Analysis of raw materials and assessment of the reactivity of china clay from Malawi for LC³ application

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Summary:
Malawi is a small land locked country in Central Africa. With a very low human development index, it generally depends on external support for its economic activity. Various types of minerals are found in Malawi which remain unexploited till date. Like other African countries, the major construction material in Malawi is as usual cement with burnt clay bricks and steel being the others. The consumption of cement in Malawi was around 370,000 tonnes in 2012. Majority of the cement produced is portland with clinker imported from neighbouring countries. Due to absence of knowledge and lack of data on minerals available, the production and supply of portland pozzolana cement remains largely unexplored. No systematic study has yet been undertaken to understand the prospects of portland pozzolana cement either in Malawi or even in Africa. The present study understands the availability of cement producing minerals in Malawi with collection and characterization of limestone, gypsum and china clay. This assessment study is to understand the preliminary feasibility of introducing pozzolana cement using calcined china clay and crushed limestone with low levels of clinker termed as Limestone Calcined Clay Cement (LC$^3$).

Secondary literature survey shows the availability of large quantities of naturally occurring minerals in Malawi to produce the LC$^3$ cement. Raw materials suitable for LC$^3$ production was procured with the help from Ceramic Department, Government of Malawi. Characterization studies and reactivity assessment of china clay and its calcined form proves that a high strength cement of existing 42.5 Grade can be made with available china clay from Linthipe. However more extensive testing and pilot production trials needs to be carried out before any definitive conclusions can be made. Based on the demand of cement in 2012 and the growth pattern it can be assumed that the present cement consumption is around 450,000 tonnes at present. Quite a substantial amount of cement is imported also. If the entire production of cement shifts from portland to LC$^3$ then it will require around 130,000 tonnes of china clay per year. Considering only the reserves in Linthipe, Malawi can sustain more than 100 years the production and consumption of LC$^3$ reducing the dependency on imported clinker from neighbouring countries. In addition to use of local natural resources, through the use of LC$^3$ Malawi can reduce the CO$_2$ emissions by around 180,000 tonnes per year considering the present quantity of consumption. Additionally production of LC$^3$ can create large number of new jobs both in the small scale mining sector and related ancillaries.
1. Introduction

With increasing affluence and development, there has been a spurt in infrastructural activities across the world. Most of the concentration of construction activities are in the developing countries e.g. China, India, Brazil, Russia and South Africa. The demand of infrastructural needs is also expected to increase the demand of cement which is expected to double by 2050. Almost 80% of this demand is projected from the developing countries\(^1\). Notwithstanding the fact that demand for cement will be primarily in developing countries, the under-developed and developing nations of Africa are not far behind.

Global sustainability with issues such as poverty and global warming are requiring urgent attention. However the most basic demand is the need of shelter. With increased growth and development of Africa, there will be emphasis on infrastructure development alongwith shelter. The need of both shelter and infrastructure will be aggravated by increased urbanization. It is seen that with an average annual growth of 3.4%, the population in several Sub-Saharan African cities will rise by 25% by 2025 (Figure 1). Going with this trend, by 2050, 60% of Africa’s population will be urbanized.

Going by this trend, the major challenge will be to provide housing and infrastructural needs for

\[\text{Figure 1: Urban population distribution and forecast in Africa}\]

the urban population, where the current population of 1.2 billion people is expected to double until 2050 and reach 4 billion by 2100. The challenge is further increased by the fact that current cement and concrete based building has a carbon footprint, which is not compatible with required reductions in carbon emissions.

It has been calculated that for every 20 tons of CO\(_2\) that enter the atmosphere due to human activity, one comes from the kilns of cement factories\(^1\). This is quite surprising that till today concrete is one of the most natural and widely used material in the earth with lowest environmental impact. However the sheer quantity of the concrete used in the world makes it\(^2\) the top contributor of human CO\(_2\) emissions.

\(^1\) Cement accounts for 3-7% of man-made CO\(_2\) emissions.

\(^2\) Every year there is around 2m\(^3\) of concrete produced for every person on the planet
The drive for development and increasing urbanization will generate an ever-increasing need for construction materials, especially in developing African countries. The low cost, low environmental footprint and ease of use of concrete compared to other construction materials make it the preferred material to meet the ever-increasing demand. For example the production of cement in China has increased 3-4 times just in the last decade and is now 60% of world production.

Within Africa also there is a vast difference in cement consumption across the regions as shown in the Figure 2.

![Figure 2: Per capita consumption vs urbanization of African countries (2008)](image)

The per capita consumption of cement varies quite largely in African countries. As per 2008 figures, North Africa was the highest with 538 kg, followed by Southern Africa at 149 kg, West Africa at 91 kg, East Africa at 50 kg and Central Africa at 39 kg. With increased emphasis on development especially in the Eastern and Central Africa, this equation is going to change drastically. This creates two main challenges for the future, which are:

- How can current resources meet the projected increasing demand?
- How can the environmental impact (CO₂ emissions) of this production increase be mitigated?

Thus any improvement in the sustainability of cement production can have a huge impact on resource efficiency and CO₂ released to the atmosphere.

The main ingredients of cement are calcium carbonate (limestone) and clay. These are also among the most abundant materials in the earth’s crust, and they have a well-balanced geographic distribution. Therefore, Portland cement can be considered a “local” material, since it can be produced almost anywhere in the world. However, in some areas existing quarries are being exhausted at a rapid rate. A key aspect of LC3 technology is the potential to produce up to twice as much cement from each ton of clinker and to better use limestone sources not suitable for clinker production.

The majority (60%) of CO₂ emissions associated with clinker production (the main ingredient in cement) come from the decarbonation of limestone (“chemical” CO₂). The consumption of fuels
makes up most of the remainder. In the end product, concrete, and accounting for aspects like transportation, the clinker component constitutes up to 90% of the CO\textsubscript{2} emissions.

Strategies to lower the environmental impact of cement manufacturing are already applied by the industry and their future potential has been evaluated in detail in by the IEA (International Energy Agency) for the Cement Sustainability Initiative (CSI) study of the World Business Council for Sustainable Development (WBCSD).

Today the three most important approaches are:

- Improvement of energy efficiency
- Use of biofuels and other alternative fuels and
- Replacement of clinker by substitute materials or supplementary cementitious materials (SCMs). Materials used include fly ash, various slags and natural pozzolans.

Beyond these approaches the CSI study imagines the remaining reduction in CO\textsubscript{2} to come from Carbon Capture and Storage (CCS) which is now widely thought to be, at best, very expensive (increasing cement prices 2-4 times). Thus this is presently and also in the future not relevant for consideration especially under African conditions. Gains from alternative fuels are projected to increase, but remain a fairly modest proportion of the total. The projected impact of the third option, substituting clinker by SCMs is limited by the forecast supply of commonly used SCMs, notably slag and fly ash. For example, although slag can substitute up to 90% and typically 70% of clinker in blends, the worldwide amount of slag available is only around 5% of the amount of clinker produced. Fly ash although available in larger amounts (around 30% of clinker worldwide, but absent in many countries) is of variable quality and much is unsuitable for cement production. So the option of clinker substitution could go much further if new sources of SCMs were available.

Low Carbon Cement can now fill the gap above. This allows a clinker substitution of around 50-60%, by a synergetic combination of calcined clay and limestone. This has been demonstrated in the collaborative research between IIT Delhi and TARA at India and at EPFL, Switzerland.

2. **Raw materials for production of LC\textsuperscript{3}**

It has been widely known that the basic components of Portland cement are clinker and gypsum. Additionally clinker is produced through a combination of clay and limestone. In India to reduce the clinker factor supplementary cementitious materials e.g. fly ash and blast furnace slag are commonly used. In fact a majority of the cement produced in India is PPC or Portland Pozzolana Cement made up of atleast 25% substitution of clinker by fly ash. However above a threshold substitution of about 30%, these materials reduce the mechanical properties, particularly at early age \cite{2}. On the other hand fly ash and blast furnace slag are limited to local availability retarding their widespread use. Consequently, alternative sources of SCM’s such as calcined clays are of interest. These are widely available in the earth’s crust and can easily be dehydroxilated at temperatures ranging between 700 – 800°C to produce metakaolin \cite{3-4}. Metakaolin demonstrates excellent pozzolanic properties \cite{5-8} and is one of the major raw materials of producing LCC.

Limestone is also an important raw material for use in production of cement. Fine limestone is also commonly added to cement and it is established that limestone additions up to around 5% can react with cement and enhance most properties \cite{9-11}. It has been shown that 45% of substitution by 30% metakaolin and 15% of limestone gives better mechanical properties at 7 and 28 days that the 100% Portland Cement reference. Results show that calcium carbonate reacts with alumina from the metakaolin, forming supplementary AFm phases and stabilizing ettringite \cite{2}. It has also been shown that gypsum addition should be carefully balanced when
using calcined clays because it considerably influences the early age strength by controlling the rapid reaction of aluminates.

In this study we map the availability of raw materials suitable for production of LC³ with particular reference to china clay availability in Malawi. This is expected to give us a broad idea on the possibility of introducing LC³ technology in Malawi and therefore make available good quality cement at an affordable rate.

3. Constraints of the mineral industry in Malawi

The development of mineral resources in Sub-Saharan Africa has been primarily directed towards the extraction and export of "metallic" ores, i.e. gold, copper, nickel, chromite uranium, diamonds, etc. As there is a ready and strategic demand for these minerals in industrialized countries, they attract the most investments from international mining houses, in contrast to the "industrial minerals" which are most abundant in Malawi. Secondly, even those minerals with export potential, such as rare earth (RE), vermiculite, bauxite and uranium, have rarely been aggressively promoted. Hence there was very little private sector exploration taking place prior to the 1980s.

Malawi's abundant resources of limestone, vermiculite, ceramic clay, graphite, pyrite and pyrrhotite could contribute significantly to local industrial development. However the failure to develop these resources to date has been due to a number of factors:

- The development of industrial minerals often goes hand in hand with that of metallic ores. Ceramic clays, kyanite, and limestone are some of the key resources in the smelting of ores, production of refractories. However, at present there is no coordinated effort to monitor potential demand and develop strategic plans for production of mineral-based commodities.

- Traditionally, Malawian manufacturers depend on the supply of raw materials from their mother companies in Europe, the U.S.A., Japan or South Africa. The demand for local raw materials is a recent phenomenon, thrust on the industry by economic difficulties.

- Often educated people are reluctant to take up business initiatives in the mining sector, and small scale mining is left to those who least understand the need for improved technologies. Consequently, inefficient and poor quality production operations are common in the small-scale mining sector.

The biggest obstacles to overcome in order to develop industrial minerals seem to be a lack of technological methods and practices; an absence of coordinated research efforts; and the inability to convert research information and results into realistic commercial ventures. Mineral commodities are only one part of the required raw materials that are necessary for the production of consumable goods. There is, however, a lack of an integrated approach in determining actual demand of raw materials.

4. Raw material availability in Malawi

No recent data exists on the mineral resources in Malawi. The only reliable data is based on the USAID report published in 1990. All the data given below has been extensively taken from this study available till date. This study describes the full range of inventories of "industrial mineral" resources found in Malawi which can be exploited in the immediate future by small to medium scale Malawian entrepreneurs. These resources include: limestone and dolomite; vermiculite; gypsum; glass sands; kaolinitic ceramic clays, graphite, gemstones and
ornamental stones; salt; pyrite/pyrrhotite; talc; rock phosphates; brick clays and kyanite. Coal, although an energy resource, is also included for its potential as a source of energy in the mining sector. However the data taken will be restricted only to those required for LC³ use i.e. limestone, kaolinitic clays and gypsum.

Industrial minerals are generally the least understood of all natural resources in developing countries. Consequently, most industrial minerals remain unexploited, while a high priority has been given to development of metallic and energy minerals for export to industrialized countries. Many Malawians do not perceive industrial minerals as a potentially lucrative investment area and thus do not invest. At present, over 80% of the consumption of industrial minerals takes place in industrialized countries. On the contrary even with vast resources, Malawi still imports a variety of commodities with a high proportion of industrial mineral materials in them. These commodities could have been easily manufactured locally by small scale industries.

It is hoped that the results of this study will throw up possibilities of large scale mining and utilization of mineral resources, thereby creating livelihood and economic opportunities.

4.1. Limestone

Malawi is blessed with large quantities of limestone and dolomite. Limestone and dolomite resources of Malawi are estimated to be over 800 million tonnes, mainly found as metamorphic marbles in the southern part of Malawi (Figure 3). All the resources may be divided into 5 types as follows:

- Metamorphic marbles: (The term marble is used in the geologic sense, and not in the trade sense of a rock)
- Carbonatites: igneous rock vents enriched in sphyrites (calcites of undoubted igneous origin
- Sedimentary limestones
- Vein calcite
- Travertine, (light coloured) tufa and (porous) soil limestones.

The most widely used limestones and dolomites are the metamorphic marbles and sedimentary limestones, hence further discussions will mainly refer to these.

For Malawi limestone is a term reserved for rocks in which the carbonate fraction is composed primarily of the mineral calcite (CaCO₃), with the Magnesian (MgO) content less than 5%.

Dolomite is reserved for those rocks which are composed primarily of the mineral dolomite, CaMg(CO₃)₂, theoretically, with 21.9% MgO and 30.4% CaO.
Most of the limestone and dolomite deposits which have been located by drilling and/or trenching have only been partially evaluated. The reserves and quality of these deposits are presented in Tables 1 and 2. Exploitation of limestone and dolomite is greatest at Lirangwe, Chenkumbi, Kholombidzo and Changalume (for cement production). However depending on the geology of the country, there are still prospects of large occurrences of various grades of limestone.
Table 1: Total estimated reserve of limestone in Malawi and their quality

<table>
<thead>
<tr>
<th>Deposit location</th>
<th>District</th>
<th>Estimated resources (Mio tonnes)</th>
<th>Quality</th>
<th>% CaO</th>
<th>% MgO</th>
</tr>
</thead>
<tbody>
<tr>
<td>Changalume</td>
<td>Zomba</td>
<td>100</td>
<td></td>
<td>0.8</td>
<td>5.8</td>
</tr>
<tr>
<td>Chenkumbi</td>
<td>Machinga</td>
<td>300</td>
<td></td>
<td>6.3</td>
<td>-</td>
</tr>
<tr>
<td>Chikoa</td>
<td>Kasungu</td>
<td>25</td>
<td></td>
<td>0.3</td>
<td>2.5</td>
</tr>
<tr>
<td>Livwezi</td>
<td>Kasungu</td>
<td>18</td>
<td></td>
<td>0.5</td>
<td>1.8</td>
</tr>
<tr>
<td>Matope/Nkula</td>
<td>Blantyre</td>
<td>180</td>
<td></td>
<td>20.0</td>
<td>-</td>
</tr>
<tr>
<td>Lirangwe</td>
<td>Blantyre</td>
<td>5</td>
<td></td>
<td>19.1</td>
<td>3.2</td>
</tr>
<tr>
<td>Makoko/Malawi</td>
<td>Nsanje</td>
<td>20</td>
<td></td>
<td>5.6</td>
<td>3.8</td>
</tr>
<tr>
<td>Golomoti</td>
<td>Dedza</td>
<td>15</td>
<td></td>
<td>1.2</td>
<td>1.7</td>
</tr>
<tr>
<td>Uliwezi</td>
<td>Karonga</td>
<td>5</td>
<td></td>
<td>1.9</td>
<td>10.6</td>
</tr>
<tr>
<td>Ngana</td>
<td>Karonga</td>
<td>6</td>
<td></td>
<td>1.3</td>
<td>16.3</td>
</tr>
<tr>
<td>Mwesia</td>
<td>Karonga</td>
<td>15</td>
<td></td>
<td>1.6</td>
<td>18.9</td>
</tr>
<tr>
<td>Chilwa Island</td>
<td>Zomba</td>
<td>25</td>
<td></td>
<td>0.2</td>
<td>3.3</td>
</tr>
<tr>
<td>Kangankunde</td>
<td>Machinga</td>
<td>5</td>
<td></td>
<td>1.2</td>
<td>10.8</td>
</tr>
<tr>
<td>Kapiri</td>
<td>Machinga</td>
<td>10</td>
<td></td>
<td>21.7</td>
<td>2.1</td>
</tr>
<tr>
<td>Lulwe Hills</td>
<td>Nsanje</td>
<td>2</td>
<td></td>
<td>0.9</td>
<td>2.6</td>
</tr>
<tr>
<td>Lisungwe</td>
<td>Blantyre</td>
<td>40</td>
<td></td>
<td>20.2</td>
<td>2.2</td>
</tr>
<tr>
<td>Nsengwa</td>
<td>Mwanza</td>
<td>30</td>
<td></td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td><strong>Total estimated resources</strong></td>
<td></td>
<td><strong>801</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 2: Mineable reserve of limestone in Malawi (proven by rotary core drilling or trenching)

<table>
<thead>
<tr>
<th>Deposit location</th>
<th>District</th>
<th>Estimated reserves (Mio tonnes)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Changalume</td>
<td>Zomba</td>
<td>25</td>
</tr>
<tr>
<td>Chenkumbi</td>
<td>Machinga</td>
<td>10</td>
</tr>
<tr>
<td>Chikoa</td>
<td>Kasungu</td>
<td>5</td>
</tr>
<tr>
<td>Livwezi</td>
<td>Kasungu</td>
<td>5</td>
</tr>
<tr>
<td>Nkula</td>
<td>Blantyre</td>
<td>0.6</td>
</tr>
<tr>
<td>Matope bridge</td>
<td>Blantyre</td>
<td>0.7</td>
</tr>
<tr>
<td>Golomoti</td>
<td>Dedza</td>
<td>0.5</td>
</tr>
<tr>
<td><strong>Total estimated resources</strong></td>
<td></td>
<td><strong>46.8</strong></td>
</tr>
</tbody>
</table>

There are many uses of limestone and dolomite, but only a few of these are appropriate in Malawi:

- **Portland cement** - Limestone is quarried and burnt in kilns at Changalume. The clinker produced is then transported to Blantyre where, after the addition of gypsum, the resulting cement is bagged and sold.
- **Lime** – 70% of the lime produced in Malawi is building lime, but it is also used for a variety of purposes including for sugar production when there are import supply...
problems. The lime is mainly obtained from limestones with an MgO content below 5%.

- Dolomite is crushed and milled in the Blantyre area for agricultural purposes and for the production of scouring powders. The particles are milled to less than 80 mesh fineness.

At present, demand for chemical grade lime in Malawi is about 3,300 tonnes per annum. 95% of this is consumed by the sugar industry, and the balance by Lilongwe and Blantyre Water Boards, and Southern Bottlers. Production and consumption of building lime in the last three years were as follow:

**Table 3: Production and consumption of building lime**

<table>
<thead>
<tr>
<th>Year</th>
<th>Production (Tonnes)</th>
<th>Sales (Tonnes)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1985</td>
<td>NA</td>
<td>1462</td>
</tr>
<tr>
<td>1986</td>
<td>2681</td>
<td>2735</td>
</tr>
<tr>
<td>1987</td>
<td>1639</td>
<td>1544</td>
</tr>
</tbody>
</table>

Thus even with a 70% production of lime, still the reserves are quite large considering the non-cement grade limestone possible to be mined.

Mining of limestone and dolomite is already well-established and production is adequate for the size of today’s market. The shortages which often occur are due to inefficiencies in the processing operations. Mining involves removing overlying soils to expose the rock. Sledge hammers are used to reduce the size of the large pieces. When the rock face is too hard or too large to remove easily, a fire is lit on the outcrop until it is hot to the touch. The hot rock is then rapidly drenched with cold water, and the rapid cooling causes the rock to crack, and the resultant pieces are broken and removed. The stockpiling of rock in 2.0 to 2.5 tonne mounds is done on a piece-rate basis.

Limestone is then crushed using small hammers to sizes ranging from 50 to 150 mm, also on a piece-rate basis. Most small miners are aware that, for lime production, only limestone (i.e. calcitic limestone) is suitable. However, quality control at the quarrying stage is a major problem. Miners must ensure that only the high-calcium limestone is quarried.

4.2. China clay

Kaolinitic clays are the natural raw materials utilized in the production of ceramic ware and refractories. Known resources in Malawi include:

- Linthipe in Dedza, 40km from Lilongwe on the MI Road
- Nkhande, 3 to 5 km from Ntcheu Barna on the Road to Lilongwe
- The headwaters of the Rivi-Rivi River in Ntcheu District
- Senzani Village, towards Ntonda in Ntcheu District
- And the banks of the Chiula River, about 3 km east of Kaseye Mission, Chitipa District.

Only the Linthipe deposit has been extensively investigated as it is the largest and most strategically located in terms of access, availability of water supplies and geological continuity over wide areas. The Linthipe clays were formed by the weathering of feldspar-rich (anorthite) rocks. They are especially rich in silicon (Si) and aluminium (Al), hence have high Aluminium to Silicon ratios which contribute to the refractory and ceramic characteristics of the clays.
There are over 14 million tonnes of kaolinitic clays at Linthipe, which have been delineated as shown in Figure 4 and Table 4. They occur near the surface as residual clays, thinly covered by grey "makande" top soil. The Linthipe clays are generally overlain by top-soils averaging 0.2 metres thick. The ratio of waste (top soil) to ceramic clay averages about 1:6.

**Table 4: Reserves of kaolinitic clays at Linthipe**

<table>
<thead>
<tr>
<th>Block</th>
<th>Area (km²)</th>
<th>Overburden thickness (m)</th>
<th>Clay thickness (m)</th>
<th>Reserves (Mio Tonnes)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>3.5</td>
<td>0.2</td>
<td>0.7</td>
<td>4.6</td>
</tr>
<tr>
<td>3</td>
<td>1.7</td>
<td>0.2</td>
<td>0.8</td>
<td>2.6</td>
</tr>
<tr>
<td>4</td>
<td>1.0</td>
<td>0.2</td>
<td>1.3</td>
<td>2.4</td>
</tr>
<tr>
<td>5</td>
<td>1.0</td>
<td>0.3</td>
<td>1.7</td>
<td>4.5</td>
</tr>
<tr>
<td>Total</td>
<td>7.6</td>
<td></td>
<td></td>
<td>14.1</td>
</tr>
</tbody>
</table>

**Figure 4: Reserves of kaolinitic clays deposits at Linthipe, Dedza**

The Linthipe clays are generally pale-grey to buff in color and are very plastic. They occur on crests and gentle upper slopes of broad interfluvies. These clay resources are characterized as being ceramic and refractory by the following parameters:

- Chemical composition
- Particle size distribution
- Plasticity as an indication of the workability of the clay, and
- Ceramic characteristics

The chemical composition of some samples from the resource blocks at Linthipe are given in Table 5.
**Table 5: Chemical analysis of Linthipe clay**

<table>
<thead>
<tr>
<th>Oxide</th>
<th>Sample 1</th>
<th>Sample 2</th>
<th>Sample 3</th>
<th>Sample 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>45.55</td>
<td>47.78</td>
<td>46.75</td>
<td>45.55</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>34.06</td>
<td>34.68</td>
<td>33.27</td>
<td>33.05</td>
</tr>
<tr>
<td>TiO₂</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>1.07</td>
<td>1.73</td>
<td>1.60</td>
<td>3.59</td>
</tr>
<tr>
<td>CaO</td>
<td>1.45</td>
<td>1.63</td>
<td>1.11</td>
<td>0.26</td>
</tr>
<tr>
<td>MgO</td>
<td>0.21</td>
<td>0.31</td>
<td>0.28</td>
<td>0.23</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.39</td>
<td>0.60</td>
<td>0.39</td>
<td>0.07</td>
</tr>
<tr>
<td>K₂O</td>
<td>0.21</td>
<td>0.23</td>
<td>0.28</td>
<td>0.14</td>
</tr>
<tr>
<td>LOI</td>
<td>10.18</td>
<td>10.09</td>
<td>11.01</td>
<td>13.64</td>
</tr>
</tbody>
</table>

**Note:**

- **Sample 1:** Buff to white plastic kaolinitic clay, Linthipe Block 1
- **Sample 2:** Grey plastic kaolinitic clay, Linthipe Block 3
- **Sample 3:** Mottled grey-to-white plastic clay, Linthipe Block 4
- **Sample 4:** Light grey, stained brown, plastic clay, Linthipe Block 5

The Linthipe clays consist principally of clay minerals of the kaolinite group. The proportion of kaolinite in the raw clays ranges from 60 to 88% of material that is finer than 63 microns (μm). This also indicates that most of the fine clay (63 μm fraction) is kaolinite (see Table 5). The SiO₂ and Al₂O₃ contents conform to compositions of commercial ball clays and fireclays in Europe and South Africa. The clays are very plastic, with PI between 24 and 41%. The wide variations in PI are the result of variable contents of non-clay minerals.

Due to the high proportion of fines, the Linthipe clays tend to have high linear firing shrinkages, ranging from 5 to 13%, when the clays are fired at 1100°C to 1250°C, respectively.

In summary, the Linthipe clays are high alumina, refractory clays. They are very plastic, with finer material accounting for more than 65% of the clay. Hence, the clays are not, on their own, suitable for pottery purposes.

The demand for Linthipe clays for the pottery and refractory business has been growing. Two Blantyre companies use the clay for production of refractory bricks in smelting recycled steel and cast iron, and as a bonding clay in foundry sands. Dedza Potteries Limited uses the clay for producing various types of pottery. There is a potentially large market for Linthipe clays in the urban areas, particularly Blantyre and Lilongwe, however, most industries are not aware of the useful properties of the clay.

The clay deposits in Linthipe are well suited to small scale mining, using the “Ndime” mining method presently being followed in Malawi. This is essentially crude strip mining, in the following stages:

- **Stage 1:** The top soil is stripped on unit plots each with 2.0m x 2.0m surface dimensions. The top soil is stockpiled at the back (opposite from the direction of advance of mining).
- **Stage 2:** The kaolinitic clay is mined and hauled to a loading area, outside the planned mining block, then clay is transported to the processing area.
- **Stage 3:** Top soil is then spread over the mined “ndimes” before re-starting Stage 1 on new section.
All the Linthipe clay deposits are close to the M1 Road and are easily reached by good secondary dirt roads. The clay deposits are generally surrounded by densely settled villages, but most of the resource areas have not been sterilised by settlements.

4.3. Gypsum

The name gypsum is derived from the Greek "gypsos", meaning "chalk" and some school board chalk is made from gypsum. A very pure, fine-grained gypsum is called alabaster. An important property that makes gypsum a very useful industrial mineral is its capacity for easy dehydration and rehydration when it has been calcined.

When gypsum is moderately heated at a temperature of 107°C for 2 hours, it changes to a calcined form, known as Plaster of Paris (POP). Mixed with water it hardens. This property makes gypsum useful for the building construction industry as a plaster. In this hardened form, it has good insulation properties (heat, sound), it is light weight, and has stability in fire. In recent years, 60-70% of gypsum (POP) has been used in the manufacture of plaster boards for partitioning walls in multi-story buildings.

Gypsum is also widely used in the production of Portland Cement in Malawi. An addition of 4% gypsum to ground cement clinker retards the setting of cement. In medicine, POP is used for making casts on sprained or fractured limbs. It is used in the production of ceramic casting moulds, ceiling boards and as a filler in paper production.

Known gypsum resources in Malawi occur as low-grade deposits in “dambos” (the swampy drainage courses accounting for over 40% of the Lilongwe and Dowa plains). Dowa and Lilongwe are the most favorable targets for gypsum exploration because of the widespread occurrence of sulphur minerals (pyrite and pyrrhotite) in the rocks. The geological model that was developed envisaged that when such rocks weathered, the sulphur they contained was released to the drainage system. Then, in an appropriate chemical environment, such as an anaerobic one, the sulphur would combine with calcium to form gypsum (CaSO₄·2H₂O). The abundant dambos, which were formed in the last 2 million years during the waning of the peneplanation processes, were selected as the most likely sites for the deposition of gypsum. So far, 50 dambos have been found to contain gypsum, but reserves have only been located in 4 dambos:

✓ Matchenche Dambo, just south of Mponela Town
✓ Katete Dambo, near Nambuma
✓ Debza Dambo
✓ Linthembwe Dambo

In these areas Gypsum occurs as discrete crystals in clays and sands, ranging in size from less than 1.0 mm to over 4 cm. The grains are generally elongated and of angular shape. They are characteristically grey and tend to contain minute, fine clay particles. The average grades of the gypsum range from 5 to 10%. These low grades make the dambo deposit unsuitable for mechanized or systematized mining techniques. The four dambos are estimated to contain some 8,000 tonnes gypsum at an average grade of 7% (Table 6):
### Table 6: Gypsum reserves of some dambos in Dowa district

<table>
<thead>
<tr>
<th>Dambo</th>
<th>Reserves (in tonnes)</th>
<th>Average grade (wt. %)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Matchenche</td>
<td>3,500</td>
<td>5</td>
</tr>
<tr>
<td>Katete</td>
<td>2,000</td>
<td>7</td>
</tr>
<tr>
<td>Dedza</td>
<td>NA</td>
<td>8</td>
</tr>
<tr>
<td>Linthembwe</td>
<td>1,000</td>
<td>10</td>
</tr>
</tbody>
</table>

In all the dambos that have been explored near Dowa, the gypsum layer is overlain by a barren horizon, 0.2 to 1.0 metres thick. The gypsum band is 0.5 to 1.5 metres thick, in heavy plastic clay. The sand content in the gypsum bearing horizon varies between 2 to 30%. The dambos are wet, often waterlogged, most of the year, except for the months of September to November when they are dry enough to work.

Mining of gypsum is based on 2 x 2 x 2 metres of excavation each day by a team of 4 men. The process starts with stripping the overburden layer, which is deposited in the direction of advance of the mining. The gypsum bearing horizon is then mined and carted to a stockpile site. The overburden material is then moved back as a top soil, and planted with grass, banana or "nsenjere". Gypsum is processed by washing. This requires an abundant source of water to wash out the clay and collect the gypsum and sand. The separation of gypsum from sand is a mineral processing problem that has not yet been resolved because the relative densities of gypsum and sand are almost the same. Consequently, one of the present criteria for determining whether a deposit is favorable for exploitation is its low content of sand.

Low grades of the gypsum deposits that have so far been found preclude the large-scale mining operations needed to meet the requirements of Portland Cement Co., amounting to some 2,000 tonnes each year. The most feasible approach would be to encourage rural homesteads to mine and process the gypsum as an off-farming season activity at production rates ranging from 20 to 200 kg of gypsum each day. The gypsum could then be sold in the same way as food crops. The advantages of this approach are:

- Low mining overhead costs for the farming community.
- Simple and low investment capital equipment: hoes, shovels, screens (2 x 2 metres) at sizes of 5 mm, 1 mm, 7 mesh, 14 mesh and 25 mesh, 2-stroke water pump and hose.
- Mining to be carried out before crop planting resumes, hence there should be little interruption of food production activities in the villages.

### 5. Sample collection details

From the above data provided in the USAID report, it is clearly seen that there are enough raw materials to produce LC³ in Malawi. All the data show very high to medium quality raw materials. However as and when feasible other qualities can also be looked at. To verify the quality of raw materials especially china clay a brief study was done in two phases.

- Phase 1: Clay collected from Dedza pottery to understand whether required quality of china clay is available in Malawi
- Phase 2: A more detailed study of the china clay including their location and assessment of the quality of other raw materials suitable for use in LC³.

Detailed report of Phase 1 is attached as Annexure 1. For Phase 2, china clay samples were collected from Linthipe with the help of Ceramic Division, Geological Survey of Malawi. The map below (Figure 5 and 6) shows the actual area from where the china clay was collected.
Figure 5: GPS plot of china clay mines and Ceramic lab in Linthipe

Figure 6: Areas of china clay occurrences and sample collection from Linthipe

6. Characterization of raw materials

All the raw materials were characterized by Differential Thermal Analysis (DTA), Thermo Gravimetric Analysis (TGA) and X-Ray Diffraction (XRD) to determine the behavior of the raw materials under heating and mineralogical compositions. Since kaolinitic clays were the principal raw materials for production of LC$^3$, thus a detailed analysis on its calcination and reactivity was also done through an Iso-thermal Calorimeter.
6.1. Limestone

From TGA and DTA analysis (Figure 7 and 8) of the limestone sample collected from Ceramic Department, Linthipe, it is seen that the thermal dissociation pattern characterizes the sample to be a dolomitic limestone. TGA shows around 44% of weight loss in between 600 to 900°C due to CO₂ evolution upon heating. DTA of sample shows a two stage dissociation pattern. The endothermic peak corresponding to around 790°C is due to dissociation of magnesium carbonate (MgCO₃) and the second endothermic peak that appears at somewhat higher temperature of 840°C is again due to dissociation of calcium carbonate (CaCO₃). Thus the combination of TGA and DTA confirms the presence of high MgCO₃ along with CaCO₃. X-ray diffraction also confirms the thermal analysis data. The highest intensity at a 2θ of 30.9 confirms the presence of MgCO₃ along with CaCO₃. Thus both the thermal and crystallographic analysis confirms the sample to be a dolomitic grade limestone.

Figure 7: TGA and DTA analysis of limestone sample collected from Ceramic Department

Figure 8: X-ray diffractogram of limestone sample
6.2. Gypsum

From the TGA curve of gypsum (Figure 9), it is observed that there is a weight loss of 19.8% in between 150°C to 300°C, which is due to removal of water. It can be inferred from weight loss that the gypsum content of the sample is around 95%. The gypsum content is further proven by the XRD pattern (Figure 10) showing a 100 intensity peak at a 2θ of 11.6.

Figure 9: TGA analysis of natural gypsum sample found in Malawi

Figure 10: X-ray diffractogram of natural gypsum sample
6.3. China clay

From the TGA graph (Figure 11) the weight loss was determined. For dehydroxilation the weight loss beyond 200°C was calculated. Weight loss between room temperature and 200°C was due to the release of free moisture. DTG also shows the mass changes corresponding to dehydroxilation. The mass loss of 8.5% between 400-750°C was due to dehydroxilation of kaolinite. Kaolinite content of the clay was calculated from the weight loss by an empirical formula developed by EPFL, Switzerland. For a corresponding weight loss of 8.5% the kaolinite content was calculated to be 61%. This is also evident from the kaolinite (K) peaks in the XRD.
analysis (Figure 12) corresponding to a 2θ of 12.3, 24.9 and 62.2. Besides kaolinite, evidence of quartz (Q) at a 2θ of 20.08, 26.5; hematite (H) at a 2θ of 35.7, muscovite (M) at 34.9 and paragonite (P) at 27.8 was also seen. Thus although the clay contained an appreciable quantity of kaolinite, considerable quantity of other impurities are also present.

7. **Calcination process**

Calcination of four clays were carried out in programmable muffle furnace at 800°C. All the samples were used in the powder form kept in crucibles. Samples were heated from room temperature to 800°C at a rate of 13°C (Figure 13). All the samples were soaked at 800°C for 30 minutes followed by normal cooling. Figure 6 shows the temperature profile of calcination. Efficiency of calcination was assessed by TGA and XRD.

![Figure 13: Temperature profile of static calcination of the china clay](image)

8. **Assessment of calcination**

8.1. **Comparative analysis of TGA of calcined and raw clay**

Comparative analysis of TGA of raw and calcined clay are given in Figure 14. Based on the weight loss data, it was concluded that the clay was calcined at 800°C properly, as there were very negligible (within experimental error) weight loss in TGA curve of the calcined clay. Thus for pilot scale production, during calcination under static or rotary conditions the material temperature should be kept at 800°C. For production scale calcination, experiments needs to be done to find the difference in temperature due to process loss and compensated accordingly.
8.2. Comparative analysis of XRD of calcined and raw clay

From X-ray diffractogram (Figure 15), in the clay after calcination, no kaolinite peaks were observed. A broad hump, representing amorphous clay or metakaolin is confirmed. Presence of crystalline peaks after calcination is due to impurities, indicating their stability even at 800°C. It is inferred that presence of these phases in calcined clays might affect reactivity. The calcination temperature was also appropriate since no mullite peaks were observed.
9. Reactivity assessment

Lime reactivity test of the calcined Linthipe clay was performed in an Isothermal calorimeter (Figure 16). It has been confirmed from previous study that lime reactivity of a clay depends mainly on kaolinite content in the clay. Kaolinite gets transformed into metakaolin on calcination and result in pozzolanicity. Combination of thermogravimetric and crystallographic analysis of clays confirm a appreciable content of kaolinite in the Linthipe clay, resulting in high reactivity in comparison to a standard Bhuj clay tested in India (Figure 16). The kaolinite content of the Bhuj clay was 56%. Thus it can be seen that the Linthipe clay is more reactive compared to the Bhuj clay due to higher kaolinite content.

![Figure 16: Comparative analysis of reactivity of raw and calcined clay](image)

10. Conclusions and recommendations

The Linthipe clay is a kaolinitic clay with other impurities. It also has a high reactivity based on the amount of kaolinite content. A high reactivity also proves that the impurity content does not have an adverse effect on the Pozzolanic properties of the clay. Based on the reactivity tests it can be definitely said that it is a good kaolinitic clay for use in LC3 application. It can be inferred from experimental knowledge on working with Bhuj clay that a 43 Grade LC3 cement can be achieved with the Linthipe clay calcined at a low temperature of 800°C. However it must be mentioned with caution that the above is based on experience on working with other clays and for any definite conclusions more tests needs to be done with an actual LC3 combination.

This type of limestone calcined clay cement having similar properties to normal portland cement will be a boon to the cement industry in Malawi in lowering down the clinker factor and reduce energy consumption and environmental emissions.

It is recommended to get around 200kg of the Linthipe clay for more extended tests and also prepare a batch of LC3 for its test as a cement. However more clays should also be explored for tests. Care should be taken for right selection of clays. The basic properties of the clays found to be suitable for use in LC3 are listed in Table 7 below.
Table 7: Suggested properties of china clay for use in LC3 production

<table>
<thead>
<tr>
<th>Sl.</th>
<th>Property</th>
<th>Value</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Loss on ignition from 300˚C to 750˚C</td>
<td>&gt;7%</td>
<td>Single peak in TGA in this temperature range, required for 50% kaolinite content</td>
</tr>
<tr>
<td>2</td>
<td>SiO2</td>
<td>&lt;65%</td>
<td>To avoid too high Quartz content</td>
</tr>
<tr>
<td>3</td>
<td>Al2O3</td>
<td>&gt;24%</td>
<td>Required for approximately 50% kaolinite content</td>
</tr>
<tr>
<td>4</td>
<td>Al2O3 to SiO2 ratio</td>
<td>&gt;0.38</td>
<td>Required for main clay phase being kaolinite</td>
</tr>
<tr>
<td>5</td>
<td>Colour</td>
<td>Not important</td>
<td></td>
</tr>
</tbody>
</table>

11. References