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Water-rock interaction during the diagenesis of mud and its prospect in hydrogeology

Yao Du [a, b], Teng Ma [a, b, *], Cong Xiao [a, b], Yanjun Liu [a, b], Liuzhu Chen [a, b], Haotian Yu [a, b]

[a] *State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Wuhan 430074, China*
[b] *School of Environmental Studies, China University of Geosciences, Wuhan 430074, China*

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ABSTRACT

Mud is a fine, saturated and unconsolidated sediment with high moisture and abundant organic matter, mineral and microbe. As the burial diagenesis of mud proceeds, the water - rock - gas - organic matter - microbe in mud interact and co-evolve, forming mudstone or shale finally. During mud diagenesis, pore water not only changes over its own properties, but also participates in diagenetic reactions actively, and involves in the production of some mineral resources (hydrocarbon, biogenic gas, methane hydrate, etc) directly or indirectly. Researching water-rock interaction during mud diagenesis will be of great theoretical and practical significance to further understanding the genesis of natural groundwater, diagenetic reactions and mineralization, and has broad prospects in hydrogeology. With diverse high temperature and high pressure devices emerging, and the rapid expansion of in-situ detecting technology, simulating forwardly diagenetic processes of mud using indoor elevating temperature and pressure technology provides a new perspective for further researching water-rock interaction during the diagenesis of mud.

1. Introduction

Mud is a kind of fine, saturated and unconsolidated sediment in modern drainage basins, in which there are abundant pore water, high organic matter content, complex mineral composition, abundant microbe and some gas (Potter et al., 2005). In traditional hydrogeology, aquifers were concerned much owing to their important resource attribute. In recent years, with the development of theory on groundwater flow system and the deepening of research on groundwater contamination, aquitards such as clay or silty clay layer have also gotten much attention (Hendry and Wassenaar, 2000, 2004; Parker et al., 2008; Wang et al., 2013; Kuang et al., 2016). Mud has high moisture but it can not release considerable water in natural state, so it hasn't become the emphasis in hydrogeology research.

In the sedimentology, mud is the origin of mudstone or shale (Aplin et al., 1999; Aplin and Macquaker, 2011). During the sedimentation, the temperature and pressure of strata increased gradually, and water, mineral, gas, organic matter and microbe interacted and co-evolved. During this process, pore water was expelled, organic matter was gradually degraded and matured, mineral was assembled and transformed, and pore was continuously compressed into micropore. As a result, mudstone or shale was formed finally (Potter et al., 2005).

During the diagenesis of mud, waters play a quite important role. Firstly, waters not only provide room for biochemical reactions, but also involve actively into diagenetic reactions. So chemical compositions of pore water in mud have been widely used to trace diagenetic processes and reactions (Hesse, 1986; Schulz, 2006). Secondly, waters involve in the formation of some mineral resources directly or indirectly, such as biogenetic gas (Golding et al., 2013) and natural gas hydrate (Hesse, 2003; Jiang et al., 2008; Yang et al., 2010, 2013) at early stage, and oil and natural gas (Shuai et al., 2012) at late stage. So it is a current hotspot in geoscience field to research the indication of pore water chemistry to mineralization. In addition, water in mud is the origin of groundwater in sedimentary formation (Shishkina, 1972). Under overlying strata pressure, considerable amount of pore water in original mud was forced out and penetrated into larger-pore sediment, which could have great influence on origin and evolution of groundwater in adjacent aquifer (Liu et al., 2017). Therefore, chemical components of pore water in original mud should be valued when researching the genesis of natural groundwater (Shen et al., 1993).

* Corresponding author. State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Wuhan 430074, China.
E-mail addresses: mateng@cug.edu.cn, matengcug@gmail.com (T. Ma).

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As such, it is clear that water in mud is an active geological agent during diagenesis, the chemical characteristics of which influence the evolution of other geological elements to a large extent. The chemical characteristics of water is closely related to experienced water-rock interaction processes. Therefore, there are important theoretical and practical significance for further knowing the genesis of natural groundwater, diagenetic reaction and mineralization, and also broad prospect in hydrogeology to research water-rock interactions during diagenesis of mud. The present paper aims to (1) review briefly the general characteristics of mud and its geological evolution, (2) discuss the indication of pore water during mud diagenesis, and (3) put forwards a new insight into the studies on water-rock interactions during mud diagenesis.

2. Physical, chemical and biological characteristics of mud

The term “mud” was widely applied in engineering geology, marine science, sedimentology, etc. The definitions of mud were diverse in different literature and was called “soft ooze” (Wang et al., 1994) or “argillaceous sediment” (Burst, 1976) in some research, depending on study objective or subject. In field of water science and environment, a term “sludge” similar to mud was widely used (Liao et al., 2016; Montalvo et al., 2016). In spite of no clear definition, there are generally some special physical, chemical and biological characteristics for mud as follows.

Mostly, mud are deposited in discharge area of groundwater system, including ocean, lake, delta, wetland, etc, which belong to retention zone of water and accumulation zone of chemical energy, and are also the information database of environmental change (Liu et al., 2002) and carbon sink of greenhouse gas (Cai et al., 2007). The mean grain size of mud is considerably small, generally less than 0.0625 mm (Folk, 1954; Aplin et al., 1999). The original porosity of mud is quite high, generally more than 70% (Lasemi and Sandberg, 1984; Hart et al., 2013). The natural moisture content of mud is also high, generally more than 75%, the existing forms of water in which include gravitational water, bound water and mineral-combined water (Siever et al., 1965; Vesilind and Hsu, 1997). Clay minerals are the major minerals in mud with detrital minerals including quartz and feldspar as minor minerals (Aplin and Macquaker, 2011). Mud is enriched in organic carbon, the content of which is generally more than 2%, primarily deriving from that remains of higher plant or plankton (Schnurrenberger et al., 2003). Mud also contains abundant microorganism, such as aerobic bacteria, sulfate-reducing bacteria and methanogens, and gas, such as oxygen and carbon dioxide (Potter et al., 2005).

3. Evolution of water-rock-gas-organic matter-microbe system during mud diagenesis

Diagenesis process refers to all changes from deposition of unconsolidated sediment to minimum metamorphism vertically, but the boundary between diagenesis and metamorphism has not been agreed on. It is generally recognized that the temperature boundary is between 120 and 200 °C, and the pressure boundary is from 100 to 200 MPa. Under the condition in which temperature and pressure are higher comparing to boundary temperature and pressure, organic matter approaches to or has reached to high-maturation stage, and almost all minerals begin to recrystal (Krumbein, 1983).

During diagenetic evolution of mud, it is a complex but ignored scientific problem how water, mineral, gas, organic matter and microbe interact. Restricted by disciplines, scientists in different fields tend to just concern on one problem or one aspect. For example, the research target of hydrogeochemists is pore water in muddy sediment (Wang et al., 1994; Bufflap and Allen, 1995) or sedimentary formation water (Kharaka and Hanor, 2004). The research in field of sedimentology mainly focus on transformation and evolution of minerals (Ruiz and Reyes, 1998; Jiang, 2003; McCarty et al., 2008). The research focus of scientists in petroleum geochemistry are the degradation and maturation of organic matter, and formation and storage of hydrocarbon (Liu, 2009; Welte et al., 2012). Scholars in microbiology field concern on biogeochemical processes during mud evolution (Hama et al., 2001; Kim et al., 2004; Dong et al., 2009). Although the research emphasis of scientists in different fields are different, they all had great contributions to deeply understand the interaction process.

The burial process of mud can be divided into physical process and chemical process in general. The physical process is primarily mediated by pressure, and chemical process is much more complex, which is mediated by microbe and temperature during shallow burial and deep burial, respectively (Potter et al., 2005).

Physical process is mainly compaction and consolidation of mud under the pressure of overlying water and sediment, for which the most significant effect are the decrease of porosity and expelling of pore water. In general, within burial depth of 300–500 m, pore water is liable to be expelled and porosity decreases sharply. When burial depth is more than 500 m, reduction of porosity slows significantly, the reason for which is that further decrease of porosity needs to be achieved by expelling the interlayer water and structural water integrated closely with clay. With depth increasing, it will also be more and more difficult to expel interlayer water and structural water (Siever et al., 1965; Jiang, 2003).

Chemical process during shallow burial of mud is a complex water-rock interaction process mediated by microbe. It will experience the evolution from aerobic zone, anaerobic zone to methanogens zone. In every zone, there is corresponding microbial community, that is aerobic bacteria, sulfate reducing bacteria and methanogen, respectively (Hesse and Schacht, 2011). In the top layer of mud, free oxygen was constantly consumed by organic matter under the effect of aerobic bacteria. When it turned into anaerobic environment due to consumption of oxygen, anoxic sulfate reduction became the leading chemical process. After sulfate was reduced absolutely, methanogens could reduce CO_2 to CH_4 (Judd and Hovland, 2007). The characteristics (salinity, oxygen level, etc) of bottom water control chemical process intensity during early burial of mud largely (Potter et al., 2005). For example, in marine and salt lake environment, salinity of bottom water is high with high content of SO_4^{2-}, for which stronger anoxic sulfate reduction could occur. On the contrast, the content of SO_4^{2-} is low in fresh water environment, for which anoxic sulfate reduction could be weak or even hardly take place (Leggett and Zuffa, 1987).

Chemical process during deep burial of mud is a water-rock interaction process under the effect of high temperature and pressure, the most representative changes in which are the transformation of clay minerals and thermal maturation of organic matter.

Clay minerals will release interlayer water and structural water under the effect of thermal reaction. Owing to the change of crystal structure, transformation of clay minerals will take place. The conversion from montmorillonite to illite (or chlorite) is the most important process, which is mainly achieved by two means. One is that detrital feldspar supplies K^+ (or Mg^{2+}) in this process: $Al_2Si_4O_{10}[OH]_2nH_2O + KAlSi_3O_8 \rightarrow KAl_3Si_3O_{10}[OH]_2 + 4SiO_2 + nH_2O$. Another is that K^+ (or Mg^{2+}) in pore water act as the reactant: $2K^+ + 3Al_2Si_4O_{10}[OH]_2nH_2O \rightarrow 2KAl_3Si_3O_{10}[OH]_2 + 6SiO_2 + 2H^+ + 3nH_2O$ (Essene and Peacor, 1995). In addition, with the burial depth increasing, kaolinite will react with K^+ in formation water, and convert gradually into illite (Sutton and Maynard, 1996).

Thermal maturation of organic matter is the most important change during deep burial of mud. In the stage of shallow burial,