

A NEW AND IMPROVED METHODOLOGY FOR QUALITATIVE AND QUANTITATIVE MINERALOGICAL ANALYSIS OF BOOM CLAY

E. Zeelmaekers¹, M. Honty^{2*}, A. Derkowski³, M. De Craen²,
N. Vandenberghe¹, M. Van Geet⁴

1. Geo-Institute, KULeuven, B-3001 Leuven-Heverlee, Belgium

(Edwin.Zeelmaekers@ees.kuleuven.be)

2. SCK•CEN, Waste and Disposal, B-2400 Mol, Belgium (mhonty@sckcen.be)

3. University of Alberta, Edmonton, Alberta, T6G 2G6, Canada (derkowski@ualberta.ca)

4. NIRAS/ONDRAF, 1210 Brussels, Belgium (mvangeet@nirond.be)

A good knowledge of the mineralogy of any host formation studied for geological disposal of high-level radioactive waste, is a prerequisite for understanding the geochemical environment which will determine the migration and retention behaviour of radionuclides. In this respect, the Boom Clay mineralogical composition has been extensively studied last decades as reference host formation (e.g. ARCHIMEDE-ARGILE project, OECD-NEA clay catalogue report) with the aim to provide reliable data for a safety assessment. However, a comparison of the available literature data clearly showed a serious discrepancy among studies, not only in the quantitative, but also in the qualitative mineralogical composition of the Boom Clay (SAFIR II). The reason for such a huge disagreement could be related, among others, to variable grain size distributions of the studied samples (sample heterogeneity) and differences in the methodological approaches. In particular, the unambiguous characterisation of clay minerals and the quantification of mixed-layer phases appeared as an everlasting problem.

This study is aimed at achieving a consensus on the qualitative and quantitative mineralogical data of the Boom Clay using the most advanced techniques currently available in the clay science. A new sampling campaign was performed in such a way that samples are (20 in total) more or less regularly distributed over Boom Clay Formation, ensuring that variations in the grain size distributions due to silty clay-clayey silt layers alternations are accounted for. The novel concept based on an analysis at two levels was applied: (1) bulk rock and (2) clay fraction analysis.

(1) A bulk rock analysis consists of conventional XRD analysis with the identification of the principal mineral phases. As a next step, the bulk rock was mixed with a ZnO internal standard and experimental diffraction patterns of randomly oriented powders were analyzed using “Quanta” (Chevron proprietary software, Mystkowski, 2002). The Quanta results were further refined with bulk rock chemical analysis (major oxides and trace elements), CEC and water adsorption at 110°C using the “BestRock” software (Chevron proprietary software, Derkowski, 2008). Bestrock provided mineral structural formulae, trace element distributions over the minerals and a set of petrophysical parameters of the individual minerals (e.g. vol.%, mineral density, dry mineral matrix density, hydrogen index, oxygen index) as an output. A chart illustrating the data flow in the course of the bulk rock analysis is shown in the Figure 1. A quantification of amorphous phases (organic matter, and poorly crystallized Al, Fe and Mn oxyhydroxides) was carried out by independent techniques as well.

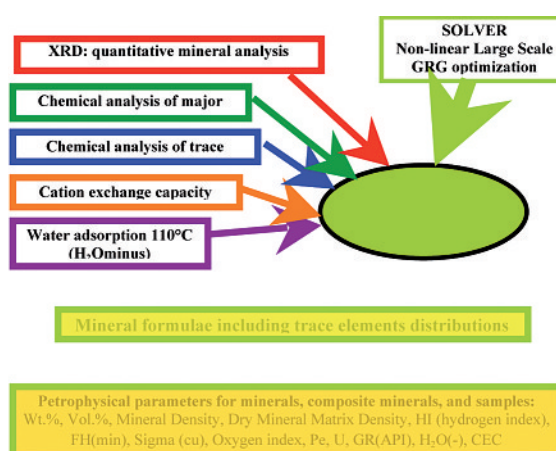


Figure 1: A flowchart showing a sequence of the bulk rock data processing in an integrated chemical-mineralogical analysis (after Derkowski, 2008).

(2) The clay fraction ($< 2 \mu\text{m}$ and $< 0.2 \mu\text{m}$) was studied in order to further refine the clay mineralogical composition. The oriented slides of air-dried and ethylene glycolated clays in Ca-form were used for conventional XRD analysis. In addition, K-, Mg- and Li- saturation and heating to 550°C was also applied in order to discriminate between various clay species. Sybilla software (Chevron proprietary software) was used as a quantification tool in order to model the experimental air-dried and ethylene glycolated diffraction patterns of the basal reflections (00 l). In addition, as an independent check the theoretical wt% K₂O and water loss at 200°C for the model was compared to the actual measured values on the clay fraction.

Altogether, an integration of the data from Quanta, Bestrock, Sybilla and chemical analysis of bulk rock and clay separates allowed for an up-date of the qualitative and quantitative mineralogical composition of the Boom Clay. Table 1 gives a qualitative and quantitative overview of a Boom Clay sample situated at a depth of 225.59 m corresponding to HADES URL level as an example.

Table 1: Quantitative mineralogical composition of the Boom Clay sample situated in the depth of HADES URL (Mol, Belgium). I-illite, S-smectite, V-vermiculite

	BESTROCK corr. (wt%)	BESTROCK Structural Formula
Quartz	21.0	<i>Fixed</i>
K-spar	3.9	$\text{K}_{0.72}\text{Na}_{0.28}\text{AlSi}_3\text{O}_8$
Plagioclase	0.4	$\text{Na}_{1.00}\text{Ca}_{0.00}\text{Al}_{1.00}\text{Si}_{3.00}\text{O}_8$
Calcite	4.2	<i>Fixed</i>
Siderite	0.3	$\text{Ca}_{0.00}\text{Mg}_{0.40}\text{Fe(II)}_{0.60}\text{Mn}_{0.00}\text{CO}_3$
Pyrite	1.9	<i>Fixed</i>
Anatase	0.8	<i>Fixed</i>
Apatite	0.2	<i>Fixed</i>
Organic matter	0.8	<i>N/A</i>
Sum Non-Clay	33.5	
Kaolinite	8.3	<i>Fixed</i>
ISV R0 (41:49:10)	22.9	<i>N/A</i>
ISV R0 (77:23:0)	15.0	<i>N/A</i>
Illite/muscovite	7.2	<i>N/A</i>
SV di- (89:11)	9.1	<i>N/A</i>
Vermiculite/Chlorite	3.9	$(\text{Mg}_{2.35}\text{Al}_{1.00}\text{Fe(II)}_{2.65}\text{Fe(III)}_{0.00})(\text{Si}_{3.00}\text{Al}_{1.00})\text{O}_{10}(\text{OH})_9$
Sum Clay	66.4	
TOTAL	99.9	

References:

- Derkowski, A., McCarty, D. K., Środoń J., Eberl, D.D., 2008. BestRock – mineralogy, chemistry, and mineral surface property optimization to calculate petrophysical properties of the mineral matrix. *Mineralogia – Special Papers*, 33, 53 (4th Mid-European Clay Conference, Zakopane, Poland, Sept 2008).
- Mystkowski, K., Środoń, J., McCarthy, D.K., 2002. Application of evolutionary programming to automatic XRD quantitative analysis of clay-bearing rocks: The Clay Minerals Society 39th Annual Meeting, abstract, Boulder, Colorado.
- NIROND, 2001. SAFIR-II. Second Safety Assessment and Feasibility Interim Report, NIROND, Brussels.