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Polymorphs of silica in the form of white pebbles detected at Lonar Crater, Maharashtra, India

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ABSTRACT

For the first time polymorph of silica in the form of white colored pebbles of spherical and elongated shapes has been detected at Crater Lake and ejecta, on a large scale which was investigated in a course of research to understand its characteristic features. Elemental composition and oxides of these pebbles through XRF analysis showed Silica (SiO_2), to be 97.985 mass % with negligible quantities of CaO, Fe_2O_3, SO_3, Al_2O_3, and Cr_2O_3. Physicochemical analysis carried out showed that these pebbles are not related to salinity or alkalinity. The high content of SiO_2 (Silica), showed that it is a polymorph of silica. Findings of silica pebbles at Lonar Crater have not been reported by earlier workers. The formation of silica pebbles is related to Lake water. Formation of silica pebbles and the presence of diatoms in the lake water indicate that the lake water contains a high percentage of SiO_2, the source of which is unknown. Pebbles are generally found near river beds or near the seashores. For which lotic water body is responsible. These pebbles contain a high percentage of CaO. Findings of large quantity of silica pebbles at Lonar crater in which SiO_2 is 98% mass, this is unusual because white pebbles containing high percentage of CaO is known, but white pebbles containing high percentage of silica (SiO_2) is unknown and has not been reported from any of the volcanic eruption sites nor has it been reported from any of the meteorite impact crater sites either, also these silica pebbles are not found in large quantity on the earth surface. Silica pebbles being a rich source of silica and found in abundance here, an industry of making glass beads may have flourished here in the 14th century. This part has not been explored in the past and is a preliminary investigation, this paper reports the first-hand information of findings of silica pebbles, a polymorph of silica and to put on record the findings. A standardless approach has been adopted.

Keywords— *Lonar crater, Silica pebbles, Lake water, Milky white, Polymorph of Silica*

1. INTRODUCTION

A circular depression excavated by a meteorite impact, as per latest studies, near a small town named Loa=nar in the southern part of Buldhana District of Maharashtra, named as Lonar Meteorite Crater. It is one of the unique Craters in the world, formed on basaltic rocks of volcanic origin. The crater has a well-preserved rim, and the circumference of the rim is ~5.652, the ejecta is also well preserved. The crater has been studied by different researchers in view of studying meteorite impact on basaltic rocks and similar crater formations on basaltic rocks of other planetary systems.

1.1 Some of the previous studies carried

Misra et al. studied impact spherules from Lonar crater and states that, the distribution of meteoritic components between the impact melt clasts and spherules is also an important aspect of the evolution of Lonar crater[1]. Newson et al. stated that the ejecta blanket at Lonar extends beyond 1350m from the rim with discontinuous patches as far as 3000m[2]. Lonar Crater is probably a meteorite crater[3]. The latitudes of which are 19°58' and longitudes 76°30', situated in a southernmost part of Buldhana District of Maharashtra. According to Nandy and Deo, it has been proved that the depth of the lake from the present crest is not less than 600 ft, whereas the maximum thickness of lamellas is known to be only 100 ft.[4]. Hagerty and Newsom stated that the comparable compositions of Lonar basalts and martian basalts suggest that similar alterations products should have been produced on Mars due to impact-induced hydrothermal activity[5]. Maloof et al. stated that unlike Barringer crater, no significant vertical displacement of the basalt flows in the crater walls was observed along identifiable tear zones[6]. Kumar stated that the impact deformation structures of Lonar are essentially similar to those at other terrestrial craters emplaced in granites and clastic sedimentary rocks[7]. Son and Koeberl concluded from their studies, the impactites and impact melt glasses collected from different locations in Lonar crater are different in shape, size, and texture[8]. A lake exists at the bottom consisting of saline and alkaline nature of water. Borul mentioned that in the present study, the data revealed that there were considerable variations in the quality with respect to their physicochemical characteristics[9]. Badve et al. reported that the pH of the lake water body ranges from 9.3-9.7[10]. Pawar recorded average pH value to be of 10.3[11], whereas Gaikwad and Sasane recorded pH value to be of 10.5[12]. Shinde and More mentioned that the lake is a closed system without any outlets and regular influents are responsible for its existence[13]. On the periphery of the lake, Vyas mentioned that temples of the 11^{th}-12^{th} century were built[14]. The continuous flow of water is seen from Dharatirtha on the eastern side of the crater, spring emerging from near Ramgaya temple is seen of which the water goes to the lake. Badve et al. mentioned that the lake basin either might have received water from the existing springs which were geothermally active and gradually lost their property in course of time or some of the hot springs got buried within the basin as the Lonar crater was lying in a structurally disturbed zone[10]. As concluded by Komatsu et al. Lonar crater is also a valuable study site on degradation processes with potential application to impact craters occurring on hydrologically active ancient Mars, or to other impact craters on earth[15]. Rajasekhar stated that meteorite impacts modify the gravity and the magnetic field at the site of impact which after the impact shows different signatures compared to the surrounding rocks[16]. Glassy objects found at Lonar were studied by Nayak and suggested that Lonar glass shows a greater resemblance with the dark glass from Henbury Crater (Australia), the dark dense glass from Ries Crater (Germany), he also stated that the glassy objects in all probability are an impact product and represent an impactite-strewn field around the Lonar crater, and the distinctive shapes reflects the splash forms that were once molten droplets[17A]. Nayak stated that the presence of impactites has been advanced as one of the chief criteria for indicating a meteorite origin of craters, and therefore, the recognition of glassy objects (resembling impact glasses) from the suspected Indian crater is of paramount significance and may eventually prove to be a reliable clue in favor of its meteoritic impact origin[17A]. A heating experiment on maskelynite conducted by Nayak, the result of which indicates that the Lonar maskelynite reverts to a crystalline state at 800°C which is lower than 900°C and 1000°C, reported for the Canadian craters[17B]. Weiss et al studied magnetic properties in glasses and concludes that paleointensity from small spherules likely provide lower limits on the paleointensity of any fields in which they formed[18]. Wright et al. studied spectra of four samples from Lonar crater and concluded that the objective of his study is to collect and report on the sample emission spectra of basaltic impact glasses and evaluate these as a possible interpretation of TIR spectra of Mars[19]. Bose et al. studied Gamma-ray mapping of the ejecta of Lonar and states that our field investigation on radioactivity around Lonar crater, however, confirms that this method can effectively be used to map the distribution of ejecta around any crater if the target is relatively old in age[20]. Stroube et al. determined 29 elements by INAA in two impact glasses and four basalts from Lonar crater and concluded that the results of this study suggest that impacts in basalt generate glasses which have an essentially unaltered composition of major rock and mineral-forming elements as compared to the parent basalts[21]. Morgan stated that Iridium is probably the most sensitive indicator element for many types of Fe-rich meteoritic material[22]. Chakrabarti et al. from Pb isotopic studies on breccias from Lonar concluded, the trace element and isotopic data presented here demonstrate that the impact breccias of the Lonar crater formed by impact-mixing of melts and clasts derived from the Deccan basalts and the Archean basement[23].

2. METHODOLOGY

White pebbles were collected randomly as a representative sample from southwest of Crater Lake as shown in Figure 1. The pebbles were washed properly with distilled water and dried in the sun as per Figure 2. Physical parameters like weight, density, size were carried out. The pebbles were weighed on a balance of making RADWAG XA-82/220/2X. The volume was taken by displacement of water method in a borosil measuring cylinders of standard quality and finally, density was calculated by using the formula M/V g/cc as shown in Table 1. For size vernier calipers of LC 0.01cm was used as per Table 1. Similarly, pebbles were collected from ejecta few meters away from Government rest house from the south-east and physical parameters were carried out as mentioned above as per Table 2. After the physical parameters were carried out, the pebbles were crushed into powdered form in a motor pestle for further analysis. The pebbles are of spherical shape while others are found to be the elongated or cylindrical shape.

Observation of powdered samples under the stereo binocular microscope of make Moticam 10 10.0MP was conducted to study its microscopic features Figure 3, followed by physicochemical analysis for pH and Chloride content as shown in Table 3 and 4. Finally, XRF analysis for elemental composition and oxides was conducted as shown in Table 5, 6 and 7.

2.1 Experimental Results

The results of observation through a binocular microscope under lower magnification showed that the powdered samples contain a tiny glassy type of crystals of unknown minerals. At higher magnification, the tiny glassy crystals were clearly seen but were unable to define the unknown minerals. The results of the physicochemical analysis Table 3 and 4 show that the samples under study have a pH value of 6.9 and Chloride content was 190mg/kg and 82mg/kg. From the results of XRF analysis, sample 1, table 5 and 6, oxides content were found to be Silica (SiO_2) - 97.985mass %, CaO - 0.571mass % and Fe_2O_3 - 1.445mass %. In sample 2, as per Table 7 oxides content were SiO_2 - 97.805mass %, SO_3 - 1.581mass %, Fe_2O_3 - 0.428mass %, CaO - 0.154mass %, Cr_2O_3 - 0.032mass %.

3. RESULTS AND DISCUSSIONS

From the results in Table 3 and 4, it infers that the pebbles do not contain any alkaline nature of salts like carbonates (CO_3) or bicarbonates (H_2CO_3), although NaCl salt is present, it is in negligible quantity. In other words, these pebbles are not related to salinity or alkalinity. White colored pebbles of CaO are generally found near river beds or near the seashores for which lotic water body is responsible for its formation. But here at Lonar white colored pebbles of silica origin are found in which CaO in sample 1 is 0.571 mass % and in sample 2 CaO is 0.154 mass % which is very negligible. In the absence of lotic water and CEO, these pebbles show a high percentage of SiO_2 which is unusual. From the results in Table 5, 6 and 7, and observation through a stereo binocular microscope, it infers that the tiny glassy like components is of Silica (SiO_2) origin. Since Lonar pebbles contain a high percentage of Silica than other oxides, hence it was referred to as Silica pebbles.

Some 65ma volcanic eruption occurred in Maharashtra. After this event volcanic activity has ceased long back. Later on some 656 thousand years back, a crater was formed[25]. As per earlier workers opinion, the formation of the crater is by a volcanic eruption. But for this assumption, no proof exists. Formation of Silica pebbles found at Lonar has not been reported from any of the workers supporting volcanic eruption theory for crater formation, nor has it been reported by workers supporting meteorite impact theory for Lunar Crater formation. In lieu of this, the formation of silica pebbles, therefore, cannot be attributed to either volcanic eruption or meteorite impact.

Quartz-pebble conglomerates have been reported by Dai et al. from Tarkwa mines, few km away from Bosumtwi impact structure[24]. Quartz pebbles conglomerates are of sedimentary origin. Silica pebbles found at Lonar are of basaltic origin and is a polymorph of silica, and can be attributed to the lake water and the surrounding conditions of the lake. Badve mentioned that the lake water contains diatoms[10], which are related to algae group, Lonar Lake is full of algae, hence the green color of the water is observed. The outer shell or outer layer of these diatoms are covered by a silica layer or in other words, it can be said that these diatoms are a rich source of SiO_2 When these diatoms die, they settle at the bottom of the lake. From the analyses of lake silt by Nandy, SiO_2 was found to higher than the other oxides[4]. Due to biodegradation, SiO_2 is released from their body and remains in the water, SiO_2 is insoluble in water hence it remains as it is but it cannot remain for a long time. The basaltic rocks lying at the lake periphery during the rainy season are submerged in the water due to an increase of water level in the lake. During this period some of the SiO_2 gets deposited on the rock surface. After the rainy season is over, the water level of the lake decreases as the month's pass. The deposited SiO_2 remains on the rock. In this way year after year this process operates. Each time some amount or quantity of SiO_2 in the water deposits on the rock and finally when the quantity or percentage of SiO_2 goes above 95% white rusting phenomena/white coating of silica on basaltic rocks is observed. The first sign of silicification seen. At this instant, the percentage of SiO_2 may be 95-97%mass. This is the first step of silicification of silica on basaltic rock or polymorphism of silica. The process continues and when the rock attains 98% SiO_2, it is a second step, which can be said of silica pebbles, because in silica pebbles, the percentage of SiO_2 is around 97-98% mass. This inference is drawn from the observation of these silica pebbles and the analysis through XRF. Hence the formation of silica pebbles can be said of the intermediate state of silicification. The third and the final step is the formation of milky white phase and glassy phase in which the deposited SiO_2 encapsulates the whole rock in which the percentage of SiO_2 is found to be 99.5-99.8mass %. At this stage, the process is almost complete and is known as silicification of basaltic rock. As basaltic rocks contain 50-52% mass SiO_2, which is a major oxide, hence it becomes easier for SiO_2 in the water to encapsulate the rock at the lake through the depositional process over the entire rock, leaching out other oxides which are present in minor quantities, thus attaining 95-99% mass SiO_2. All the three steps are of the same event i.e., silicification or polymorphism of silica i.e., the existence of silica in different forms.

The formation of silica pebbles being the intermediate process is observed at Crater Lake. But a question arises, if the white pebbles are related to water, i.e., large quantity of these silica pebbles should be found at the lake periphery, but it is observed that lesser quantity of these silica pebbles are found at the lake shore and large quantities of these pebbles are found on the rim and the ejecta for reasons unknown.

Vyas mentioned that a civilization existed here were glass beads work or the making of glass beads was carried out. It is possible that due to the availability of a rich source of silica in abundance at that time, in the form of white pebbles found at Lonar Crater Lake, making of glass beads industry may have flourished. The temples at Crater Lake were built in the 12th - 14th-century [14]. Hence it is possible that during this period silica pebbles were used by the ancients for making glass beads or it can also be said that some kind of work related to silica pebbles maybe were involved or engaged among the civilization, where these silica pebbles were used. Because Vyas mentioned that pieces of pebbles found on the upper part of the circular region of the hilly area of Lonar crater i.e., on the ejecta which was found to be cut intentionally in a precise manner or was cut precisely or purposefully[14]. Though the silica pebbles are of milky white color, their size is approximately 1cm and the shape of spherical, elongated or cylindrical and are found in large quantity on the crater rim instead of a crater lake, which means that the civilizations existing here as per Vyas were engaged in some kind of work related to silica, i.e., the rocks of milky white phase must have been cut into proper shape and size and may be, were used as glass beads or other products unknown, because after the production of glass beads, the leftover product may be the silica pebbles and since the work was carried on the rim, the byproducts which are silica pebbles were left on the rim as it was unused byproduct. This may be possible. Otherwise, there are no reasons for such a large amount of silica pebbles to be found on the rim and ejecta.

In nature, any component does not have the same shape or size. In silica pebbles, it is observed that the size of all the pebbles under study was found to be around 1cm and shape spherical and some to be cylindrical or elongated. Also in none of the volcanic eruption site findings of silica pebbles been reported, nor has it been reported from any of the meteorite impact crater sites either. But in volcanic eruption site, white rusting (silica coating) phenomena have been observed and reported and at Lonar lake also white rusting phenomena has been observed and reported[26].

It is to be stated here that in our earlier paper on “white rusting of rocks” The phenomena observed at Lonar crater lake, the formation of white rusting is due to a volcanic eruption and the environmental conditions existing at Lonar crater has been mentioned[26]. As per detail work and study conducted on white rusting/white coating and silica pebbles. It is found that the formation of white rusting/white coating found on basaltic rocks is not due to volcanic eruption, neither is it due to environmental conditions existing at Lonar Crater, but it is due to lake water, because volcanic activity has ceased long back, there has been no volcanic activity after the volcanic eruption of 65Ma. Hence due to the absence of any volcanic activity, white rusting phenomena cannot be attributed to volcanism. At the time of the volcanic eruption, volcanic rocks do contain a high percentage of SiO_2. In due course, it disintegrates and the SiO_2 content present at the time of volcanic activity may not be the same as it was at that time. The process for formation of white rusting, high percentage of SiO_2 is required around 95-97% mass, the source of SiO_2 for the formation of white rusting cannot be fulfilled by volcanism which took place 65Ma. Since the basaltic rocks coated by silica found at the Crater Lake, it shows that water is the factor responsible for its formation, because nowhere has white rusting phenomena been observed on Deccan Plateau. Hence it is to be stated here that the formation of white rusting/white coating of silica is not related to volcanism but to the lake water and its environment existing at the lake because the formation of white rusting/white coating of silica and formation of silica pebbles are one and the same event, the process is also the same only the percentage of SiO_2 goes on increasing as deposition of SiO_2 in the water goes on increasing and various forms or stages of silica is observed which is known as silicification or polymorphism of silica. Hence white rusting formation can only be attributed to the lake water and not to volcanic eruption as mentioned in our earlier paper and through this study silica pebbles has also been observed which shows that this process may be related to white rusting and being a polymorph of silica it may be formed similarly to the white rusting process as mentioned above and not as mentioned in our earlier paper of white rusting.

In lieu of this, therefore, the findings of silica pebbles on a large scale at crater rim and ejecta and to a lesser extent at Lake Basin cannot be attributed to meteorite impact of which crater was formed because at the impact site around the globe, no rocks containing a high percentage of silica has been reported. In stony meteorites as per analysis by XRF of Chelya Binsk (a stony meteorite), SiO_2 is found to be 49.6 mass % and an iron meteorite, analysis by XRF of Campo del Cielo (iron meteorite), Si was found to be 0.798 mass % and in an unknown iron meteorite analyses by XRF shows that Si is found to be absent No oxides were detected in these two iron meteorites. Hence the formation of silica pebbles must be related to the silicification process at the lake.

Although the formation of silica pebbles is a natural process and may have been formed from basaltic rocks, cutting process and shaping process of these pebbles related to some work i.e., glass beads work is manmade or artificial. What was the work of ancient civilization carried here that gave silica pebbles the precise circular shape and size of 1cm is unknown? But this is the best explanation offered as to the findings of silica pebbles at Lonar Crater on a large scale. At the time of the volcanic eruption, the basaltic rocks have high % of SiO_2 or basalts are found to be rich in SiO_2 content. In due course, disintegration takes place due to weathering conditions where SiO_2 bonds become loose and leachate out. Hence at present, if one observes that the value of SiO_2 and other oxides are not the same as it was during a volcanic eruption. Formation of silica pebbles, therefore, cannot be attributed to volcanic activity either meteorite impact.

Since the pebbles contain 97% silica, and silica is white in color, hence the pebbles of Lonar has attained milky white color. The high content of SiO_2 in splash forms of glass from Lonar which is related to tektites has been reported[27]. Presence of white rusting on basaltic rocks at Lonar Crater Lake and further formation of silica pebbles shows that all the three steps formed i.e., white rusting, silica pebbles and milky white phase and the glassy phase are of one and single event and the process for their formation operates at Lake water.

As per Chemtob studies on Kilauea flow in Ka'u Desert of Hawaiian volcanic islands, (silica coating) white rusting phenomena has been observed where volcanic eruption has taken place recently[28]. At Lonar due to absence of any volcanism, the formation of white rusting/white coating of silica, formation of silica pebbles and finally formation of milky white phase and the glassy phase are observed and can only be attributed to the lake water where, the source of SiO_2 is found to be in large quantity due to the presence of diatoms having a rich source of SiO_2 in their body. The formation of silica pebbles a polymorph of silica, therefore, cannot be attributed to volcanism or is not related to volcanism.

3. CONCLUSION

The findings of white colored pebbles/Lonar pebbles (Silica pebbles) at lake basin and subsequently on ejecta of Lonar Crater, the physical parameters carried out, observations through binocular microscope, physicochemical analysis carried and finally through XRF (X-ray fluorescence) for oxide composition, showing high percentage of SiO_2 (silica), and other oxides in minor quantities, through discussion, and the search for the formation, source, and origin of silica pebbles, a polymorph of silica, can be attributed to the lake water. Formation of white rusting/white coating of silica on basaltic rocks and formation of silica pebbles and presence of diatoms in the lake water indicates that the lake water contains a high percentage of SiO_2, the source of which is unknown. It may also be due to environmental conditions existing at Lonar lake. From the discussion, it confirms that findings of SiO_2 in large quantity than other oxides in these pebbles is unusual because it is not found in any of the volcanic eruption site or meteorite impact site or anywhere on earth accept near river beds and near the seashores, where pebbles containing high percentage of CaO is generally observed, but pebbles containing a high percentage of silica has not been reported from anywhere on earth. These silica pebbles, found in abundance or is distributed in large quantity around the affected part of the crater, it must be or it is emphasized that it must be formed by the process of polymorphism, which is a unique and highly specific physiochemical process or rare process which has not been reported from any of the craters formed on earth. Finally, it can be concluded that silica pebbles formation is a product of polymorphism or silicification.

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APPENDIX

Table 1: The physical parameter of Crater lake pebbles, Lonar

S. No.	Parameters	Description /Value
1	Color	Milky white
2	Shape	Spherical/elongated
3	Size (approximate)	1 cm
4	Average wt. of pebbles	0.79g
5	The average density of pebbles	2.38g/cc

Table 2: Physical parameters of pebbles from ejecta, Lonar crater

S. No.	Parameters	Description/Value
1	Colour	milky white
2	Shape	Spherical/elongated
3	Size (approx.)	1cm
4	Avg. weight of pebbles	0.7914 g
5	Avg. density of pebbles	2.075 g/cc

Table 3. Physicochemical analysis of the powdered form of Lonar lake pebbles

S. No.	Test Parameter	Result	Unit	Test method
1	pH(10% Solution)	6.9		SW-846-9045-C
2	Chloride	190	mg/kg	IS:3025(P-32)1988

Table 4. Physicochemical analysis of pebbles from ejecta, Lonar crater

S. No.	Test for Parameter	Result	Unit	Test method
1	pH (5 % solution)	6.9		SW-846-9045-C
2	Chloride	82	mg/kg	IS:3025(P-32)1988

Table 5: Elemental composition of a powdered form of Lonar lake pebbles by XRF

S. No.	Atomic Number	Symbol	Elements	Mass %
1	14	Si	Silicon	95.46
2	20	Ca	Calcium	1.26
3	26	Fe	Iron	3.28
			Total	100

Table 6: Elemental composition of oxides of Lonar lake pebbles by XRF

S. No.	Atomic Number	Symbol	Elements	Mass %	Formula	Mass %
1	14	Si	Silicon	45.8	SiO_2	97.985
2	20	Ca	Calcium	0.41	CaO	0.571
3	26	Fe	Iron	1.01	Fe_2O_3	1.445
4	8	O	Oxygen	52.78		
			Total	100		100.00

Table 7. Elemental composition and oxides of pebbles from ejecta, Lonar by XRF

S. No.	Elements	Mass (%)	Oxides	Oxide content	Mass (%)
1	Silicon	96.25	SiO_2	97.805	45.714
2	Sulphur	2.18	SO_3	1.581	0.791
3	Iron	1.1	Fe_2O_3	0.428	0.299
4	Calcium	0.39	CaO	0.154	0.11
5	Chromium	0.08	Cr_2O_3	0.032	0.022
			Oxygen		53.063
	Total	100		100	99.999

Fig. 1. White colored (Silica pebbles), before cleaning found at Lonar

Fig. 2: White colored (Silica pebbles) after cleaning found at Lonar

Fig. 3: Microscopic observation of silica pebbles powdered form

