recorded over the complete drilled stratigraphic sequence. These data are regarded as extremely important as "the relative concentrations of the (mineral) constituents have the potential to make or break a potential resource play" (Alexander et al., 2011).

In this paper, the mineralogy, and particularly the clay mineralogy of a wide variety of unconventional reservoirs in the USA is reviewed, with the broad aim of trying to establish a better understanding of the relationship between individual clay minerals and petrophysical properties,