## Processing of a Zimbabwean petalite to obtain lithium carbonate

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#### ABSTRACT

Processing of petalite concentrate from the Bikita deposits in Zimbabwe for production of high purity Li<sub>2</sub>CO<sub>3</sub> has been studied. XRF and ICP-OES analysis showed that the concentrate consists of oxides of Li, Si and Al as major components, with an average Li<sub>2</sub>O content of 4.10 %. XRD examination confirmed that the sample is a petalite. Processing of the petalite involves roasting the pre-heated concentrate with concentrated H<sub>2</sub>SO<sub>4</sub> followed by water leaching of the resulting Li<sub>2</sub>SO<sub>4</sub>, solution purification and precipitation of Li<sub>2</sub>CO<sub>3</sub>. The effects of roasting temperature, stirring speed, solid to liquid ratio, leaching temperature and time on the lithium dissolution are reported. The dissolution rates are significantly influenced by roasting temperature and stirring speed. Water-washed lithium carbonate with a purity of 99.21 % (metal basis) was produced. Synthesised and commercial Li<sub>2</sub>CO<sub>3</sub> samples were characterised and compared using X-ray diffraction (XRD) and thermogravimetric analysis (TGA).

Keywords: Petalite; Lithium; Lithium extraction; Lithium carbonate; Bikita Minerals

### 1. Introduction

Lithium is found in minerals, lake brines, clays, seawater and oil residues. The commercial sources of lithium are minerals, brines and seawater (Amouzegar et al., 2000). The lithium minerals of economic importance are spodumene (LiAlSi<sub>2</sub>O<sub>6</sub>), petalite (LiAlSi<sub>4</sub>O<sub>10</sub>), lepidolite ((Li,Al)<sub>3</sub>(Al,Si)<sub>4</sub>O<sub>10</sub>(F,OH)<sub>2</sub>), amblygonite (Li,Al(F,OH)PO<sub>4</sub>) and zinnawaldite (K(Li,Al,Fe)<sub>3</sub>(Al,Si)<sub>4</sub>O<sub>10</sub>F<sub>2</sub>) (Kondás and Jandová, 2006; Wietelmann and Bauer, 2003; Demirbaş, 1999).

In Zimbabwe, petalite deposits are found in the Bikita area east of Masvingo. Pegmatite deposits in Zimbabwe contain significant amounts of lithium and are one of the world's largest lithium deposits (Veasey, 1997). Zimbabwe is among the largest producers of lithium concentrate in the world. Jaskula (2010) reported that major producers of lithium concentrates are Australia, Canada, Zimbabwe, and Portugal. Lithium deposits at Bikita were estimated at 23,000 mt Li in 2009 (Jaskula, 2010). Other minor lithium deposits in Zimbabwe occur in the Insiza, Matobo, Mazoe, Mutoko, Harare, Mutare, Kamativi and Hwange districts (Cooper, 1964). Petalite is one source from which lithium is produced commercially (Garrett, 2004).

Lithium carbonate is the precursor for all other lithium compounds including lithium chloride, a raw material for lithium metal production (Jandová et al., 2010). Its oldest application is in the glass and ceramic industry. The addition of lithium carbonate in glass and ceramic production lowers the process melting point, reduces energy consumption, increases furnace refractory life, improves the strength of the glass product, reduces the coefficient of thermal expansion as well as viscosity (Ebensperger et al., 2005; Garrett, 2004). Lithium and its compounds have recently found use in energy storage devices such as rechargeable lithium- ion batteries (Brandt and Haus, 2010).

In the aluminium industry, lithium carbonate is added during electrolysis of alumina  $(Al_2O_3)$  to lower the melting point of the cryolite bath, allowing a lower operating temperature for the cells, increasing the electrical conductivity, decreasing the bath viscosity, reducing the consumption of cryolite, reducing the consumption of anode carbons, and reducing fluorine emissions from the electrolytic cells (Ober, 2007; Ebensperger et al., 2005; Nicholson, 1978). High-purity lithium carbonate is used in the treatment of manic-depressive psychosis (Ober, 2007) and production of electronic grade crystals of lithium niobate (Amouzegar et al., 2000).

Processing of lithium minerals relies on chemical roasting of the concentrate with the use of sulphuric acid (Kondás and Jandová, 2006; Wietelmann and Bauer, 2003), hydrochloric acid (Wietelmann and Bauer, 2003), limestone and or lime (Wietelmann and Bauer, 2003),

sodium and or potassium salts and calcium sulphate with calcium hydroxide (Kondás and Jandová, 2006). The objective of all the processing techniques is to convert lithium minerals into soluble form. Roasting of lithium minerals with H<sub>2</sub>SO<sub>4</sub> gives a high yield of lithium and has favourable energy consumption compared with other processes (Wietelmann and Bauer, 2003). In addition, H<sub>2</sub>SO<sub>4</sub> is cheaper and easier to handle than HCl. Roasting with HCl is not attractive because of complexity of purification. Wietelmann and Bauer (2003) reported that the alkaline process and other processes have a relatively high energy requirement and give a lithium yield appreciably below that of the sulphuric acid process.

Jaskula (2008) reported that the global market for lithium-ion batteries has increased by more than 20 % per year in the past few years and that the use of lithium batteries in upcoming electric and hybrid vehicles could further increase demand for the metal. The growing demand for lithium to meet the raw material needs of the energy storage devices is one of the reasons for studying the upgrading of Zimbabwean petalite into Li<sub>2</sub>CO<sub>3</sub>. In addition there is very limited work or published data, particularly on the chemical leaching and processing of Zimbabwean petalite. One major work is that of Cooper (1964), which was preliminary in nature and was on the geology of Bikita pegmatite and petalite characterisation. The work presented here is thus expected to be of interest.

This study gives detailed data on the characterisation, concentrate roasting with  $H_2SO_4$ , subsequent water leaching of the resulting  $Li_2SO_4$ , solution purification, and precipitation of  $Li_2CO_3$ .

## 2. Materials and methods

#### 2.1. Materials

Laboratory tests were carried out with a petalite concentrate, as received from Bikita Minerals (Pvt) Ltd in Zimbabwe. A multi-element standard (ICP grade) and analytical grades Na<sub>2</sub>CO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub>, HCl, CaCO<sub>3</sub> and Ca(OH)<sub>2</sub> from Merck Chemicals (Pty) Ltd were used. Distilled water was used in the preparation of all aqueous solutions.

#### 2.2. Instrumentation

The following instruments were used in this study: for X-ray fluorescence (XRF) a Thermo ARL9400 XP instrument; for inductively coupled plasma optical emission spectrometry (ICP-OES) a SPECTRO ARCOS; for X-ray diffractometry (XRD) a PANalytical X'Pert PRO diffractometer; for mass measurements a Mettler PM2000MC balance; for pH measurements a pH meter cripson GLP 21; and for thermogravimetric analysis (TGA) a TA instrument DSC-TGA SDT Q600.

#### 2.3. Methods

#### 2.3.1. Particle size analysis

Dry sieving was performed on a 100.00 g sample of petalite concentrate to determine particle size distribution using a Fritsch-sieve shaker, with universal test sieves SABS ISO 310. Masses were determined with an analytical balance.

#### 2.3.2. Characterisation of the concentrate

Elemental analysis of the concentrate was done using XRF and ICP-OES. The petalite sample was dissolved completely using a method adapted from Stoch (1986) to determine % Li<sub>2</sub>O content. 0.5 g of petalite was decomposed by HF-H<sub>2</sub>SO<sub>4</sub> followed by dissolution of soluble salts by boiling with water. XRD analysis was performed using Fe filtered CoK<sub> $\alpha$ </sub> as a radiation source. The phases were identified using X'Pert Highscore Plus software. The semi-quantitative phase amounts (weight %) was estimated using the reference intensity method in the X'Pert Highscore Plus software.

### 2.3.3. Calcination and roasting of petalite

The finely powdered petalite concentrate was first heated in a laboratory furnace at 1100 °C for 2 hrs with exact temperature regulation. Heating petalite to high temperatures

results in an irreversible phase change to a  $\beta$ -spodumene-SiO<sub>2</sub> solid solution which is more reactive to acid and base attack (Wietelmann and Bauer, 2003; Kamiensiki et al., 2005; Garrett, 2004). A weighed amount of ground pre-heated petalite sample was mixed with concentrated H<sub>2</sub>SO<sub>4</sub> in a porcelain crucible. An amount of acid 15 % in excess of the lithium equivalent was added. The roasting temperatures were varied from 200-300 °C. The mixture in a crucible was placed in a laboratory furnace when the desired temperature was attained. On completion of the roasting process the samples were removed, cooled at room temperature and ground to less than 75 µm. Reaction of  $\beta$ -spodumene with H<sub>2</sub>SO<sub>4</sub> is shown in equation (1) (Mcketta, 1988).

$$Li_{2}O.Al_{2}O_{3}.4SiO_{2(s)} + H_{2}SO_{4(conc)} \rightarrow Li_{2}SO_{4(s)} + Al_{2}O_{3}.4SiO_{2(s)} + H_{2}O_{(g)}$$
(1)

#### 2.3.4. Leaching studies

This study discusses the conditions such as temperature, leaching time, solid/liquid ratio and stirring rate which affect leaching of lithium. Leaching experiments were performed in a 600 mL flat-bottomed pyrex glass beaker as a reactor. Accurately measured volume of distilled water (150 mL) was added to the reactor and heated to the desired temperature within ±1 °C by a thermostatically controlled hot plate magnetic stirrer at atmospheric pressure. A teflon coated stirring bar was used. When the temperature reached the pre-set value and remained stable, the acid roasted product was added to the reactor and heated together with the water. During leaching, samples were withdrawn at selected time interval. After various leaching experiments had been conducted, ICP testing was done to calculate the extraction rate.

### 2.3.5. Solution purification

Acid roasting and leaching of petalite concentrate is not entirely selective as co-dissolution of impurities also occurs. The resulting leach liquor contains aluminium, calcium, magnesium, iron and other impurities which can be removed by pH control and addition of a carbonate (Averill and Olson, 1977). CaCO<sub>3</sub> was added little by little to the acidic leach solution at laboratory temperature to obtain a pH of 5.5-6.5. At this pH Fe and Al were precipitated and the cake was filtered and washed with water. Mg was precipitated using  $Ca(OH)_2$  and filtered. The filtrate was treated with  $Na_2CO_3$  at 25 °C which produced a pH between 11-12.  $Na_2CO_3$  equivalent to the amount of calcium present was added. This precipitated the Ca, which was removed by filtration. The flow sheet of the purification procedure is shown in Fig.1. A pH meter was used during the procedure for the pH value analysis. ICP-OES was used to determine the concentration of lithium and impurities at each stage. Reproducibility of the process was tested by running the experiments in triplicate. Errors for each were below 3 %. The purified solution was adjusted to pH 7-8 using  $H_2SO_4$  (1 mol L<sup>-1</sup>) in order to precipitate the carbonate (Kamiensiki et al., 2005).

### [INSERT Fig. 1]

#### 2.3.6. Precipitation of lithium from leach liquor

The purified leach solution was evaporated to about 50 % of its original volume and filtered to remove the calcium residue. The evaporation process was continued until the concentration of Li was more than 11 g L<sup>-1</sup>. A hot saturated solution of Na<sub>2</sub>CO<sub>3</sub> was added drop-wise to the filtrate at 95-100 °C giving a white precipitate of Li<sub>2</sub>CO<sub>3</sub>. The precipitate was filtered and washed thoroughly with water (95-100 °C) to remove residual Na<sub>2</sub>SO<sub>4</sub> and excess Na<sub>2</sub>CO<sub>3</sub>. The water-washed Li<sub>2</sub>CO<sub>3</sub> was dried at 250 °C. The flow sheet for the lithium recovery procedure is shown in Fig. 2. Changes of the lithium and impurity concentration during the processing of leach liquor were recorded by means of ICP. Na<sub>2</sub>SO<sub>4</sub> in the mother liquor was crystallised by chilling the solution at 0-8 °C followed by pressure filtration (Garrett, 2004). The filtrate which contains some lithium and residual Na<sub>2</sub>CO<sub>3</sub> was recycled to the evaporator.

## [INSERT Fig. 2]

### 2.3.8. Characterisation of lithium carbonate

The dried  $Li_2CO_3$  was subjected to phase, thermal and chemical analysis. Phase and thermal analysis was done by XRD and TGA respectively. XRD analysis was performed using X-ray diffractometer with Fe filtered  $CoK_{\alpha}$  as a radiation source. During TGA investigations,

samples of (10 ±1.5) mg were measured in a temperature range of 25-1090 °C at a rate of 20 °C min<sup>-1</sup> in flowing N<sub>2</sub>. Comparison was made for the synthesised and commercial Li<sub>2</sub>CO<sub>3</sub> from Alfa Aesar, a Johnson Matthey Company. Chemical analysis was done by means of ICP to determine the purity of Li<sub>2</sub>CO<sub>3</sub> 0.50 g of processed Li<sub>2</sub>CO<sub>3</sub> was dissolved with concentrated HCl at 90 °C for 30 minutes and made up to 100 mL with distilled water in a volumetric flask. The solution was subjected to ICP analysis to determine the lithium and impurity concentration.

#### 3. Results and discussion

### 3.1. Particle size distribution

The particle size distribution of as-received petalite concentrate is shown on Table 1. It is revealed that the concentrate is fine with more than 91 % of the concentrate particles smaller than 75  $\mu$ m. Small mineral particle size enhances the leaching rate since larger surface area of the mineral particle is exposed to chemical attack. The influence of mineral particle size thus cannot be overlooked in hydrometallurgical treatment.

#### [INSERT Table 1]

#### 3.2. Mineralogical composition of the concentrate

The mineral phases recorded from the XRD examination of the lithium pegmatite is provided in Table 2. XRD examination confirmed that the sample is a petalite. Furthermore the XRD data showed the presence of associated minerals, viz. spodumene, bikitaite, lepidolite, quartz, albite and microcline.

[INSERT Table 2]

#### 3.3. Elemental analysis

The results of the XRF and ICP-OES showed Li<sub>2</sub>O (4.10 %), SiO<sub>2</sub> (76.11 %) and Al<sub>2</sub>O<sub>3</sub> (17.76 %) as major oxides. The chemical composition of petalite concentrate is shown on Table 3.

### [INSERT Table 3]

#### 3.4. Roasting studies

#### *3.4.1. Effect of roasting temperature and time*

A number of experiments were run by varying the roasting temperature in the range of 200-300 °C. The results of this investigation are presented in Fig. 3. An increase in temperature from 200 to 300 °C brings about an increase in dissolution of lithium, which is attributed to the fact that raising the temperature will increase the reaction rate. From the graph maximum extraction was achieved at 300 °C in 60 minutes.

[INSERT Fig. 3]

#### 3.5. Leaching studies

#### *3.5.1. Effect of stirring rate*

The effect of agitation on the dissolution of lithium was investigated in water at 50 °C, using stirring speeds of 0-400 rpm with solid/liquid ratio of (1/7.5 g/mL). The results in Fig. 4 show that stirring speed has a significant effect on the dissolution of lithium. Agitation is required to keep the solids in suspension as well as to exclude the influence of external mass transfer (diffusion through liquid boundary layer). Above 320 rpm, the stirring rate no longer influences the lithium dissolution. On the basis of the data, subsequent experiments were carried out at a stirring speed of 320 rpm.

[INSERT Fig. 4]

## 3.5.2. Effect of solid/liquid ratio

The effect of solid/liquid (g mL<sup>-1</sup>) ratio was also examined. The results of different solid to liquid ratios are illustrated on Fig. 5. It can be seen from Fig. 5 that the percentage lithium extracted decreases with increasing solid-to-liquid ratio. A decrease in solid to liquid ratio decreases the viscosity of the system and as a result decreases the mass transfer resistance in liquid-solid interface. The solid-to-liquid ratio of (1/7.5 g/mL) gave the highest dissolution and was kept for further use in this study.

## [INSERT Fig. 5]

## 3.5.3. Effect of leaching temperature and time

The effect of temperature on the dissolution of lithium was investigated with solid/liquid ratio of (1/7.5 g/mL), stirring speed of 320 rpm at temperatures 50 and 90 °C. From the results in Fig. 6, it can be shown that temperature has very little effect on the extraction of lithium. The highest extraction of around 97.30 % was realised after 60 minutes for all the temperatures. To avoid significant loses of liquid by evaporation the temperature of 50 °C was used for subsequent experiments.

#### [INSERT Fig. 6]

## 3.6. Solution purification

Changes of lithium and impurity concentration during processing leach liquors are described in Table 4. It is indicated from the table 4 that  $CaCO_3$  is effective in removing Al and Fe by precipitation. The same implies to precipitation of Mg and Ca with  $Ca(OH)_2$  and  $Na_2CO_3$  respectively. Lithium losses during the purification process were 7.64 %.

[INSERT Table 4]

#### 3.7. Precipitation and characterisation of lithium carbonate

Precipitation of Li<sub>2</sub>CO<sub>3</sub> was done at 95-100 °C because the solubility of lithium carbonate decreases with increasing temperature (Wietelmann and Bauer, 2003). Lagos and Becerra (2005) reported that precipitation with sodium carbonate is the most effective since sodium sulphate that also precipitates is eliminated by washing with hot water. Lithium carbonate is precipitated from an aqueous lithium solution using Na<sub>2</sub>CO<sub>3</sub> according to the following reaction (2):

$$Li_2SO_{4(aq)} + Na_2CO_{3(aq)} \rightarrow Li_2CO_{3(s)} + Na_2SO_{4(aq)}$$
(2)

Analytical results show that about 86.00 % of the lithium was recovered as a precipitate relative to the concentrated leach liquor.

The thermogram for both synthesized and commercial  $Li_2CO_3$  is shown in Fig. 7. The thermogravimetric curve for the synthesised  $Li_2CO_3$  is almost similar to that of commercial powder. Both thermograms show that below 700 °C the curves are smooth and the mass changed very little. At about 725 °C  $Li_2CO_3$  starts to decompose releasing  $CO_2$  according to the following reaction (3):

$$\text{Li}_2\text{CO}_3 \rightarrow \text{Li}_2\text{O} + \text{CO}_2 \tag{3}$$

Mass losses of 58.43 % and 60.83 % for synthesised and commercial  $Li_2CO_3$  respectively were recorded, these corresponds well to the expected stoichiometrical mass loss of 59.55 %.

[INSERT Fig. 7]

A comparison between X-ray pattern for synthesised and commercial  $Li_2CO_3$  powder is shown in Fig. 8. As can be seen, the X-ray patterns for the synthesised and commercial powder are very similar. The results also indicate that no other phases were identified except the lithium carbonate. A slightly difference in peak intensities might be due to the difference in concentration of the powders.

## [INSERT Fig. 8]

The purity of the synthesised product is indicated in Table 5. From the results, the purity of the recovered powder is 99.21 % (metal basis). The analysis shows that the major impurity is Na which may be attributed to the residual  $Na_2SO_4$  and excess  $Na_2CO_3$  that were not removed completely during the washing process.

[INSERT Table 5]

## 4. Conclusions

The XRF and ICP-OES technique showed that the petalite concentrate consists of oxides of Li, Si, and Al as major components, with an average Li<sub>2</sub>O content of 4.10 %. XRD examination on the other hand confirmed that the predominant mineral is petalite. It also revealed that presence of associated minerals, viz. spodumene, lepidolite, bikitaite, quartz, albitite and microline. The extraction rates of lithium are significantly influenced by roasting temperature, stirring speed and solid liquid ratio. Maximum extraction of 97.30 % was realised at a roasting temperature of 300 °C, solid/liquid ratio (1/7.5 g/mL), stirring speed of 320 rpm at leaching temperature of 50 °C in 60 minutes. The experimental results show that purity of Li<sub>2</sub>CO<sub>3</sub> produced by this process was 99.21 % (metal basis). XRD of the synthesised powder indicate that no other phases were present except of zabuyelite (Li<sub>2</sub>CO<sub>3</sub>). The thermograms for synthesised and commercial Li<sub>2</sub>CO<sub>3</sub> where almost identical with both powder starts decomposing around 725 °C.

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Fig. 1. Flow sheet for leach liquor purification.



Fig. 2. Flow sheet for precipitation of  $Li_2CO_3$ .



**Fig. 3.** Effect of roasting temperature on the extraction rate of lithium. Experimental conditions: leaching temperature 50 °C, leaching time 60 minutes, stirring speed 320 rpm and solid/liquid ratio (1/7.5 g/mL).



**Fig. 4.** Effect of stirring speed on the extraction rate of lithium. Experimental conditions: acid roasting temperature 300 °C, acid roasting time 60 minutes, leaching temperature 50 °C, leaching time 60 minutes and solid/liquid ratio (1/7.5 g/mL).



**Fig. 5:** Effect of solid/liquid ratio on the extraction rate of lithium. Experimental conditions: acid roasting temperature 300 °C, acid roasting time 60 minutes, leaching temperature 50 °C, leaching time 60 minutes and stirring speed 320 rpm.



**Fig. 6.** Effect of leaching temperature on the extraction rate of lithium. Experimental conditions: acid roasting temperature 300 °C, acid roasting time 60 minutes, stirring speed 320 rpm, leaching time 60 minutes and solid/liquid ratio (1/7.5 g/mL).



Fig. 7. Thermograms of synthesised and commercial  $Li_2CO_3$ .



Fig. 8. X-ray patterns of synthesised and commercial  $Li_2CO_3$ .

Particle size distribution.

Particle	Weight	Weight retained	Cummulative
size (µm)	retained	%	% passing size
75	8.136	8.14	91.86
53	26.732	26.73	65.13
45	16.095	16.10	49.04
38	42.139	42.14	6.90
-38	6.898	6.90	0.00

Semi-quantitative mineral content of the petalite (XRD analysis).

Mineral phase	(Wt. %)
Petalite (LiAlSi <sub>4</sub> O <sub>10</sub> )	57.0
Spodumene (LiAlSi <sub>2</sub> O <sub>6</sub> )	3.0
Quartz (SiO <sub>2</sub> )	4.0
Albite (NaAlSi <sub>3</sub> O <sub>8</sub> )	17.0
Bikitaite (LiAlSi <sub>2</sub> O <sub>6</sub> (H <sub>2</sub> O))	2.0
Lepidolite (K(Al_{0.62}Li_{0.38})_2Li_{0.92}Si_4Al_{0.42}O_{10}(OH)_{0.485}F_{1.51})	10.0
Microline (KAlSi <sub>3</sub> O <sub>8</sub> )	7.0

Component (%)	ICP-OES	XRF (Normalised)	Combined	
SiO <sub>2</sub>	-	79.37	76.11	
$Al_2O_3$	-	18.52	17.76	
Li <sub>2</sub> 0	4.10	-	4.10	
Fe <sub>2</sub> O <sub>3</sub>	-	0.05	0.05	
Na <sub>2</sub> O	-	0.57	0.54	
K <sub>2</sub> 0	-	0.47	0.45	
CaO	-	0.12	0.11	
Rb <sub>2</sub> O	-	0.09	0.09	
P <sub>2</sub> O <sub>5</sub>	-	0.03	0.03	
Cs <sub>2</sub> 0	-	0.03	0.03	
MnO	-	0.02	0.02	
F	-	0.01	0.01	
CeO <sub>2</sub>	-	0.09	0.09	
Cr <sub>2</sub> O <sub>3</sub>	-	0.00	0.00	
MoO <sub>3</sub>	-	0.01	0.01	
LOI* (1000 °C)		0.62	0.59	
Total		100.00	100.00	

Chemical composition of the petalite (XRF, ICP-OES analysis).

\* Loss on ignition

Solution	pH	Elemental concentration (mg L <sup>-1</sup> )							
		Li	Са	К	Na	Al	Si	Fe	Mg
Original leach liquor	0.95	5716.82	145.14	451.60	1422.70	10.05	33.21	37.95	12.24
Leach liquor on addition of	10.25	5500.72	559.50	147.80	1015.30	<0.025	<0.01	<0.01	<0.01
$(CaCO_3 and Ca(OH)_2)$									
Leach liquor on addition of	11.56	5227.65	11.36	131.90	2163.30	< 0.025	<0.01	<0.01	<0.01
(Na <sub>2</sub> CO <sub>3</sub> )									
Concentrated leach liquor	8.00	11998.62	7.29	492.93	5362.32	0.04	< 0.01	< 0.01	<0.01
Mother liquor (filtrate)		1120.15							

## Concentration of elements in processed solutions.

Content of impurities in lithium carbonate.

Li <sub>2</sub> CO <sub>3</sub>	Purity (%)	Content of major impurities (%)						
		Ca	K	Na	Al	Si	Fe	Mg
	99.21	<0.01	0.031	0.760	<0.01	<0.01	<0.01	<0.01