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Geochemical Assessment of Antalo Limestone for Cement Manufacturing.

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***ABSTRACT**: Limestones are one of the most important of all the sedimentary rocks. Limestones are composed mostly of the mineral calcite (CaCO3). They may also contain some other carbonates minerals and several non-carbonate impurities. Limestones are the raw materials widely used by throughout industry, although the limestone is the first raw materials for cement making industry where chemical properties are important. Portland cement is produced by calcining finely ground raw meal consisting of a mixture of about 75% limestone and 25% of clay, at about 1450°C in a rotary kiln to form a calcium silicate clinker which is then ground and mixed with a small amount of gypsum which acts as a setting retardant. The compositional chemistry of cement depends largely on geochemistry of its raw materials, i.e., limestone. Approximately 75% of the cement's raw material consists of lime (CaO)-bearing material.*

Cement is the prime ingredient used in the construction industry. Cement consumption has a direct correlation to economic growth and improvement in the living standards of society. Energy and capital intensity nature of the industry necessitate large investments that require a long-term perspective on financing and returns. Besides, production and consumption of cement are mainly subject to economic and construction cycles, resulting in volatility of operating costs and revenues. Addition to that Portland cement clinker production is one of the major sources of CO2 and other greenhouse gases within the contribution of 5 % of the annual global atmospheric CO2 emission. The increasing demand for cement has also provided the desired boost to the cement industry leading to a quite visible growth of additional production capacity and need to find out other cement replacing potential materials. As a result, the study amid to assess on cement production practice and potential cement replacing materials in Ethiopia to reduce the high cost of cement in order to provide sustainable and cost-efficient structure for the public and private sectors. This paper documents the geochemical assessment of limestone for cement manufacture.

Key words: *Limestone, Calcite, Carbonates, Calcium silicate, Gypsum, Geochemistry*

I.INTRODUCTION

1.1 Background of the study area

The geology of Ethiopia consists of Precambrian basement complexes, late Paleozoic to the Early Tertiary sedimentary rocks and Cenozoic volcanic rocks in an ascending order. The Paleozoic and Mesozoic sedimentary successions are present in three distinct provinces including; the Mekelle outlier in the north (Beyth, 1972; Bosellini et al., 1997); the Blue Nile Basin in the central part (Jepsen, 1964; Beauchamp, 1977; Russo et al., 1994); and the eastern margin of the Ethiopian Rift Harrarghe, and the adjacent Ogaden Basin (Kazmin, 1973; Merle et al., 1979; Bosellini et al., 2001; Asfawossen As rat, 2015). In the Mesozoic times, continental Jurassic sedimentation in Ethiopia was interrupted by the transgression of the Ocean. This sea level rise is documented in the Antalo Formation (Russo et al., 1994; Balemwal Atnafu, 2003; Gila Michael Kidane Mariam et al., 2009). During Regression started in the Late Jurassic and resulted in the deposition of the predominantly continental sequences of the Mug her Mudstone and the Debre Lebanese formations of the Abiy River Basin (Getaneh Assefa, 1991). The topmost oolitic limestone of the Antalo Formation (Lagajima Limestone) includes invertebrates of Kimmeridgian age (Russo et al. 1994).

1.2 Location and accessibility of the study area

The study area is found in the Blue Nile Basin, particularly located in the Blue Nile Gorge, hitch is exposed along the Dejen-Gohatsion area near to Abiy River in main road and is about 270 km from Bahir Dar and 290 km from Addis Ababa. The main target of this work focused in the antalo limestone formation which overlain on the Gohatsion formation. The study area enclosed between 402000m-414000m easting and 1120000m-1110000m northing.

1.3Population and Settlement

The study area is dominantly populated by Amara People. The rural people made live on agricultural and animal farming. The land farm is highly exposed for erosion and land slide thus decreasing the fertility of soil for crop production. They grow maize, sorghum, ―Teff‖, and wheat.

1.4 Objectives of the Study

1.4.1 General objective

The major aim of the study is to analyze, assessing and discuss the suitability of antalo limestone for cement manufacturing.

1.4.2 Specific objectives

the study was geared to attain the following specific objectives.

- ✓ *To investigate quality of antalo limestone for cement manufacturing*
- ✓ *To determine the modal composition of antalo limestone*
- ✓ *Produce Portland limestone cement as an alternative cement type having comparable characteristics to that of OPC and PPC*
- ✓ *To classify the different types of limestone;*

1.5 Statement of the problem

Some researchers have been done on the sedimentation, depositional environment and digenesis of antalo limestone in the Blue Nile Basin (Wolela, 2008, 2010). Getaneh (2002) However, the source rock composition, source area weathering and tectonic setting of the antalo limestone in the Blue Nile Basin are yet not well studied. Hence this work investigates parent rock assemblages of the antalo limestone of the Blue Nile Basin of their depositional tectonic setting; provenance and source area weathering, quality of antalo limestone for cement manufacturing, geochemical approach and compare with the antalo limestone in Mekelle Basin.

1.6 Scope of the study

The scope of the study focuses on chemical and physical characterization of the limestone resource that is assumed to be used as one of main inputs in cement manufacturing or qualitative and quantitative evaluation of the minable parts of the limestone deposit,

II.LITRATURE REVIEW

2.1 Regional Geologic Setting

To understand the regional geological setting of Blue Nile Basin, there are different stage and event of sedimentary basin evolution as follows:

2.1.1 Peneplain stage: *This stage corresponds to the Pan-African metamorphic peneplain, which covered with a veneer of pediment sediments (Russo et al., 1994). It represents the situation before any tensional effect and before the Gondwanaland break-up.*

2.1.2 Intercontinental rift stage: *It is equivalent to the early stage of Karoo rifting and as a consequence of Gondwana break- up in late Paleozoic-Jurassic times, continental rifts were developed especially along the border of the mega continent as a result of NW-SE tensional stresses with a correspondent thinning of the continental crust produced graben and half-*

grabens and a number of rift basins stretching from South Africa to eastern Ethiopia (Baseline et al., 1989). The early stage of Karoo rifting initiated the formation of N-S, NW-SW and NE-SW oriented rift basins such as Ogaden and Blue Nile Basins (Wolela, 1997; Hun gnaw, 1998 and reference therein). The Blue Nile Basin has NW-SE trending in the cause of NE-SW tensional or extensional forces. The early sedimentation of the Karoo rift in eastern Ethiopia commenced with the deposition of the alluvial fan deposits of the Caleb sandstone formation, the lacustrine Bokeh shale formation, and the fluviatile Gambaro sandstone formation (Wolela, 2004).

2.1.3 post-rifting stage: *This stage corresponds to the deposition of the Adigrat sandstone formation (Russo et al., 1994). This stage consists the following events: -*

2.1.3.1. Early Flooding: *Corresponds to the early transgression by the sea and flooding of the craton, due to the rifting and thermal subsidence of the East African continental margin. The thermal subsidence and eustatic sea level changes caused the southern arm of Tethys to spread westward over the Arabo-Ethiopian shield (Bosselini et al., 1989). The beginning of the marine transgression is documented by the deposition of the Gohatsion formation in the Blue Nile basin (Assefa, 1987).*

2.1.3.2 Drowning of the craton: *This stage related to a major transgression, dated as Callovian-Early Oxfordian over the entire East Africa, is documented by the Antalo Limestone (Russo et al., 1994). This is probably related to the drifting phase and major sea level high stands.*

2.1.3.3 At the end of the Jurassic time*: the sea began to withdraw from the Horn of Africa probably as result of the intraplate effect of the separation of South America and Africa (Manspeizer., 1988; Bosellini et al., 1989). The withdrawal of the sea initiated the deposition of the transitional facies, Mug her mudstone and Debre-Libanose sandstone units. From late Cretaceous to Lower Miocene there was successive uplifting and subsidence events including the volcanic eruption which related to the Afar mantle plume. At the end of cretaceous rifting began in the Gulf of Aden area and the then the formation of red sea and the Main Ethiopian Rift (Bunter et al., 1998, Korma et al., 2004 and the reference therein). According to (Wolfenden et al.,2004) the earliest volcanism may have initiated between 32 and 33 Ma with the greatest eruption rates and volumes occurring ~31 to 28 Ma, and the total duration of the flood volcanism was at least 4 Ma.*

The Blue Nile Basin is one of the sedimentary basins, which covers an area of about 120,000km2 (Wolela, 2008). Today the large parts of Mesozoic sediments are exposed on the Eastern Ogaden, central dissected plateau areas in the Blue Nile River Basin and in Northern Tigray around Mekelle. At the central part of the Blue Nile Basin (Gohatsion-Degen area), the sedimentary succession reaches a maximum thickness of 3000m (Assefa, 1991; Russo et al,1994; Wolela, 1997).

2.2 The Lithological Description of The Blue Nile Basin

Generally, the Blue Nile Basin of central Ethiopia consists of Neoproterozoic basement rocks, Paleozoic and Mesozoic sedimentary succession and the Quaternary volcanic rocks exposed in the Blue Nile canyon. According to Dawit (2010) the complete Paleozoic and Mesozoic sedimentary succession in the Blue Nile Basin are

- ✓ *Per-Adigrat (I and II),*
- ✓ *Adigrat Sandstone Formation,*
- ✓ *Gohatsion Formation,*
- ✓ *Antalo Limestone formation,*
- ✓ *Mug her Mudstone and*
- ✓ *Debre Libanos Sandstone bottom to top respectively.*

2.2.1 Basement Rocks

The basement rocks of the Blue Nile Basin consist of the metamorphosed quartzofeldspathic schist and gneisses, migmatites and plutonic rocks (Gani et al., 2009). The age of these rocks considered to Neoproterozoic, ranging from 850 to 550 Ma as documented from U-Pb and Rb-Sr geochronologic studies further south of the study area (Ayalew et al. 1990).2.2.2. Adigrat Sandstone

This formation rests unconformable on the Pre-Adigrat (I and II) Paleozoic sediments and consists approximately 300 m thick in Degen to Gohastion area of Blue Nile basin (Dawit, 2010). The Adigrat Sandstone is widely distributed in the Blue Nile, Ogaden, and Mekelle basins. It covers an extensive area in the Blue Nile Basin and forms vertical cliff exposures in Degen, Gohatsion, Amur and Jarty areas etc. In the Blue Nile Basin, the Adigrat Sandstone Formation reaches thickness ranges from 120m Arjo (Assefa, 1987) to of 800m in Amuru-Jarty (Tamrat and Tibebe, 1997).

2.2.3 Glauconitic Sandy Mudstone

According to (Gani et al., 2009) this unit overlain on the Adigrat Sandstone formation, which have approximately 30 m thick of greenish color. This unit reported for the first time by (Gani and Abdulsalam, 2006) found as sandwiched between Adigrat Sandstone and Gohatsion formation. This characterized by hummocky cross-stratification and wave ripples in upper parts of this unit, indicating storms and waves in marine environment.

2.2.4 Gohatsion Formation

The Gohatsion Formation was consisting of a cyclic repetition of facies successions that are composed, from bottom to top, of alternating dolostones, marlstones and shales, bioturbated mudstones with thin siltstone intercalations, fine-grained coquinoidal cross-laminated sandstones and thick beds of gypsum (Assefa, 1981 and Russo et al, 1994). According to (Russo et al., 1994) the presence of scattered small bivalves and gastropods (Corbiculae, Lucinids and other) indicates peri tidal environments with associated lagoon and pond waters bodies. This formation corresponds to the initial flooding of the craton, which is largely related to rifting and subsidence of the African continental margin (Russo et al., 1994).

2.2.5 Antalo Limestone

The Antalo limestone conformably overlies the Gohatsion Formation, which contain about 420m thick carbonate succession (Russo et al., 1994 and Atnafu, 2003), and can be subdivided into three parts.

The lower part has 180 m thick and composed of burrowed mudstones that grade upwards into oolitic and coquinoidal limestones with or without intercalated marl beds, and massive limestones with scattered patches of corals, and stromatoporoids, which was indicated a shallow water environment.

The middle part also contains 200 m thick consists of highly fossiliferous interbedding of marly limestones and marls. The presence of ammonite fauna (e.g., Lithoscopes sp. and Subplanites spathe), with brachiopods (e.g., Terebratula pelagic a and Nanogyra) and other in faunal siphone feeders (Anisocardia, Venilicardia and Somalirhynchia somalica and Zeillleria latifrons) suggests a shelf to open marine environment (Russo et al. 1994, Atnafu, 2003).

The upper part of Antalo Limestone contains about 50m thick comprises planar laminated oolitic and refal limestone, which was interpreted to indicate the return of shallow water conditions (Dawit, 2010).

2.2.6 Mug her Mudstone

The name of this formation derived from Mug her River, it consists mainly mudstone and sandstone with gypsum and dolomite. The unit exposed in mugger valley, zaga wedon, wesena adabai and Jemma River section (Russo et al., 1994 and Assefa, 1991), but it pinches out towards the abay river gorge (Wolela, 2004).

2.2.7 Debre Liberoes Sandstone

This formation conformably overlies the mug her mudstone formation and is in turn overlain by volcanic rocks. The unit dominantly exposed in the Zega Wodem river and its tributaries, which have large and small-scale planar tabular and asymmetrical through cross-beds, convolute beds, flat beds, scoured and channel surface and massive beds. Some fining upward sequence occur from medium to fine grained sandstone up to laminated clay stones and this unit interpreted as a deposit of sandy-braided rivers on a broad alluvial plan (Assefa, 1991).

2.2.8 The Volcanic Rocks

The volcanic rocks are post Oligocene age (Hoffman et al., 1997) and about 5500 meters of maximum thickness (Assefa, 1991). This unit unconformably overlies the Antalo Limestone at Degen-Gohastion section and it consist basalt, trachyte, and rhyolite with beds of truffe, paleosols and lacustrine sediment (Assefa, 1991). Generally, the Blue Nile Basin sedimentary succession of chrono and lithostratigraphic unites are shown (Figure 2.1).

2.3 Lithological Description of Antalo Limestone

The name Antalo Limestone was first entitled by Blandford (1869, 1870) after the town Antalo in northern Ethiopia of the Tigray region. Later on, the unit was well-described by Levitte (1970), Beyth (1972a, b), Merle et al. (1979), Bosellini et al. (1997) and Matire et al. (2000). The limestone unit is represented by three formations.

✓ *The lower cliff-forming unit is composed of white, pale yellow limestone with shale intercalation and contains fossils of bivalves & brachiopods.*

✓ *The middle cliff forming limestone is intercalated with marl & mud rock. It forms gentle slopes and is the thickest part attaining up to 350m thickness. The limestone is micritic, rich with fossils mainly of brachiopods and bivalves. It is characterized by horizontally bedded sedimentary structures.*

✓ *The upper cliff forming limestone unit has thin layers of marl and shale intercalations in the lower part but is pure limestone at the upper part. This limestone is more fossiliferous than the lower cliff forming limestone and is crystalline, medium to coarse-grained with interbeds of calcite nodules.*

According to Getaneh Assefa (1991) the carbonate unit consists of approximately about 600m thick of fossiliferous dominated carbonate interbedded with marl, shale and mudstone. The top most part of this unit is oolitic, massive and cliff forming limestone.

It is found in the SW-flowing segment of the Blue Nile sandwiched between the Early– Middle Jurassic Shaly and gypsum unit, and either the Late Jurassic–Early Cretaceous Upper Sandstone unit (Mug her mudstone or the Debre Libanos sandstone) or in some vicinity to Early–Late Oligocene volcanic rocks.

2.3.1 Lower Limestone: *An Early Jurassic age was therefore assigned to this unit based on its stratigraphic position. The upper part of this unit is characterized by hummocky cross-stratification and wave ripples, indicating storms and waves in a marine environment. A trough cross-stratified shoreface sandstone interval has also been identified within the upper part of this unit. Presently, the glauconitic unit is preserved as mound-shaped erosional remnants which appear festoon-shaped in map view (Gani and Abdulsalam 2006). This unit is interpreted to be deposited in an offshore to shelfful marine environment.*

2.3.2 Lower Limestone and gypsum unit: *This unit, 450 m thick, also known as the Gohatsion Formation, is of Early–Middle Jurassic age, as determined from micro– and mega–fossil studies by Assefa (1981). It is exposed along the SW-flowing segment of the Blue Nile where it is underlain by the glauconitic sandy mudstone unit or the Triassic–Early Jurassic Sandstone and overlain by a Middle–Late Jurassic Upper Limestone unit. The unit consists of a lower thinly bedded (average 20 cm) limestone interval and an upper interval of alternating*

The gypsum beds are characterized by mottled texture, and are inter-bedded with glauconitic mudstone beds and rare thin sandstone beds. Deposition of the Lower Limestone indicates deepening of the basin. However, the alternation of gypsum and limestone in the upper part of the unit indicates repetitive drying and flooding of an evaporitic basin. The Lower Limestone is cross-cut by NW-trending normal faults, NE- and NW-trending dilutional fractures and less-frequent NE-trending normal faults.

2.3.3 Upper Limestone: *This 400 m thick unit comprises thinly bedded (average 10 cm) to massive limestone. It is also known as the Antalo Limestone, which is of Middle–Late Jurassic* *age. It is found in the SW-flowing segment of the Blue Nile sandwiched between the Early–Middle Jurassic Lower Limestone unit, and either the Late Jurassic–Early Cretaceous Upper Sandstone unit or Early–Late Oligocene volcanic rocks. Although the base of this unit is not exposed in the S-flowing segment of the Blue Nile, it is overlain by the Late Jurassic–Early Cretaceous Upper Sandstone unit.*

The deposition of the Upper Limestone indicates a second major marine transgression in the Blue Nile Basin. The Upper Limestone is affected by NW- and NE-trending normal faults, the throws of which generally range between a few cm and 60 m, but with one fault having a 400 m throw. Fault zones range from a few cm to 50 m wide. Fractures within this unit are dilutional and dominantly N-trending with subordinate ENE- and NW-trending sets.

2.4 Description of Limestone

Limestone is a sedimentary rock composed primarily of calcium carbonate (CaCO3) in the form of the mineral calcite. It most commonly forms in clear, warm, shallow marine waters. It is usually an organic sedimentary rock that forms from the accumulation of shell, coral, algal, and fecal debris. It can also be a chemical sedimentary rock formed by the precipitation of calcium carbonate from lake or ocean water.

2.4.1 Environment of Formation

2.4.1.1 Limestone-Forming Environment: Marine

Most limestones form in shallow, calm, warm marine waters. That type of environment is where organisms capable of forming calcium carbonate shells and skeletons can easily extract the needed ingredients from ocean water. When these animals die, their shell and skeletal debris accumulate as a sediment that might be lithified into limestone. Their waste products can also contribute to the sediment mass. Limestones formed from this type of sediment are biological sedimentary rocks. Their biological origin is often revealed in the rock by the presence of fossils. Some limestones can form by direct precipitation of calcium carbonate from marine or fresh water. Limestones formed this way are chemical sedimentary rocks. They are thought to be less abundant than biological limestones.

Today Earth has many limestone-forming environments. Most of them are found in shallow water areas between 30 degrees north latitude and 30 degrees south latitude.

2.4.1.2 Limestone-Forming Environment: Evaporative

Limestone can also form through evaporation. Stalactites, stalagmites, and other cave formations (often called "speleothems") are examples of limestone that formed through evaporation. In a cave, droplets of water seeping down from above enter the cave through fractures or other pore spaces in the cave ceiling. There they might evaporate before falling to the cave floor.

When the water evaporates, any calcium carbonate that was dissolved in the water will be deposited on the cave ceiling. Over time, this evaporative process can result in an accumulation of icicle-shaped calcium carbonate on the cave ceiling. These deposits are known as stalactites. If the droplet falls to the floor and evaporates there, a stalagmite could grow upwards from the cave floor. The limestone that makes up these cave formations is known as "travertine" and is a chemical sedimentary rock. A rock known as "tufa" is a limestone formed by evaporation at a hot spring, lake shore, or other area.

2.4.2 Types of Limestone

Limestone is by definition a rock that contains at least 50% calcium carbonate in the form of calcite by weight. All limestones contain at least a few percent other materials. These can be small particles of quartz, feldspar, clay minerals, pyrite, siderite, and other minerals. It can also contain large nodules of chert, pyrite, or siderite. There are many different names used for limestone. These names are based upon how the rock formed, its appearance or its composition, and other factors. Here are some of the more commonly used varieties.

2.4.2.1 Chalk: *A soft limestone with a very fine texture that is usually white or light gray in color. It is formed mainly from the calcareous shell remains of microscopic marine organisms such as foraminifers, or the calcareous remains from numerous types of marine algae.*

2.4.2.2 Coquina*: A poorly-cemented limestone that is composed mainly of broken shell debris. It often forms on beaches where wave action segregates shell fragments of similar size.*

2.4.4.3 Fossiliferous Limestone: A limestone that contains obvious and abundant fossils. These are normally shell and skeletal fossils of the organisms that produced the limestone.

2.4.2.4 Lithographic Limestone: *A dense limestone with a very fine and very uniform grain size that occurs in thin beds which separate easily to form a very smooth surface. In the late 1700s, a printing process (lithography) was developed to reproduce images by drawing them on the stone with an oil-based ink and then using that stone to press multiple copies of the image.*

2.4.2.5 Oolitic Limestone: *A limestone composed mainly of calcium carbonate "oolites," small spheres formed by the concentric precipitation of calcium carbonate on a sand grain or shell fragment.*

2.4.2.6 Travertine: *A limestone that forms by evaporative precipitation, often in a cave, to produce formations such as stalactites, stalagmites, and flowstone.*

2.4.2.7 Tufa*: A limestone produced by precipitation of calcium-laden waters at a hot spring, lake shore, or other location.*

2.5 Uses of Limestone

Limestone is a rock with an enormous diversity of uses. It could be the one rock that is used in more ways than any other. Most limestone is made into crushed stone and used as a construction material. It is used as a crushed stone for road base and railroad ballast. It is used as an aggregate in concrete. It is fired in a kiln with crushed shale to make cement. Some varieties of limestone perform well in these uses because they are strong, dense rocks with few pore spaces.

These properties enable them to stand up well to abrasion and freeze-thaw. Although limestone does not perform as well in these uses as some of the harder silicate rocks, it is much easier to mine and does not exert the same level of wear on mining equipment, crushers, screens, and the beds of the vehicles that transport it. Some additional but also important uses of limestone include:

2.5.1 Dimension Stone: *Limestone is often cut into blocks and slabs of specific dimensions for use in construction and in architecture. It is used for facing stone, floor tiles, stair treads, window sills, and many other purposes.*

2.5.2 Roofing Granules: *Crushed to a fine particle size, crushed limestone is used as a weather and heat-resistant coating on asphalt-impregnated shingles and roofing. It is also used as a top coat on built-up roofs.*

2.5.3 Flux Stone: *Crushed limestone is used in smelting and other metal refining processes. In the heat of smelting, limestone combines with impurities and can be removed from the process as a slag.*

2.5.4 Portland Cement: *Limestone is heated in a kiln with shale, sand, and other materials and ground to a powder that will harden after being mixed with water.*

2.5.5 AgLime: *Calcium carbonate is one of the most cost-effective acid-neutralizing agents. When crushed to sand-size or smaller particles, limestone becomes an effective material for treating acidic soils. It is widely used on farms throughout the world.*

2.5.6 Lime*: If calcium carbonate (CaC03) is heated to high temperature in a kiln, the products will be a release of carbon dioxide gas (CO2) and calcium oxide (Cao). The calcium oxide is a powerful acid-neutralization agent. It is widely used as a soil treatment agent (faster acting than aglime) in agriculture and as an acid-neutralization agent by the chemical industry.*

2.5.7 Animal Feed Filler*: Chickens need calcium carbonate to produce strong egg shells, so calcium carbonate is often offered to them as a dietary supplement in the form of "chicken grits." It is also added to the feed of some dairy cattle who must replace large amounts of calcium lost when the animal is milked.*

2.5.8 Mine Safety Dust: *Also known as "rock dust." Pulverized limestone is a white powder that can be sprayed onto exposed coal surfaces in an underground mine. This coating improves illumination and reduces the amount of coal dust that activity stirs up and releases into the air. This improves the air for breathing, and it also reduces the explosion hazard produced by suspended particles of flammable coal dust in the air.*

Limestone has many other uses. Powdered limestone is used as a filler in paper, paint, rubber, and plastics. Crushed limestone is used as a filter stone in on-site sewage disposal systems. Powdered limestone is also used as a sorbent (a substance that absorbs pollutants) at many coal-burning facilities.

Limestone is not found everywhere. It only occurs in areas underlain by sedimentary rocks. Limestone is needed in other areas and is so important that buyers will pay five times the value of the stone in delivery charges so that limestone can be used in their project or process.

2.6 Description of Cement

2.6.1Typesand features of cement

Cement is a fine product used to bind a substance which sets and hardens others materials together. Cement means to chemically combine where water is the trigger that gets the chemical reaction going (i.e., hydration). Cement is one of the backbones for the development of an economy serving as a basic raw material for the development of infrastructures, construction of small, medium and large-scale industries, construction of residential and non-residential buildings and others. Numerous types of cement are currently produced worldwide to meet a range of requirements and needs. The types of cement are differentiated based on the composition of ingredients (raw materials) used and other factors. The following major types of cement are being produced across the world. Ordinary Portland Cement; Portland Pozzolana Cement; Moderate Heat Portland Cement; Rapid Hardening Cement; Low Heat Portland Cement; Sulphate Resisting Cement; and White Cement.

In Ethiopia only Ordinary Portland Cement (OPC) and Portland Pozzolana Cement are being produced. Portland cement is the generic term for the type of cement used in virtually all concrete work. (Ministry of trade and industry 2011/ 2012) Based on interview with cement producers the inputs required in cement production include limestone, clay, pumice and gypsum. These four inputs contain four important and essential elements called calcium, silicon, aluminum and iron. Generally the production process follows the following steps: Individual raw materials are crushed with the required size typically 50 mm, Then the raw materials are roughly blended in a pre- homogenization pile, The raw materials are next ground together in a raw mill on which controlled proportions of each material are delivered onto the belt by weigh feeders which produce the raw mix, The raw mix undergoes clinkering using a high temperature, 1450 – 1500 degree centigrade, in a kiln that drives off carbon dioxide and fuses the mixture and The last step involves adding gypsum to the cooled clinker and grinding the resulting mixture to produce cement and it conveyed by belt to a silo for storage.

Table 1: Production capacity of cement plants (2011/12)

NO	***Plant Name***	***Attainable Capacity in ton***	***Produced amount in ton***
1	*Mug her Cement*	*900,000*	*540,000*
2	*Messe Bo Cement*	*900,000*	*540,000*
3	*National Cement*	*150,000*	*90,000*
4	*Jemma Cement*	*15,000*	*9,000*
5	*Abyssinia Cement*	*90,000*	*54,000*
6	*Red fox cement*	*100,000*	*60,000*
7	*Haunching cement*	*435,000*	*261,000*
8	*Zhongshan cement*	*250,000*	*150,000*
9	*MidrocDejen*	*90,000*	*54,000*

10	Debresina cement	90,000	54,000
11	Hwaiu	150,000	90,000
Total		**3,170,000**	**1,902,000**

Source: Ministry of Trade and Industry (2011/12)

2.6.2 Role of Limestone for Cement Manufacturing

Portland limestone cement is thus, one of the alternative cement types that can be produced in our cement industries. The raw materials for production of such cement type is sufficiently available, requires lesser energy for production, and has less CO2 and NO2 emissions. It is also economical for countries like Ethiopia where OPC and PPC cement types are the only types of cement used for all construction purposes. This study is thus attempting to make use of these limestone fines in the production of Portland limestone cement. An experimental investigation is thus carried out to examine the impacts of adding these fines on the physical and chemical properties of the cement paste such as consistency, setting time, and compressive strength.

2.6.3 Limestone in Cement Production

Limestone is made up of varying proportions of chemicals such as calcium carbonate (CaCO3), Magnesium carbonate (MgCO 3), Silica (SiO2), Alumina (Al 2O3), Iron oxide (Fe2O3), Sulphate (SO3), and Phosphors (P2O5) with calcium and magnesium carbonate being the two major components. Limestone is the most common form of calcium carbonate which is used extensively for the manufacture of cement. Cements in different types are made mainly by calcining a mixture of about 75% limestone and 25% clay to form a calcium silicate clinker which is then ground and mixed with a small amount of gypsum. According to European standard (EN 197) limestone used as a main constituent in cement production shall meet the following requirements

i) The Cacao 3 content shall be ≥ 75% by mass

ii) Clay content methyene blue absorption should be 1.2 g/100 g, and

iii) Total organic material content (TOC) shall be ≤ 0.2% by mass.

Limestone is also one of the well-known ingredients used for the production of Masonry cement by intergrading with Portland cement together with plasticizing material.

2.6.4 The Effects of Limestone Fillers on Cement Properties

2.6.4.1 Fineness: *Limestone fillers have many effects on the cement properties due to its fineness. Inclusion of this fine material will significantly accelerate the hydration of alite and aluminates of the cement, because the particles act as nucleation sites for the formation of the hydration products. Another effect of finely divided additions is their action as fillers between the cement grains producing a denser paste and densifying the interfacial zone between the aggregate and cement paste.*

2.6.4.2 Consistency: *The effect of limestone powder on the water requirement of OPC and blended cement has been studied extensively and a majority of findings are in favor of a better workability of mortar and lowering the water requirement for neat paste containing limestone. The improvement in the workability of paste and mortar is due to suitable texture fineness and particle size distribution of cement containing limestone.*

2.6.4.3 Soundness*: The recent findings show that the addition of calcareous material (limestone) up to the range of 5-7% in cement mortar have smaller influence on shrinkage as compared to siliceous additives. It is also confirmed by different investigators that there is no remarkable effect on the soundness of OPC paste with up to 10% replacement by limestone additives.*

2.6.4.4 Hydration: *Many research papers on influence of limestone powder on hydration of Portland cement have reported that the C3S hydration rate is accelerated when the amount and fineness of CaCO3 is increased. This is due to the fact that they generate a large number of nucleation sites for precipitation of the hydration products.*

2.6.4.5 Compressive strength: *It has been found that addition of limestone powder into cement paste and mortar increases the strength at early ages without changing the workability of mortar. It has also been investigated that blending of Portland cement with 10 - 40% finely ground limestone improves the early strength. Findings of research works on the strength reveals that, irrespective of grinding methods (intergrading or separate grinding of limestone and other materials) up to 5%, the strength of limestone cement at 3 and 7 days were slightly higher than the pure ordinary Portland cement since increase in strength is directly related to the increase in rate of hydration of cement obtained due to the addition of limestone fillers. However, findings also reveal that, as the percent of substitution of limestone in OPC increases, the compressive strength development of resultant cement decreases.*

2.6.4.6 Grinding: *Studies have shown that intergrading clinker with different proportion of limestone resulted in improved grinding behavior of clinker resulting in saving of grinding time and decrease in fuel and electric consumption cost. Thus, the European cement industries allowed using mineral addition to economize the production of cement under the specified standard. It has also been proved that intergrading has resulted in better particle size distribution for the same energy level than that of separately grinding of raw materials with clinker.*

2.6.4.7 Environmental impact: *It has been recognized that cement industries release different gases to the atmosphere including greenhouse gas emissions, Dioxin, NO2, SO2, and vibration during operating machinery and blasting in quarries. It is an established fact that 0.9 ton of CO2 is emitted per ton of cement. However, there are strategies for the reduction of carbon dioxide which aimed at lowering emissions per ton of cement, even though there is inherent emission of carbon dioxide during chemical breakdown of the limestone in cement kilns during production of Portland cement clinker.*

One of the strategies of decreasing CO2 emission is intergrading or blending limestone with Portland cement which offers key advantages in reduction of CO2 emissions, climate change, economic and technical benefits.

2.6.5 Properties of Materials Used in The Investigation

2.6.5.1 Clinker: *The clinker used for this investigation was Portland cement clinker produced by Degen cement factory. The suitability of this clinker was investigated from its chemical and mineralogical compositions which are the major properties responsible for the quality of cement. To this effect, the clinker chemical and mineralogical compositions were tested in Degen cement enterprise materials quality testing and assurance laboratory and the test results are shown in*

Table 2 and 3, respectively.

Table 2: Chemical compositions of Degen Portland cement clinker

Chemical composition	DEJEN clinker (%)
SiO_2	22.15
Fe_2O_3	3.43
Al_2O_3	5.76
Cao	65.05
MgO	1.05
SO_3	1.04
LOI (Loss on Ignition)	0.08
IR (Insoluble residue)	0.05
F-Cao (Free calcium)	0.67
SM (silica Modules)	2.41
AM (Aluminum Modulus)	1.68
100LSF (Lime Saturation Factor)	91.93
CI (Coating Index)	32.35
LPH (Liquid phase)	26.19
$CaCO_3$	76.39

Table 3: Mineralogical compositions of Degen cement clinker

Mineralogical composition	Percent (%)
C_3S	47.11
C_2S	27.96
C_3A	9.46
C_4AF	10.44

The above-mentioned clinker was produced from over burden limestone, clay and sand stone which were collected from the nearby raw materials sources of the factory located 2.5, 7.5 and 5km distances from the plant site, respectively.

2.6.5.2 Limestone filler: *The limestone filler used in the test program was obtained from the source of Degen limestone raw material exploration site which is 2.7 km away from the factory. While selecting the sources, the potential for being used in the cement industry was also taken into consideration. Laboratory test results indicated that, the $CaCO_3$ content was 93.75% which* *is greater than the standard requirement (75%) as given by EN 197 and the clay content, was found to be 1.15g/100g which is also below the standard requirement (1.2 g/100g).*

2.6.5.3: CEN Standard Sand: *The sand used for the study to determine the strength of cement was CEN standard sand which is well graded rounded particles having a silica content of 98 % as specified in EN 196-1 standard requirements. This CEN standard sand is delivered in plastic bags with a content of 1350 g.*

2.6.5.4 Water: *Throughout the investigation, tap water supplied for drinking consumption at Degen were used for curing the hardened mortar samples and also distilled water were used for all physical and chemical analysis works.*

2.6.5.5 Chemicals: *For the determination of the chemical and mineralogical composition as well as sulphate and insoluble residue, different chemicals were used as per the specified method of testing cement based on the European standard EN 196-2.*

2.6.6 Experimental Program

The experimental program was composed of the following major sections: Determination of the effects of fineness and percentage additions of limestone fillers on various properties of Degen Portland cement clinker, and Study of the effects of fineness ranges and percentage additions of limestone fillers on Degen Portland cements to determine the contents of the chemical composition.

2.6.7 Grinding of the raw materials

In the preparation of laboratory samples, a laboratory ball mill having 420 mm diameter was used for grinding processes. The revolution rate for the grinder was about 45 revolutions per minute. The grinding elements used were balls of different diameters. The grinding elements weight and distribution filled into the ball mill was identical in all grinding operations.

2.6.8 Suitability of Limestone for The Production of Portland Limestone Cement

Laboratory test results indicated that (Table below), the calcium carbonate content of the limestone filler is above 93% and its clay content is 1.15gm/100gm satisfying EN 197-1 standard requirements. Furthermore, the test results also fulfill the requirements of researchers which determine the chemical composition of limestone for favorable utilization as additive in Portland cement to be with CaCO3 > 9O %, Al 2O3 < 2 %, MgO < 5% and SO3 < 0.5.

Table 4: Laboratory test results on chemical composition of limestone powder samples

Chemical composition	Norm used by the Factory (%)	Test result of Limestone Powder (%)
CaCO3	≥ 90.0	93.75
SiO2	≤ 5.0	2.58
Fe2O3	≤ 0.80	0.12
Al 2O3	≤ 1.50	1.03
Cao	≥ 50.0	53.16
MgO	≤ 0.80	0.4

SO3	*≤ 0.30*	*Negligible*
K2O	*≤ 0.30*	*Negligible*
Na2O	*≤ 0.30*	*Negligible*
LOI	*≥ 40.0*	*41.76*

Comparing the test results shown in Table above with EN 197-1 standard, and with past investigators report, the limestone raw material quarried from Degen cement factory source is suitable for utilization as addition to cement clinker in the production of Portland limestone cement under controlled and specified cement quality standards.

2.6.8.1 Grinding Time: *As expected, more time is required for fine grinding than normal fineness. However, the addition of limestone filler reduces the grinding time and also decreases energy consumption of the mills as compared to the production of the same cement quality of pure Portland cement of the same fineness. This decrease in grinding time is due to softness of limestone and easiness to grind than pure Portland cement clinker.*

2.6.8.2 Consistency of Cement Paste: *Normal consistency tests were conducted to observe the changes in water requirements of pastes due to limestone additions. The test results reveal that the consistency of cement pastes slightly decreases with increase of blain fineness values and with increased proportion of limestone content of the cement (Table 5). This decrease in consistency is due to the addition of limestone which increases the plasticity of cement paste. This may be attributed to the effect of limestone as an active component in the hydration of Portland cement, i.e., the rate of hydration increases and the amount of the hydration products enhances. The limestone fillers act as a nucleating agent which increases the hydration rate of cement paste. And also, the limestone forms monocarbo aluminate hydrate that needs less water than that of ettringite.*

Table 5: Consistency of the cement paste as a function of percentage substitution of limestone

Mix no.	*Mix Code*	*Consistency (%)*	
4000 – 4300 cm2/gm	*4301 – 4500 cm2/gm*		
1	*M1*	*26*	*26*
2	*M2*	*32*	*28*
3	*M-05*	*25*	*24*
4	*M-10*	*24*	*24*
5	*M-15*	*23*	*23*
6	*M-20*	*23*	*23*
7	*M-25*	*23*	*22*
8	*M-30*	*23*	*22*
9	*M-35*	*22*	*22*

It is observed from the test results that for the same range of blain fineness value the water requirement of DPPC is higher than that of DOPC and limestone added cement. This increase of water requirement is due to increase of surface area of cement particles of DPPC, a slightly lower hydration reaction of DPPC to that of DOPC and limestone added cements. However, *for the 5% addition of limestone fillers the water requirement doesn't show significant difference comparing to DOPC.*

2.6.8.3 Setting Time: *The EN 197-1:2000 limits the initial setting times for composite Portland cement not to be less than 45 minutes. Ethiopian standards also specify initial and final setting time for Portland Pozzolana cement (ES C.D5.202, Section 4.2.4) to be 45 minutes and 600 minutes, respectively. Comparing the obtained test results of investigation indicated in Table 8, all limestone added cement produced satisfy the requirements specified by both European and Ethiopian standards.*

The test results also indicate that, limestone addition into Portland cement increases the initial and final setting times considerably compared to Degen OPC due to dilution of C3S and C3A content in the cement. However, Degen PPC shows slightly higher setting time compared to limestone added cement. As expected, it is also noticed that as the fineness increases from Blain fineness value ranging from 4000-4300 cm2/gm to Blain fineness value range of 4300-4500 cm2/gm, the setting time of the limestone added cement is slightly accelerated.

2.6.8.4 Soundness: *Le-Chattier apparatus was used to conduct soundness test. No remarkable effects are observed on the soundness for replacement of cement clinker up to 35% by limestone additives. This negligible soundness effect can be attributed to less content of MgO and free Cao in the raw materials, fineness of the cement, good chemical and mineralogical composition of the clinker and good burning temperature in a kiln which favor a decrease of free periclase to occur.*

III. METHODOLOGY

To finish this paper, we use Secondary data from the following sources:

- ✓ *The annual Statistical Abstract (report) of the FDRE, Central Statistical Agency;*
- ✓ *Surveys, researches and other resources from the Ministry of Construction and Urban Development and Ministry of Industry of the FDRE;*
- ✓ *Relevant bulletins, statistical reports and reference books.*
- ✓ *Topographic map of the study area with 1:250,000 scale.*
- ✓ *Previous reports and publications relevant to the study area.*
- ✓ *Field structural and lithological mapping and*
- ✓ *Our sedimentary field work of Degen-Gehatsion area*

The researcher's collected samples during field work were taken to laboratory for treatment and standard laboratory preparation prior to analysis. Geochemical analysis of major elements was done using an X-Ray Fluorescence Spectrophotometer (XRF) at the deem Portland Cement factory Laboratory. Analytical precision is better than 3% for the major oxides. Total iron was expressed as Fe2O3

IV.CONCLUSIONS AND RECOMMENDATIONS

4.1 CONCLUSIONS

The future demand of Ethiopia for cement, like many other construction materials is a function of a number of interrelated variables. Some variables that is essential in determining the magnitude and trend of demand for cement are-

- *The overall economic development level and growth trend of the country,*
- *The pattern and growth trend of the construction industry,*
- *Expected technological changes that affect the structure of the construction industry,*
- *Government policies and regulations that have impact on the future level and trend of construction activities, and*
- *Size of population and its growth rate etc.*

Generally, review of sources put forward that Ethiopia has greater cement demand driver's potential for higher cement consumption to come mainly due to continued and robust GDP growth. Some of those demand driver's factors are; -

- ✓ *Political stability,*
- ✓ *increasing per capita income and emergence of middle class,*
- ✓ *increasing government capital budget expenditures,*
- ✓ *rapid urbanization (4.3 %),*
- ✓ *high rate of population growth (2.6 %),*
- ✓ *significant housing shortages,*
- ✓ *large infrastructural gap and*
- ✓ *development of industrial parks (29.37 million square meters).*

Besides, in order for the country to realize the dream to join middle-income countries by 2025, several other mega projects will be undertaken in years to come. The total estimated annual local production is 26.21 mln of MT. but our country's production capacity is around 60 % of their annual estimated production capacity. As a result, the country's' annual production is 15,726,000 MT.

Generally, the general trend of cement production and consumption in Ethiopia has been increasing. Yet the prevalence of underutilized cement production capacity remains outstanding challenge for Ethiopian's cement industry. Currently, the average cement production capacity utilization rate in the country is about 50 %. The level of capacity utilization even compared to global average of 60 to 70 % or recommended acceptable optimum production capacity utilization rate that ranges between 80 to 85 % is sustainably low Recommendations

4.2 RECOMMENDATIONS

The following recommendations are forwarded.

1. Portland limestone cement (limestone added Portland cement) is one of the alternative cementing materials well suited to our construction industry with technical, economic and environmental benefits. Therefore, concerned bodies should be made aware and promote production and use of these cement type for appropriate functions.

2. Since there is abundant quantity of limestone with necessary chemical composition, it is possible to produce Portland limestone cement at Degen cement factory.

3. In Ethiopian construction industry, little is known on availability of different alternative cement types except OPC and PPC. Therefore, cement production companies as well as political decision makers together with professionals particularly material engineers should take an active part to exploit the existing resource for using it economically.

4. Some basic physical and chemical properties of limestone added cements were investigated. However, further investigation can be made on the following topics:

a) Effects of different fineness values and compositions of limestone fines on Portland cements

b) Study the durability characteristics of the produced cements such as sulfate resistance and reactivity with alkali reactive aggregates.

c) Properties of fresh and hardened concrete prepared with limestone added Portland cements.

d) Cost benefit analysis of Portland limestone cements.

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