

## DECIPHERING THE GENETIC EVOLUTION OF EPISODIC DIAGENETIC MODIFICATIONS IN CRETACEOUS KAWAGARH FORMATION, HAZARA BASIN, PAKISTAN; INSIGHTS FROM GEOCHEMICAL INVESTIGATIONS

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### Abstract

The Cretaceous Kawagarh Formation is exposed in various parts of Hazara Basin. The Formation consist of micritic limestone and dolomite. Field investigations, petrographic studies, Mineralogical studies, geochemical analysis and isotopic analysis are used to decode the formation and mechanism of dolomites in the Kawagarh Formation. Two sections were sampled (Bagnothar and Kala Pani) to find out the mechanism of dolomitization. Three types of dolomites were recognized according to grain size and grain textures i.e. medium to coarse grain matrix replacive planar-p dolomite (RD1), medium grain matrix replacive subhedral dolomite (RD2) and medium to coarse grain matrix replacive nonplanar dolomite (RD-3). Beside replacive dolomites, two type of dolomite cements i.e. fracture filling fine grained dolomite cements (CD1) and coarse grain dolomite cements (CD2) were also reported. Voids and fracture filling cementing calcites (CC) i.e. Twin and syntaxial were also reported. XRD study shows that all the phases' dolomites are stoichiometric and of high order ranging 0.925945 to 0.952155 (with an average value of 0.921275). The analytical results demonstrate that dolomites have high Fe and Mn contents but low in Na and K content. Isotopic analysis indicates that dolomite phases show a varied range  $\delta^{18}\text{O}$  signature. Dolomites are highly depleted in  $\delta^{18}\text{O}$  ranging -3.89‰ to -4.94‰ V-PDB (with an average value of -4.66‰ V-PDB). Dolomites and calcites support the diagenetic alteration of the Kawagarh Formation by dolomitization temperatures. The stable carbon isotope  $\delta^{13}\text{C}$  values of all dolomite phases slightly depleted from the original ranging 0.77‰ to 1.32‰ V-PDB (with an average value of 1.08‰ V-PDB). The  $\delta^{13}\text{C}$  values of carbonates rocks is less affected by temperature. Compactional features in field observation, grain to grain contacts and medium to coarse grained dolomites having irregular boundaries between dolomites crystals in petrographic studies are the clear indication of deep burial. Geochemistry, mineralogical studies and isotopic results also support the deep

*burial dolomitization.*

## INTRODUCTION

The geology of dolomite and dolomitic rocks is a major field of research. Dolomite formed from the diagenetic alteration of limestone, calcium in limestone partially replaced by magnesium, usually the magnesium rich fluids move through in the limestone. Carbonate diagenesis resulted in the formation of dolomites. 50% of the world reservoirs rock for oil and gas are carbonate rocks (Zenger et al., 1980). The production and deposition of carbonate rocks strongly effect by tectonic, hydrodynamic regimes, space of accommodation, temperature, ocean water composition and siliciclastic input (Li et al., 1999, 2000; Schalger, 2000; Pirllet et al., 2010). After the burial of carbonate rocks changes occur in the fabric, composition and rigidity and porosity in the form of Neomorphism, Compaction, Dolomitization and Dissolution (Smith and Simo, 1997; Machel, 2004; Nichlos, 2009; Lapponi et al., 2013).

This article demonstrated the comprehensive study of cretaceous Kawagarh Formation in the Hazara Basin. This study is based on Field observation, Transmitted light Petrography, Mineralogical study,

Geochemistry and Carbone, Oxygen Isotopic data were used to analyze the genesis of dolomite in the Kawagarh Formation in the Hazara Basin. In the Hazara Basin, previous research studies have only documented about the environment of deposition on the basis of fossils studies (S. J. Sameeni et al., 2007; Saif et al., 2016;) however these researchers have not comprehensively investigated the origins of dolomite.

### Geological Setting:

The Kawagarh Formation stratigraphically widespread in the Kohat-Potwar and Hazara basin. Cretaceous-Tertiary (C-T) Boundary stratigraphically marked by the Kawagarh Formation because of late cretaceous age. Tectonically the uppermost part of Kawagarh Formation marks the period of initial collision between Indian and Eurasia and any light on its depositional and sedimentary history will have important bearing on the tectonic history of the Indian plate margin. Tectonically the study area lies in between the Main Boundary Thrust (MBT) and Panjal Thrust (Fig. 1).

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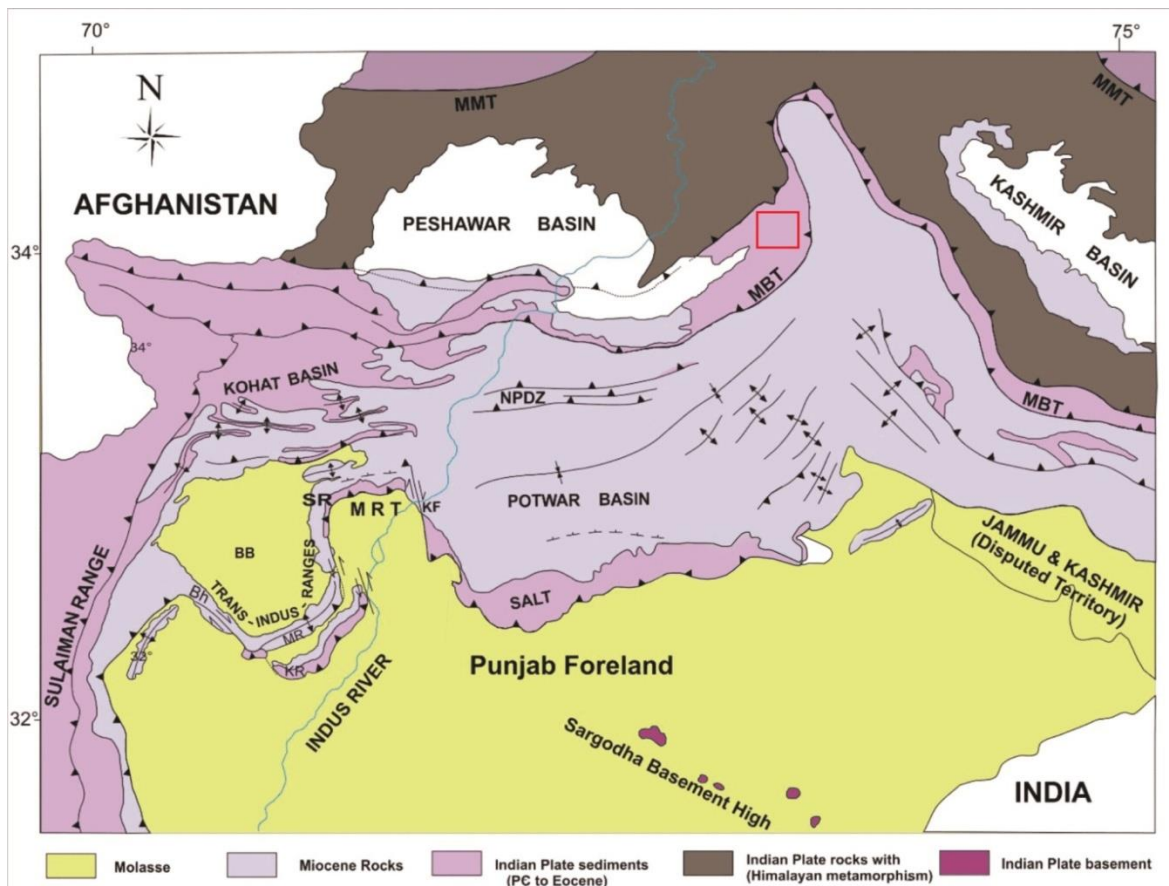


Figure 1. Geological map of northern Pakistan (Kazmi and Rana,1982), showing the location of study area as indicated by red box.

Stratigraphy of the Study Area is ranging in age from Jurassic to Eocene (Umer et al., 2014, Fig. 2). Jurassic age Samana Suk Formation is exposed, which comprise of Limestone, thin bedded Marl, Shale and also the fossiliferous bed of shell legs. Cretaceous age rocks are Chichali Formation (consist of dark green, greenish grey sandstone with dark grey sandy, silty, glauconitic shale and clayey nodules), Lumshiwal Formation (consist of grey color of limestone and

dolomite, fine grained to medium grained sandstone) and Kawagarh Formation (consist of limestone, interbedded dolomite, sandy dolomite and shale). Paleocene age rocks are Lockhart Formation (comprise of nodular limestone and marl) and Patala Formation (comprises of Khaki shale and occasional limestone bands).

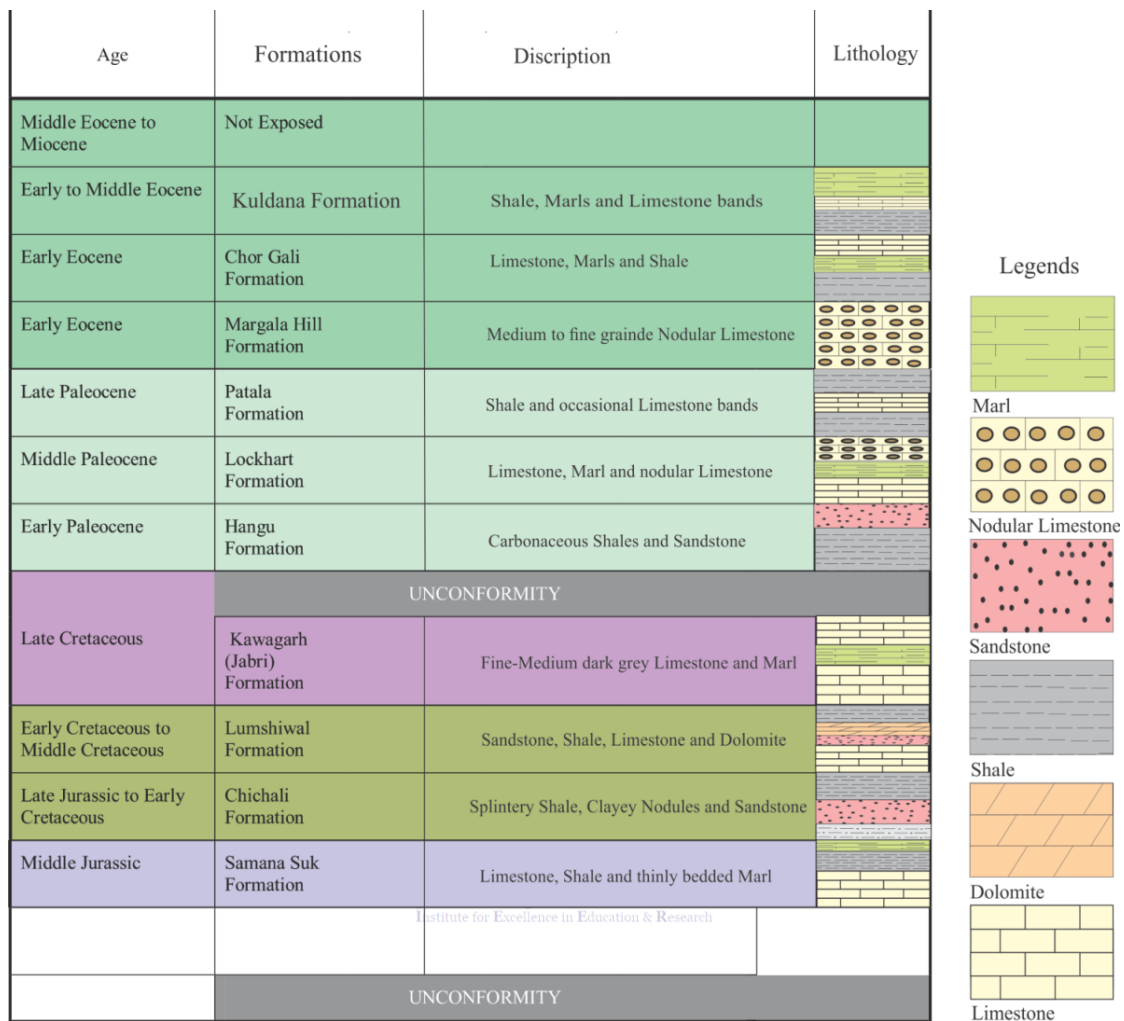


Figure 2. Generalized stratigraphy column of the Hazara Basin (modified after Latif, 1970).

**Materials and Methods:**

Two sections were studied and sampled i.e. Bagnothar and Kala Pani. Bagnothar has latitude 34° 07' 38" N and longitude 73° 20' 36" E, and Kala Pani has latitude 34° 12' 35" N and longitude 73° 19' 51" E respectively. Both sections were sampled on the variation base i.e. Color, Textures and Grained size. Kawagarh Formation has beds of different colors and have different features i.e. Stylolite, Calcite veins and fractures. In the proposed area, the Kawagarh Formation comprised of limestone, interbedded dolomite, sandy dolomite and shale. Finger like weathering present which is the diagnostic feature of Kawagarh Limestone and the dolomite shows butcher chub weathering. The Kawagarh Formation is known to contain upper

Cretaceous Planktonic Foraminifera (Latif, 1970). Different species of Globotruncana are commonly distributed throughout the Formation. Thin sections were prepared from the representative samples and stained with the Alizarin Red S solution Dickson (1966). Staining was used to differentiate the calcite phase from the dolomite. The petrographic analysis is based on thin section study, using digital camera, fitted Nikon petrographic microscope. The standard classification of Friedman (1967) and Sibley et al., (1987) were followed for the studied thin sections. The petrography was based on detail study of Allochem, Matrix, Crosscut relationship, Cementation, Textural features and Fossils.

XRD analysis was conducted in the Geosciences Lab of Geological Survey of Pakistan (GSP) and in the National Center for Physics (NCP). For XRD 5g samples of each observed phases of dolomite and calcite were crushed to 200 mesh grain sizes in agate mortar. The samples were then analyzed through the PANalytical Instrument Inc. XRD peaks were generated from the resulted data by using Xpert Pro. Elemental analysis was done in the Environmental Geochemistry lab of Chemistry department, Quaid-i-Azam University Islamabad. The atomic absorption method and Mass spectroscopy methods were applied to the selected samples for the determination of Ca, Mg, Fe, K, Na, and Mn. About 0.5 to 1 gram of crushed sample was dissolved in 1:3 of HNO<sub>3</sub> and HCL. The dissolved samples for the further digestion were put on hot plate on temperature 110-120 °C. Following the mechanism further after the digestion samples was filter using filter papers-42. And then add 50ml of de-ionized water to the digested samples. These digested samples were highly concentrated so dilute it further for the determination limit of machine.

Oxygen and Carbone analysis were performed in the Pakistan Institute of Nuclear Sciences and Technology (PINSTECH). Stable C and O results obtained from dolomite by focusing a laser beam with a wavelength of 1064nm and diameter 20µm onto a sample placed in a vacuum chamber to ablate a small area on the sample and release CO<sub>2</sub> gas. The CO<sub>2</sub> gas was led directly into a Finnegan MAT 252 mass spectrometer (Thermo Electron Corporation, Waltham, MA, USA) for isotopic analysis. The O and C isotopic data herein were recorded using conventional δ<sup>18</sup>O and δ<sup>13</sup>C notation with respect to V-PDB standard respectively.

## Results and Discussion:

### 4.1. Petrography:

Petrography of the Kawagarh Formation was carried out on prepared thin sections of the collected rock samples. Diagenetic events i.e. micritization, neomorphism, chemical compaction, mechanical compaction, dolomitization and cementation. Three dolomite phases were distinguished on the bases of crosscut relationship: Medium to coarse grain matrix replacive planar-p dolomite (RD-1), Medium grain matrix replacive subhedral dolomite (RD-2) and Medium to coarse grain matrix replacive nonplanar dolomite (RD-3).

#### 4.1.1 Medium to coarse grain matrix replacive planar-p dolomite (RD-1):

RD1 is composed of euhedral crystal and have clear boundaries. Crystals of RD1 are inclusion rich. Crystal size range 50 to 100µm. The intensity of dolomitization varies from isolated rhombs floating in the micritic matrix (Figs. 3A-C) to idiotopic and hypidiotopic mosaic texture. RD1 came after the CC1 directly (Fig. 3A).

#### 4.1.2 Medium grain matrix replacive subhedral dolomite (RD-2):

RD2 is composed of subhedral crystal and unimodal. RD2 is mostly concentrated and tightly packed (Figs. 3D-F). Crystal size range 50 to 100µm. Crystal of RD2 is inclusion rich.

#### 4.1.3 Medium to coarse grain matrix replacive nonplanar dolomite (RD-3):

RD3 is composed of non-planar sucrosic dolomite crystal (Sibley and Gregg, 1987) of uniform size occurring as replacive phase. Crystal size range 50 to 150µm. These crystals of RD3 display interlocking pattern (Figs. 3G-I) and has cloudy appearance indicating the presence of fluid inclusion and micropores. Regarding of crystal shape this phase is fabric destructive resulting in the complete overprint of the precursor limestone.

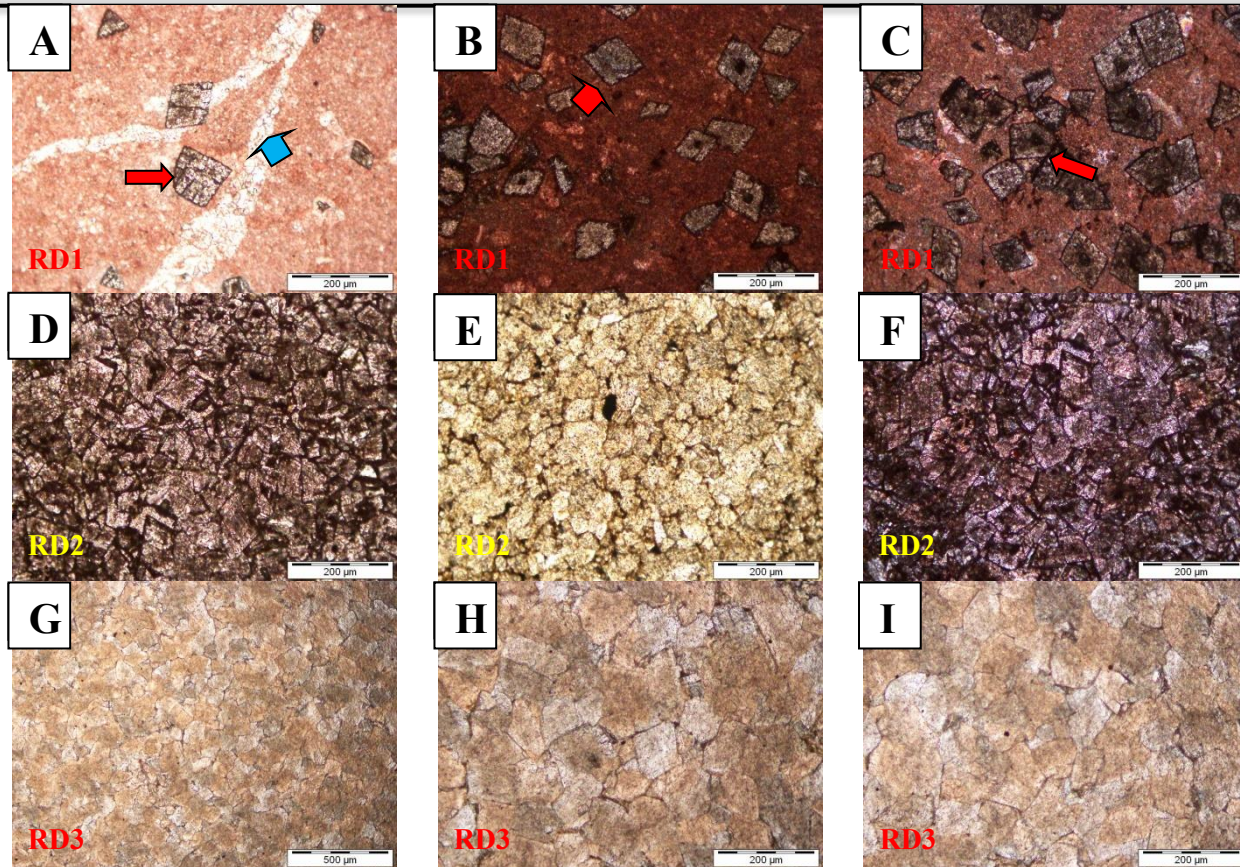


Figure 3. Plain and cross polarized light photomicrograph of the representative thin sections demonstrates different type of dolomite observed in the Cretaceous Kawagarh Formation. (A,B,C) Medium to coarsely crystalline matrix selective replacive planar-p dolomite (RD-1) indicating by red arrows while the blue arrow points out the CC1 in A. (D,E,F) Indicates medium crystalline matrix replacive pervasive dolomitization with subhedral texture (RD-2). (G,H,I) Exhibits medium to coarsely crystalline matrix replacive nonplanar dolomite (RD-3).

#### 4.1.2 Dolomite Cements:

Fine grained fracture filling dolomite cements CD1 and coarse-grained pores filling dolomite cements are mostly CD2 present in the medium to coarse grain matrix replacive nonplanar dolomite (RD-3). Fine grained dolomite CD1 cements mostly reported from fractures and the crystal size ranging 5 to 10 $\mu$ m

(Figs. 4A-C). Coarse grained dolomite cements CD2 composed of 50 to 300 $\mu$ m crystalline euhedral to subhedral dolomite rhombs with moderately unimodal crystal (Figs. 4D-F). Coarse grained dolomite cements volumetrically in minor amount throughout the Kawagarh Formation in the Hazara Basin.

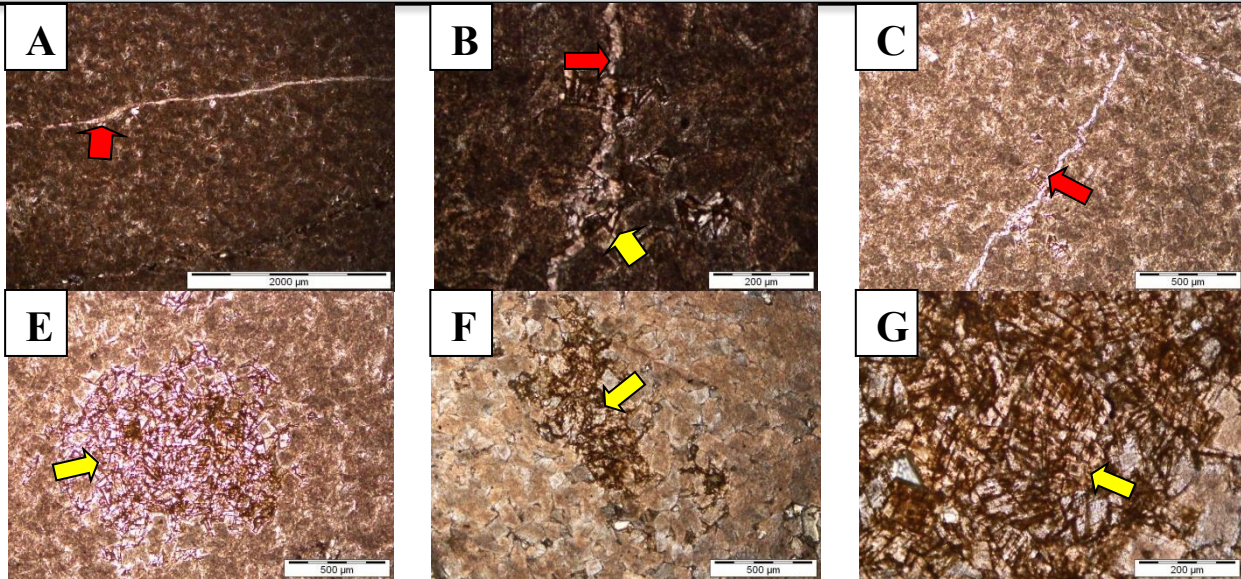


Figure 4. Photomicrograph shows different type of dolomite cements. (A,B,C) shows fine grained fracture filling dolomite cements (CD1) red arrows point out the CD1 and yellow arrow in B pointing the CD2, (E,F,G) shows the coarsely crystalline pores filling dolomite cements are mostly (CD2).

4.2 X-Ray Diffraction Analysis:

XRD data of different dolomite phases of Bagnother section do not indicate any remnants of parent rock while results from the Kalapani samples indicate the precursor limestone. Bagnother section is mostly dolomitized from 40 to 60% and stoichiometric and have high order. Sharp peaks were obtained for all the dolomite samples for stoichiometry. Dolomites reported in Kawagarh Formation from Kalapani section are non-stoichiometric. Lumsden (1979) equation was used to find out the stoichiometry. The results are shown in the (Table 1).

Medium to coarse grain matrix replacive planar-p dolomite (RD-1) are nearly stoichiometric (48.0537

%mol Ca), medium grain matrix replacive subhedral dolomite (RD-2) are non-stoichiometric (33.247 %mol Ca) and medium to coarse grain matrix replacive nonplanar dolomite (RD-3) are stoichiometric (50.03 %mol Ca). Stoichiometry versus  $d_{104}$  show in (Fig. 5). All of the dolomite is of high order (from 0.915945 to 0.952155). Commonly degree of order positively related to the intensity of dolomitization, crystallinity and temperature Zhang, Z.X et al., (2012). In deep burial environment temperature increase and  $Ca^{2+}$  ions replaced by  $Mg^{2+}$  ions, so the degree of order is increasing. Therefore, the deep buried formed dolomites have the higher degree of order (Fig. 6).

Table 1. Exhibits the mineralogical analysis (X-Ray Diffractometry) values of different diagenetic phases.

S. No	Samples	Phases	$d_{104}$	a=b(Å)	c (Å)	%mol Ca	015/110
1	KP-1	Partially Dol-	2.82621	4.996	16.964	30.874	0.916261
2	KP-4	Partially Dol-	2.82501	4.991	16.972	29.6705	0.916364
3	KP-5	Limestone	2.82379	4.993	16.917	29.2639	0.916102
4	BK-9	Limestone	2.83547	4.991	16.922	33.247	0.916008

5	BK-1	RD-2	2.88609	4.805	16.022	50.03	0.915969
6	BK-14	RD-1	2.88538	4.808	16.022	49.793	0.915945
7	BK-15	RD-3	2.88016	4.8038	16.006	48.0537	0.952155

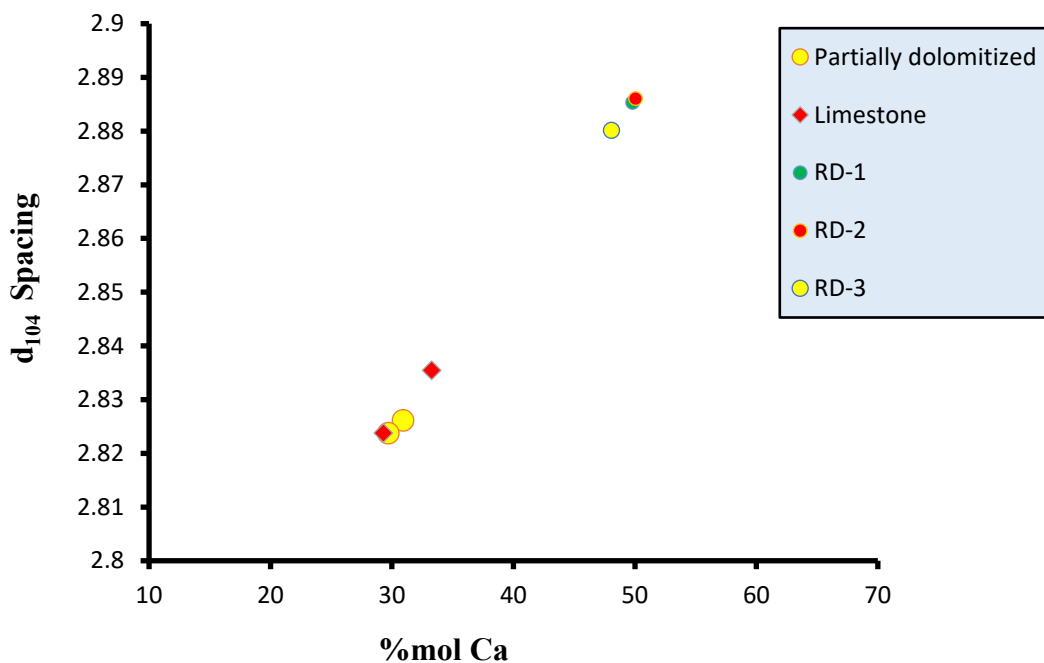


Figure 5. Cross plot  $d_{104}$  versus stoichiometry of the dolomite bodies observed in Kawagarh Formation.

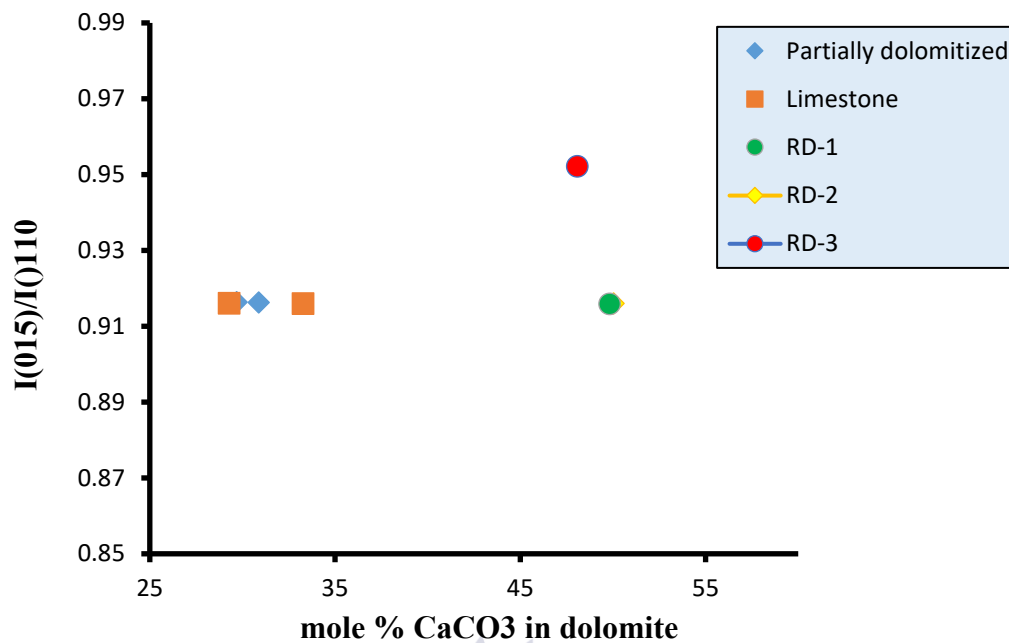


Figure 6. Cross plot showing degree of ordering versus stoichiometry of Kawagarh Formation dolomites (I-integral intensity of XRD peaks)

#### 4.3 Major Elements and Trace elements:

The dolomite of Kawagarh Formation shows the wide range of Ca and Mg ranging from 166.5ppm to 314.7ppm (in percentage it is 1.66% to 3.147%) and 48.47ppm to 126.5ppm (in percentage 0.004847% to 0.01265%). Variation in Ca and Mg ratios shows the degree of calcite replacement by dolomite Zheng Jianfeng et al., (2012). The value of Ca and Mg is less than the theoretical values in stoichiometric dolomite. The lower Ca and Mg values and exhibit a negative correlation between Ca and Mg indicate that the formation of dolomite occurred after deep burial (Fig. 7). The dolomite of Kawagarh Formation has high Fe and Mn content and low Na content (Table 2). Fe and Mn content range from 28.57ppm to 80.2ppm (with an average value 43.8227ppm) and 1.468ppm to 2.317ppm (with an average value

1.38085ppm) (Fig. 7). Fe and Mn in dolomites show the intensity of diagenesis and burial depth. Intensity of diagenesis and higher content of Fe and Mn are associated with greater depth Reinhold, C (1998). Mn and Fe contents are very low in the sea water while it is in diagenetic pore fluids Zhao Weiwei et al., (2011). Na is the best indicator of paleosalinity in the ancient fluids and the most abundant cation in marine water. Dolomite reported from the Kawagarh Formation has the lowest Na content with average 3.356ppm. The major and trace elemental results of Kawagarh Formation in the Hazara Basin indicating that the dolomite are formed after burial.

Table 2. Shows the elemental results of different facies and phases observed in the Kawagarh Formation in Hazara Basin

S. No	Samples	Phases	Ca	Mg	Na	K	Fe	Mn	Mg
1	BK-1	RD-2	171.7	125.9	3.375	4.028	80.2	2.317	125.9
2	BK-4	RD-1	314.7	118.2	1.383	3.039	29.96	1.598	118.2
3	BK-5	RD-2	215	126.5	4.356	1.561	76.55	1.526	126.5
4	BK-9	limestone	368.5	48.47	9.604	3.42	51.78	0.457	48.47
5	BK-14	RD-1	384.9	118.4	1.333	3.126	28.57	1.468	118.4
6	BK-15	RD-3	166.5	126.4	4.184	5.144	76.68	1.703	126.4
7	KP-4	limestone	312.3	65.92	1.552	0.637	1.604	0.339	65.92
8	KP-5	limestone	286.2	49.29	1.061	0.69	5.238	1.639	49.29

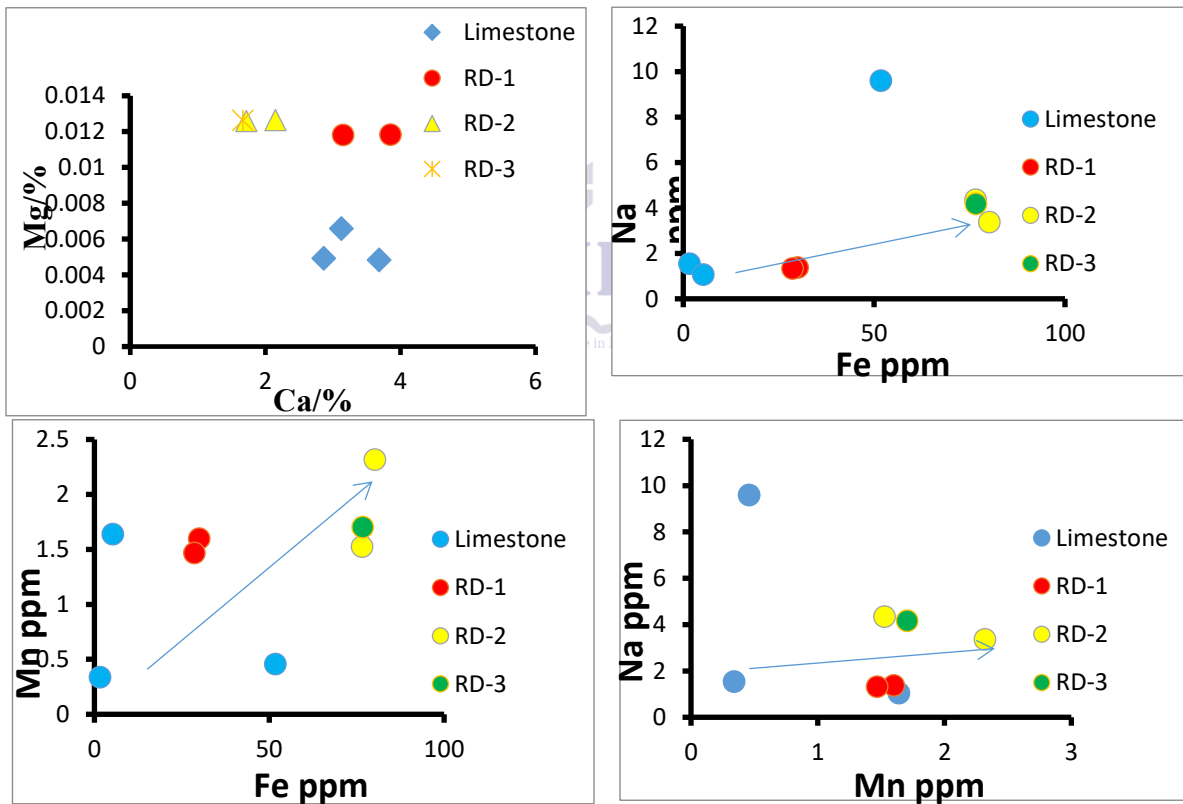


Figure 7. Cross plots of major and trace elemental compositions i.e., Na and Fe, Mn and Fe, Na and Mn contents observed in the representative samples of the Kawagarh Formation.

4.4 Isotopic Analysis of Carbon and Oxygen:

Selected samples of different phases of dolomite were analyzed for the  $\delta^{18}O$  and  $\delta^{13}C$ . Stable isotopic

analysis of  $\delta^{18}O$  and  $\delta^{13}C$  were used to find out the changes and variation that alter the Kawagarh

Formation at different sections. Samples for isotopes of  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  were selected on the basis of petrographic studies and were prepared through dentist drill machine from different phases. The cross plots of  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  values from both Bagnothar and Kalapani areas showing marine isotopic signatures range of cretaceous seawater ( $\delta^{18}\text{O} = -2.0$  to  $-2.5$  V-PDB ‰ and  $\delta^{13}\text{C} = +2.0$  to  $+3.0$  V-PDB ‰) considered as starting values of the host limestone prior to interaction with magnesium rich fluids (Fig. 8) (Moss and Tucker, 1995).

Numerical isotopic values of different phases of dolomite and calcite observed during geochemical investigations (Table 3). The replacive dolomite RD-2 numerical values of carbon and oxygen varies from

(1.32 V-PDB ‰) and (-3.89 V-PDB ‰) respectively and RD-3 numerical values of carbon and oxygen varies from (1.15 to 0.77 V-PDB ‰) and (-4.57 to -4.94 V-PDB ‰). The calcite cement phase, carbon and oxygen isotopes values ranges from (2.1 to 0.94 V-PDB ‰) and (-6.48 to -7.16 V-PDB ‰). KP-4 and KP-1A stands for Limestone the numerical isotopic values of Carbon and Oxygen ranges from (1.5 to 2.1 V-PDB ‰) and (-6.48 to -6.71 V-PDB ‰). Depleted values of  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  from the original marine signatures i.e.  $\delta^{18}\text{O} = -2.0$  to  $-2.5$  V-PDB ‰ and  $\delta^{13}\text{C} = +2.0$  to  $+3.0$  V-PDB ‰ show the deep burial condition for dolomite.

Table 3 Exhibits the stable Carbon and Oxygen isotopic signatures of different phases of dolomite

S.No	Samples ID	Different Phases	$\delta^{13}\text{C}$ ‰ (V-PDB)	$\delta^{18}\text{O}$ ‰ (V-PDB)
01	BK-1	RD-2	1.32	-3.89
02	BK-5	RD-3	1.15	-4.57
03	BK-6	Calcite	0.94	-7.16
04	BK-9A	Calcite	1.64	-7.14
05	BK-15	RD-3	0.77	-4.94
06	KP-1A	Limestone	2.1	-6.48
07	KP-1B	Calcite	1.75	-6.71
08	KP-4	Limestone	1.5	-6.71

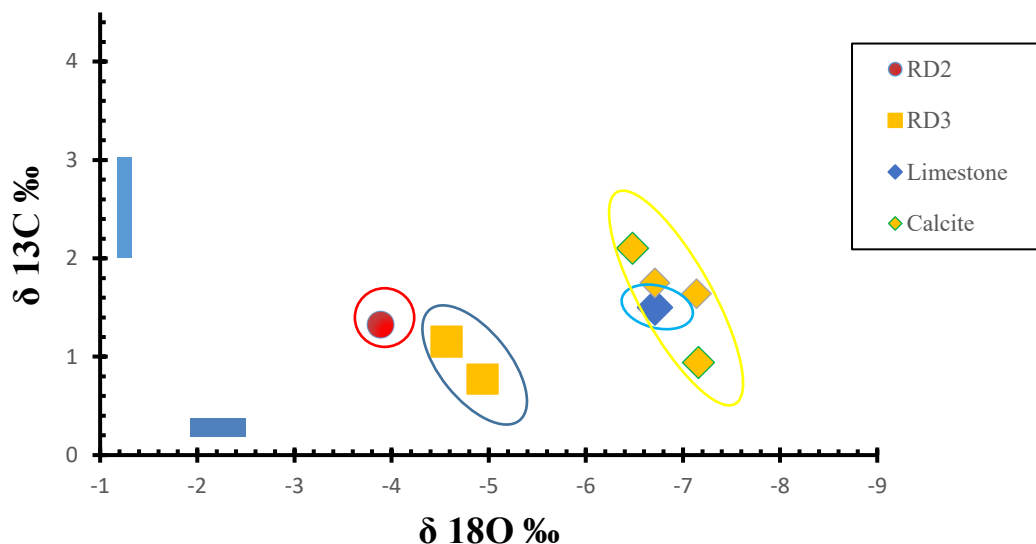


Figure 8. Demonstrates stable isotopic cross plot of various phases observed in the studied formation

#### 4.5 Proposed Dolomitization Model:

Dolomitization model for Kawagarh Formation was developed based on petrographic studies and geochemical analysis. Both field and petrographic studies of Kawagarh Formation shows no signs of evaporate/anhydrites minerals. As there no such evaporitic minerals reported from the field and thinsection of the Kawagarh Formation reveals that hypersaline model has no role in the dolomitization of the formation. Hydrothermal model cannot be proposed to the formation, because field observation and geological map of the study area shows no evidence of igneous intrusion, any faulted contact of the formation or within the formation. Furthermore, the stable oxygen isotope  $\delta O^{18}$  value are slightly depleted from the range of cretaceous marine signature indicating that hydrothermal fluids were not responsible for the formation of dolomite in the Kawagarh Formation.

Petrographic observations also show that coarse grained euhedral to subhedral dolomite are mainly clustered along the stylolite. The occurrence of dolomite along the stylolite shows that dolomitization postdates chemical compaction and the stylolite suture facilitates the passage of brines

that carry the ions to precipitate coarse grained euhedral to subhedral dolomite crystals. The geochemical results also support the previous petrographic observations. The stable isotopic values of  $\delta^{18}O$  and  $\delta C^{13}$  values of all dolomite phases are depleted (Fig. 8).

Field observation, Petrographic evidences, mineralogical study and isotopic analysis suggested the burial model for dolomitization. The coarse grained dolomite with non-planner surface is the evidence for deep burial and shows temperature above  $60^{\circ}C$  (Spotl and Pitman 1998). Coarse crystalline and stoichiometric dolomites with depleted isotopic values of  $\delta^{18}O$  and  $\delta C^{13}$  (Wilson et al., 1994; Nielsen et al., 1998; Boni et al., 2000; Gasparini et al., 2003; Wilson et al., 2007; Shah et al., 2010) fulfill the requirements for dolomite formed in deep burial condition.

Due to thick sedimentation (round about 2232m as per published stratigraphic sequences) over the Kawagarh Formation, the Mg-rich fluids expel upward from the underlying Lumshiwal Formation and Samana Suk Formation as basinal brines and caused dolomitization in the Kawagarh Formation (Fig. 9).

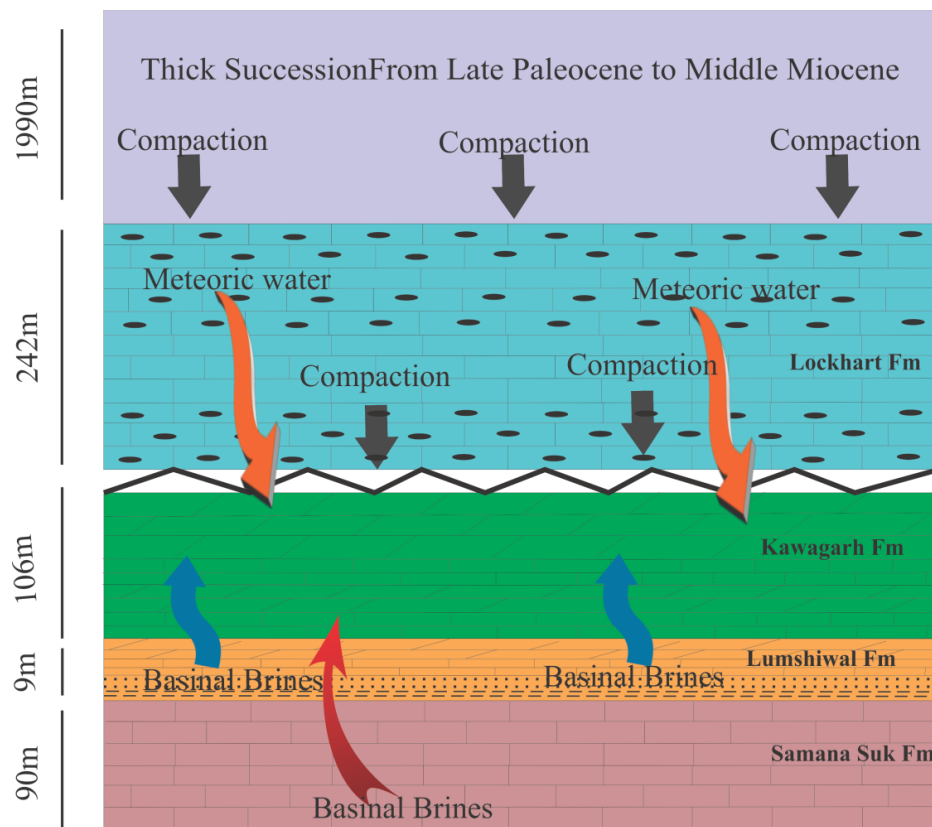


Figure 9. Sketch illustrating a proposed diagenetic model for Kawagarh Formation

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