

Consideration of Mineralogy and Permeability of Sand/Bentonite Mixes

B. Makgekgenene, Department of Civil Engineering, University of Cape Town, Cape Town, South Africa.
makgekgeneneb@jgi.co.za

F. Scheele, Dr. - Ing. F. Scheele & Assoc, Cape Town, South Africa

ABSTRACT

The disposal of municipal, commercial and industrial waste contained in near impervious lining systems still poses challenges in terms of sustainability and long-term durability.

This paper assesses the engineering behaviour and chemical performance of selected sand/bentonite mixes to be used in the context of an environmentally inert landfill liner. Numerous test series in the laboratory focused on the permeability as a consequence of the physio-chemical characteristics of the selected mixes of two South African Bentonites and locally available sands. Several exploratory techniques (i.e. Cation Exchange Capacity; Scanning and Transmission Electron Microscopy) were employed to obtain the respective properties of the test materials while a flexible membrane test setup in connection with the falling head principle was used to determine the permeability of the mixes. A wide variety of bentonite percentage mixes (by mass) and a range of all round pressures were investigated.

The results of the exploratory techniques reveal valuable information on the quality and uniqueness of the two bentonites and allow comparative studies of their performance within a technical application. A visual presentation of bentonite content, density of the mixes, confinement and coefficient of permeability is also given.

1. INTRODUCTION

Traditionally, landfills operated on the dilute and attenuate principle. This principle allowed leachate to leak from sites into the underlying strata where contaminants would be attenuated by a suite of physical, biological, and chemical processes. Due to the ever present fact of aquifer contamination a more sustainable approach has been adopted which relies on the advances in modern barrier liner designs. This paper focuses on the selection of appropriate materials for the specification of sand/bentonite mixes in the application of hydraulic barriers in landfills.

1.1 Mineral Structure and Geotechnical Properties of Bentonite

Bentonite is a name used to identify naturally occurring clay minerals comprising the Montmorillonite mineral. The clay mineral is found in deposits of volcanic ashes at shallow wet sites in various locations around the world (Grim, 1968). Two types of bentonites exist, i.e. Calcium Bentonite and Sodium Bentonite. The mineral is mostly valued for its crystalline structure responsible for swelling. This swelling is facilitated by the 2:1 expandable silicates layers which are composed of two tetrahedral sheets separated by an octahedral sheet (Grim, 1968). Bentonite swells by inhibiting water or polar organic solvents between the minerals quasicrystals and between the individual layers within the quasicrystals. Laird (2006) suggests that smectites swelling should be viewed as an orchestra of different mechanisms operating in concert. These mechanisms are crystalline swelling, double-layer swelling, formation and break-up of quasi crystals, cation demixing and Brownian swelling. The mechanisms are controlled and operated by different physicochemical conditions such as cation positions and type, rate of cation exchange, attraction and repulsive forces, isomorphous substitutions and layer charges.

1.2 South African Bentonites

In South Africa, relatively pure deposits of bentonite exist. Sodium Montmorillonite deposits occur in the Heidelberg area whilst Calcium Montmorillonite deposits occur in the Koppies area and near Plettenberg Bay, respectively. The mining process of the Calcium bentonite involves acid-activation to enhance surficial properties of the clay (Agnello, 2004). Acid-activation of clays refers to the controlled partial dissolution of the raw materials by the acid giving rise to a mixture of protonated clays and amorphous hydrous silica phases (Komadel et al., 1997; Komadel, 1999; Gates et al., 2002). Studies have shown that the effects of acid activation, i.e. dissolution rates depend on the composition of the smectite type. The resulting product is an increase in surface area and porosity at the expense of the 2:1 expandable silicates layers (Komadel et al., 1997). The mining process and distribution of the Heidelberg bentonite does not involve acid activation.

2. SAND/BENTONITE MIXES

Sand/bentonite mixes are widely used as barriers to control the movement of liquids from waste disposal facilities due to their enhanced geophysical properties (Chapuis, 1989). Sand/bentonite mixes, in general, exhibit high strength, low compressibility and very low hydraulic conductivities. In a sand/bentonite mix the sand accounts for the structural integrity and stability while the hydrated bentonite fill the voids within a compacted matrix. Depending on the grain size distribution, texture and grain shape of base sand, the bentonite particles settle in various distinctive coating patterns. Furthermore, the morphology of the bentonite influences the void closure and distribution of the bentonite within the resulting fabric of the mixture.

The aim of this research was to explore the use of sand/bentonite mixes as an impervious lining system for hazardous waste landfills. The selected sands comprise two geologically different local sands, a Klipheuwel sand and a Philippi Dune sand. Klipheuwel sand is a weathered Cape Granite deposits from the Malmesbury area, whilst Philippi Dune sand is an aeolian sand from the Cape Flats area. The bentonites are from the Heidelberg deposit, courtesy of the Cape Bentonite Mine and Envirobent is a commercial product from Koppies in the Free State, supplied by G&W Base minerals.

3. INVESTIGATIVE TECHNIQUES

Geochemical tests were performed in various laboratories at the University of Cape Town. Investigative techniques of the bentonite involve the use of X-Ray Diffractometer (XRD), Cation Exchange Capacity (CEC) and High Resolution Transmission and Scanning electron microscopy (TEM & SEM). A Falling-Head Triaxial Permeability test stand was developed in the Geotechnical Laboratory of the Department of Civil Engineering. The permeability tests were carried out in accordance to the ASTM D5084 protocol.

4. GEOCHEMICAL OF RESULTS RESEARCH MATERIALS

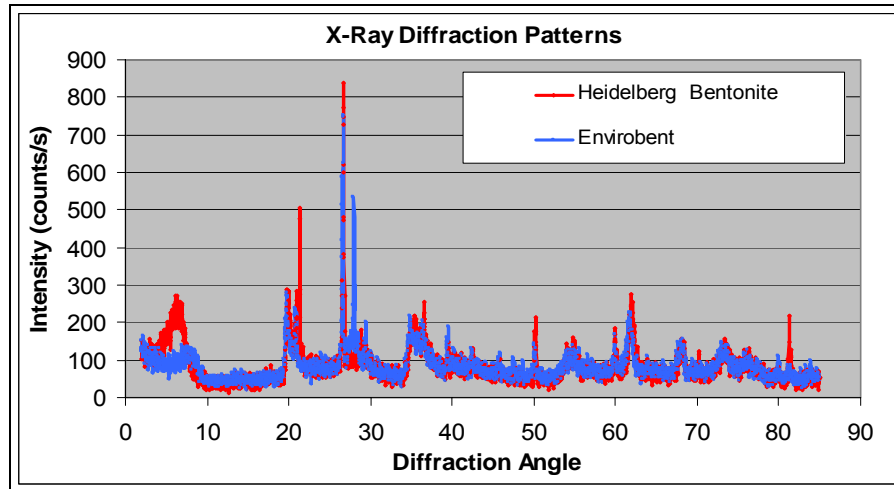
The geophysical properties of Heidelberg Bentonite and Envirobent are listed in Table 1.

Table 1: Geophysical properties of Heidelberg Bentonite and Envirobent

Property	Envirobent	Heidelberg
Specific Gravity, G_s	2.60	2.50
Natural Moisture Content (%)	4.00	39.00
Particle Range μm	0.05 - 48.27	0.05 - 76.32
Mean Grain Size, D_{50} μm	2.46	6.24
Coefficient of Uniformity, C_u	5.26	5.11
Coefficient of Curvature, C_z	1.03	0.20
Plastic Limit (%)	39	41
Liquid Limit (%)	142	160
Plasticity Index (%)	103	119
Free Swell (%)	100	110

4.1 X-Ray Diffraction

X-ray diffraction patterns for pressed Bentonite powder were obtained using a Pan Alytical PW 3830 Diffractometer, operating at 40kV with 25mA. A Phillips X'Pert Software was used to identify minerals by a PDF number and colour coded mineral check lists. Graph 1 displays the diffraction patterns of Heidelberg Bentonite and Envirobent.



Graph 1: X-ray Diffraction pattern of Heidelberg Bentonite and Envirobent

4.2 Transmission Electron Microscopy

The high resolution transmission electron micrographs (see Image 1 and 2) show the arrangement of individual layers within a smectite quasicrystal of Envirobent and Heidelberg Bentonite. The bentonites were treated with Hexal-Alkylammonium cations and prepared for transmission of electrons according to the procedure described by Laird & Fleming 2008. Image 1 displays Hexal-Alkylammonium cation saturated Envirobent and Image 2 displays Hexal-Alkylammonium cation saturated Heidelberg Bentonite.

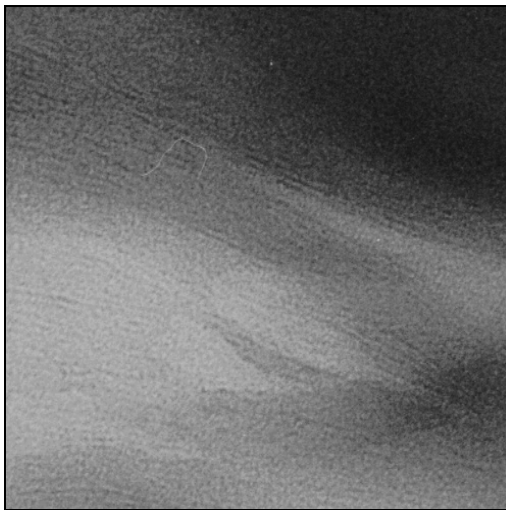


Image 1: High resolution transmission electron micrograph of Hexal-Alkylammonium cations saturated Envirobent quasicrystals (Scale 1:20nm)

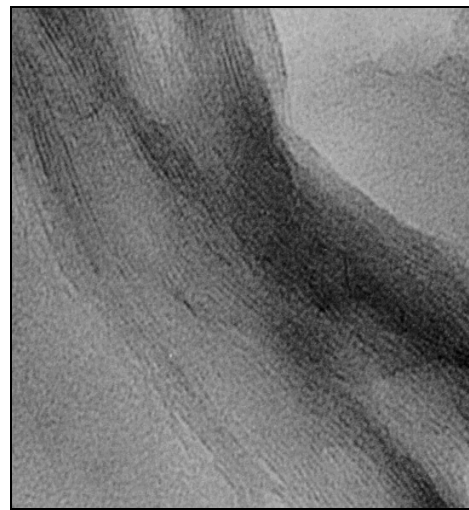


Image 2: High resolution transmission electron micrograph of Hexal- Alkylammonium cations saturated Heidelberg Bentonite quasicrystals (Scale 1:20nm)

4.3 Fabric of Sand/Bentonite mixes

Image 3 and 4 are scanning electron micrographs of Klipheuvel sand/bentonite mixes displaying possible combinations of the fabrics of the sand and the two different Bentonites. Image 3 is a Klipheuvel sand mixed with 9 % Envirobent at a moisture content of 8% and Image 4 shows Klipheuvel sand mixed with 9% Heidelberg Bentonite at a moisture content of 13%.

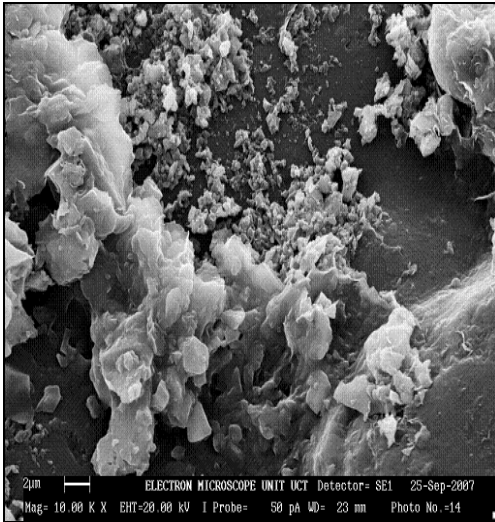


Image 3: SEM image of mix of Klipheuvel Sand/Envirobent (magnification 10000)

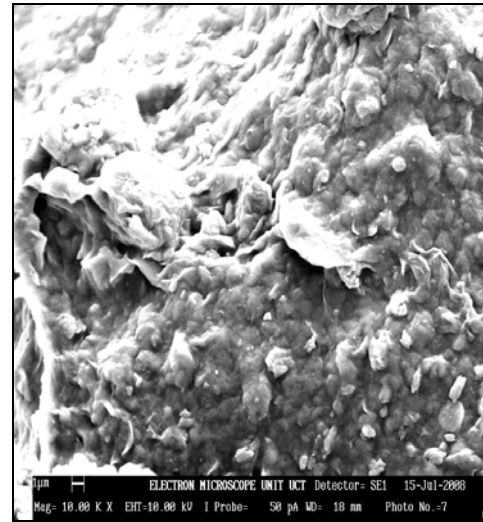
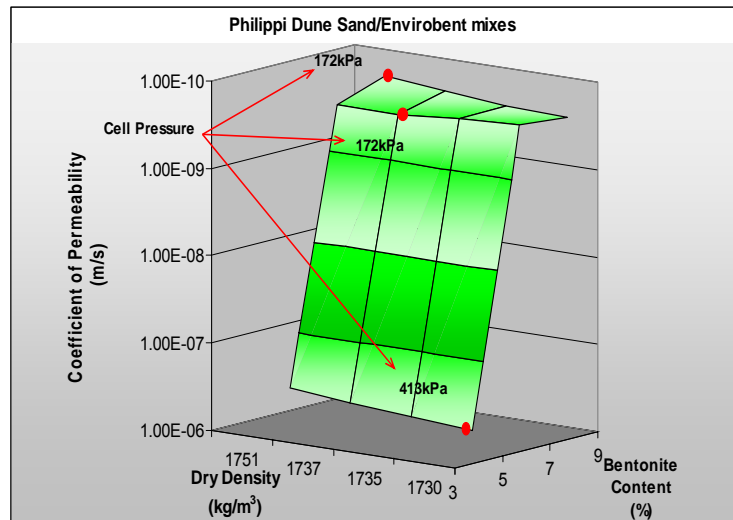


Image 4: SEM image of mix of Klipheuvel Sand/Heidelberg Bentonite (magnification 10000)

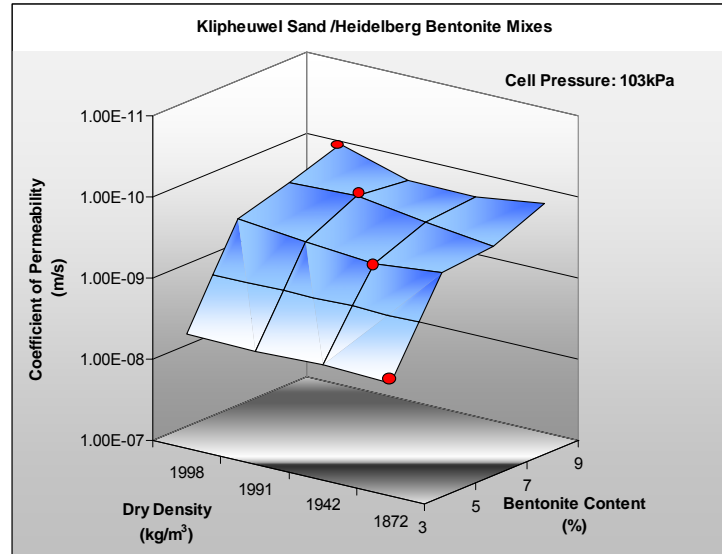
5. GEOTECHNICAL RESULTS OF RESEARCH MATERIALS

5.1 Permeability Results

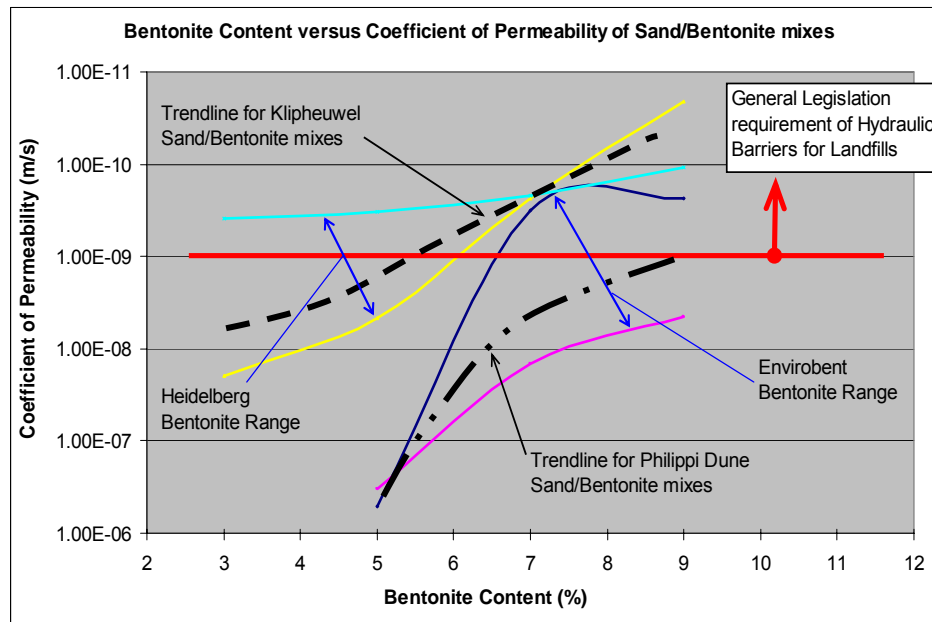
Three dimensional graphs of bentonite content, dry density, coefficient of permeability are shown in Graphs 2 and 3. In Graph 2 the results of Sand/Envirobent mixes are shown whilst Graph 3 displays the responses of Sand/Heidelberg Bentonite mixes. The applied cell pressures are also given in the graphs.



Graph 2: Dry Density versus Bentonite Content versus Coefficient of Permeability of Phillippi Dune Sand/Envirobent mixes



Graph 3: Dry Density versus Bentonite Content versus Coefficient of Permeability of Klipheuwel Sand/Heidelberg mixes



Graph 4: Effect of Sand type on the Coefficient of Permeability of Sand/Bentonite mixes

6. DISCUSSIONS AND CONCLUSIONS

6.1 Discussion addressing Geochemical Observations

The diffraction patterns of Envirobent sample as shown in Graph 1 display no peaks close to the diffraction angles of a Calcium Montmorillonite mineral. The pattern also reveals significant variation in the diffraction angle range of 8° to 20°. The mineral Illite and traces of Biotite were identified within the Envirobent sample. Illite is a three layered clay mineral which does not exhibit swelling

characteristics. The diffraction pattern of the Heidelberg Bentonite revealed presences of pure Sodium Montmorillonite mineral with the chemical formula, $\text{Na}_{0.3}(\text{Al}, \text{Mg})_2\text{Si}_4\text{O}_{10}(\text{OH})_2 \cdot 4\text{H}_2\text{O}$, a d-spacing of 13.6Å and a diffraction angle of 6.49°.

Acid activation causes a considerable amount of cationic activity within the physico-silicate layers of clay minerals. These activities involve atomic substitution, i.e. hydrogen cations, resulting in increases the specific surface area of the clay mineral (Babaki et al., 2007; Noyan et al., 2007). Horn et al. (2000) point out that the Koppies deposit is a trioctahedral smectite which dissolves much faster in acidic conditions than their dioctahedral counterparts (Komadel et al., 1997). The increase in surface area is a result of the combination of the smaller pores which bind together to create larger pores as a function of the acid concentration, i.e. higher acid concentrations causes greater alterations within the morphology of the clay.

Transmission electron microscopy of the individual layers of the quasicrystal structure of Envirobent treated with Hexal-Alkylammonium is displayed in Image 1. The image appears blurred and presents very few visible basal spaces of the individual layers. Some parts of the images appear as solid non-transmissible layers. Vali et al., (1994) points out that Illitic structures are not affected after n-Alkylammonium treatment. Furthermore, the image displays non-electron transmissible portions and inconsistent expansive patterns. This is evidence of Illitic behaviour as opposed to the uniform expansion of Calcium Montmorillonite layers.

In Image 2, the arrangement of individual layers of the quasicrystal structure of the Hexal-Alkylammonium treated Heidelberg Bentonite is displayed. The clear dark fringes represent the 2:1 physico-silicate layers of the clay mineral. These layers appear to be curvy suggesting flexibility and continuity, representing a “mosaic morphology” as described by Grim, (1968) and Laird, (1989).

The scanning electron micrographs show two different fabrics of sand/bentonite mixes (shown in Image 3 and 4). Envirobent displays a stacked “broken sheet morphology”, the Klipheuwel particles being trapped in between the layers. Towards the top-middle side of Image 3, clay-sized quartz particles are assembled in isolation. On the top right corner of the image silt-sized quartz particles and broken layers of Envirobent are visible. In Image 4 a continuous “blanket” of the Heidelberg Bentonite layers cover the particles of the Klipheuwel, i.e. the sand particles are completely embedded in bentonite.

Lithium Chloride solution was used as the leaching agent to remove the cations from the surface of the clay hence facilitating Atomic Absorption Spectrometry for concentration measurements. The measured Cation Exchange Capacity of Envirobent was 48meq/100g and 125meq/100g of Heidelberg Bentonite, respectively.

6.2 Discussion regarding Geotechnical Behaviour

Three dimensional representations of bentonite content versus dry density versus moisture content of the various sand/bentonite mixes are given in Graphs 2 and 3.

The research has shown that Philippi Dune sand/Bentonite mixes are less impervious than Klipheuwel sand/Bentonite mixes. It was found that above 7% Envirobent content no significant increase in dry density can be achieved (applying the same compaction effort). Therefore, the coefficient of permeability appears to be constant. Even an increase in the confinement pressure does not affect the coefficient of permeability. This is shown in Graph 4 whereby, Philippi Dune/ Bentonite mixes show no significant decrease in coefficient of permeability as the bentonite content increases.

Klipheuwel/Heidelberg Bentonite mixes display an increase in dry density (with constant compaction effort) as the bentonite content increases. It is also noted that the coefficient of permeability decreases as the bentonite content increases (see Graph 3). Furthermore, these observations are also presented in Graph 4.

Therefore, it is concluded that the quality of sand/bentonite mixes in the context of hydraulic barrier depends on the quality of the bentonite, i.e. mining process, mineralogy and geochemical composition. The selection base sand should be focused on the geotechnical properties of the sand, i.e. grain size distribution, shape and texture.

ACKNOWLEDGEMENTS

Financial support from Jeffares & Green Consulting Engineers (Cape Town) and academic contribution from Dr J Rogers (Department of Geological Sciences) was much appreciated.

REFERENCES

- Agnello, V.N. (2004). Bentonite, Pyrophyllite and Talc in the Republic of South Africa. Directorate; Mineral Economics, 1-67
- ASTM D5084. Standard test method for measurement of hydraulic conductivity of saturated porous materials using a flexible wall permeameter. American Society for Testing and Materials, [Online] Available: www.astm.org
- Bakaki, H., Salem, A. & Jafarizad, A. (2007). Kinetic model for the isothermal activation of bentonite by sulphuric acid. *Material Chemistry and Physics*, Vol. 108, 263-268
- Chapuis, R. (1989). Sand-bentonite liner: predicting permeability from laboratory tests. *École Polytechnique de Montréal, Québec, Canada*
- Gates, W.P., Anderson, J.S., Raven, M.D. & Churchman, G.J. (2002). Mineralogy of bentonite from Miles, Queensland, Australia and characterisation of its acid activation products. *Applied Clay Science*, Vol 20, 189-197
- Grim, R.E (1968). *Clay Mineralogy*. 2nd Ed, Mc Graw Book Company. New York
- Horn C.F.J & Strydom J.H. (2000), *Clay. The mineral resources of South African* 6th Ed. Council for Geosciences South Africa
- Komadel, P., Janek, M., Madejova J., Weekes, A., & Breen C. (1997). Acidity and catalytic activity of mildly acid-treated Mg-rich montmorillonite and hectorite. *J.Chem. Soc., Faraday Trans*, Vol 23, 4207-4210
- Komadel, P. (1999). Structure and Chemical characteristics of modified clays. *Institute of Inorganic chemistry, Slovak Academy of Sciences*, 3-18
- Laird, D.A., Fenton T.E., & Scott, A.D. (1989). Evaluation of the Alkylammonium method of determining layer charge. *Clays and Clay Minerals*, Vol.37, No 1, 41-46
- Laird, D.A. (2006) Influence of layer charge on swelling of smectites. *Applied Clay Science*, Vol 34, 74-87
- Laird, D. & Fleming, P. (2008). Analysis of layer charge, cation and anion exchange capacities and synthesis of reduced charge clays. *Soil Science Society of America, Madison, USA Chapter 16*, 485-508
- Noyan, H., Önal, M., & Sarikaya, Y. (2007). The effect of sulphuric acid activation on the crystallinity, surface area, porosity, surface acidity and bleaching power of bentonite. *Food Chemistry*, Vol 105, 156-163
- Vali, H., Hesse, R. & Martin, F.R. (1994). A TEM-based definition of 2:1 layer silicates and their interstratified constituents. *American Mineralogist*, Vol 75, 644-653