Hardness and compositional variations in the siliceous mudstones of Akita and Tsugaru Basins

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Fine sedimentary formations such as shales and mudstones are characterized by natural fracture systems. These systems alter the porosity and permeability properties of the sedimentary formations and can favor the migration of fluid geo-resources such as hydrocarbons. In this context, the analysis of hardness variations is fundamental to define the susceptibility of a sedimentary formation to the formation of brittle fractures. Hardness variations are, generally, controlled by the relative percentages of quartz, carbonate, and clay minerals. Siliceous formations distributed in several areas in the world including North America, and Asia are important source rocks for oil and gas, and they are characterized by several variations in the mineralogical composition which could affect also the hardness variations. In this context, the siliceous formations from Akita and Tsugaru Basins in northern Honshu Island represent important case studies to analyze the relationship between hardness and composition.
In this study, we are going to present results from the analysis of the middle-late Miocene Odoji and Onnagawa siliceous mudstones, located respectively in the Tsugaru (southwestern Aomori Prefecture) and Gotanzawa (central Akita Prefecture) areas. Samples of siliceous mudstones have been collected, observed, and analyzed with the XRF method to estimate the percentage of major elements. In this study, percentages of Si, Al, and Ca were considered as proxies for quartz, clays, and carbonates. Moreover, biogenic Si (BSi) was calculated as the difference between total Si and Si associated with Al, using the formula $BSi = Si_{sample} - [(Si/Al) * Al_{sample}]$, where Si/Al was considered equal to 3.11 (average shale Si/Al). On the same samples, the hardness was measured using a hardness tester. The average hardness values were calculated from five measurements, and are expressed as Leeb hardness values (HL scale).
Samples of the analyzed Odoji and Onnagawa Formations are composed of light gray and olive-gray massive mudstones. All samples were characterized by high values of Si (36–41 wt. % in the Odoji Formation, 23–41 wt. % in the Onnagawa Formation), moderately low values of Al (1.4–4.2 wt. % in the Odoji Formation, 1.5–7.2 wt. % in the Onnagawa Formation and low values of Ca (0.3–0.9 wt. % in the Odoji Formation, 0.2-1.2 wt. % in the Onnagawa Formation). Most of the Si in the two formations was composed of BSi (21–34 wt. % in the Odoji Formation, 16–36 wt. % in the Onnagawa Formation). The average hardness is a range of 580–776 HL in the Odoji Formation and in a range of 549–781 HL in the Onnagawa Formation.
By comparing the variations of BSi, Al, and Ca with hardness, a positive correlation (R2 = 0.50–0.70) between the BSi and hardness was found in both formations. In contrast, the hardness resulted negatively correlated with the Al contents (R2 = 0.62–0.73). Finally, the Ca did not show any correlation with the hardness values. Based on these correlations, we hypothesize that the brittle behavior in the Odoji and Onnagawa Formation was mostly controlled by the content of BSi. However, the presence of consistent contents of Al (i.e., clay minerals) would have reduced the brittleness of these formations. The presence of highly brittle horizons within these formations may have determined the formation of fracture systems. Lastly, the comparison between the hardness ranges in the Odoji and Onnagawa Formations with other notable siliceous formations such the Monterey, Horn River, and Woodford, reveals a similar trend in hardness between these formations, suggesting a comparable degree of brittleness.

Keywords: Akita and Tsugaru Basins, Biogenic silica, Clay minerals, Hardness, Siliceous mudstones

