Nature and composition of interbedded marine basaltic pumice in the ~52–50 Ma Vastan lignite sequence, western India: Implication for Early Eocene MORB volcanism offshore Arabian Sea

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The recognition of pyroclasts preserved in sedimentary environments far from its source is uncommon. We here describe occurrences of several centimetres-thick discontinuous basaltic pumice lenses occurring within the Early Eocene Vastan lignite mine sedimentary sequence, western India at two different levels – one at ~5 m and the other at 10 m above a biostratigraphically constrained 52 Ma old marker level post-dating the Deccan Volcanism. These sections have received global attention as they record mammalian and plant radiations. We infer the repetitive occurrence of pumice have been sourced from a ~52–50 Ma MORB related to sea-floor spreading in the western Arabian Sea, most plausibly along the Carlsberg Ridge. Pyroclasts have skeletal plagioclase with horsetail morphologies $\pm$ pyroxene $\pm$ Fe–Ti oxide euhedral crystals, and typically comprise of circular polymodal (radii ≤ 10 to ≥ 30 μm), non-coalescing microvesicles (>40 – 60%). The pumice have undergone considerable syngenetic alteration during oceanic transport and post-burial diagenesis, and are a composite mixture of Fe–Mn-rich clay and hydrated altered basaltic glass (palagonite). The Fe–Mn-rich clay is extremely low in SiO_2 , Al_2O_3 , TiO_2 , MgO , alkalis and REE, but very high in Fe_2O_3 , MnO , P, Ba, Sr contents, and palagonitization involved significant loss of SiO_2 , Al_2O_3 , MgO and variable gain in Fe_2O_3 , TiO_2 , Ni, V, Zr, Zn and REE. Bubble initiation to growth in the ascending basaltic magma (liquidus ~ 1200 – 1250°C) may have occurred in ~ 3 hr. Short-distance transport, non-connected vesicles, deposition in inner shelf to more confined lagoonal condition in the Early Eocene and quick burial helped preservation of the pumice in Vastan. Early Eocene Arabian Sea volcanism thus might have been an additional source to marginal sediments along the passive margin of western India.

1. Introduction

The record of millimetre to centimetre-sized small pyroclastic detritus, preserved in sedimentary environments far from its source is largely unrecognized

in the geologic record (e.g., Carey *et al.* 2001; Risso *et al.* 2002). Wadia (1944) was one of the first to identify abundant pumice fragments in coastal Sri Lanka, an island where no inland volcanic eruptions have occurred. Records of submarine volcanism

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supplying volcanic detritus to continental margins are increasingly recognized in recent years (e.g., Bryan *et al.* 2004; Brasier *et al.* 2011; Rotella *et al.* 2013; Jutzeler *et al.* 2014). Although pumice is susceptible to destruction and alteration, lighter vesicular pumice can be transported far from the submarine volcanic source in a short time and thus have potential for accretion and preservation in sedimentary depositional systems in passive continental margins elsewhere. For example, rafts of pumice formed during the 1883 Mt. Krakatoa eruption, Indonesia, travelled about 3000 km in less than one year and deposited along the Pacific coast (Brasier *et al.* 2011). The stranding of sea-rafted pumice along sea coasts and passive margins thus call for better recognition and identification in geologic history, because these are excellent accessible sedimentary inventories that may provide valuable clues about the nature of remote submerged volcanic eruptions. This is important because volcanic contributions to sedimentary records, though poorly constrained, are enormous; at a production rate of 6–8 km³/yr, at least $\approx 3.4\text{--}4.5 \times 10^9$ km³ of pyroclastic material may have been added to the surface since ~ 4.5 Ga (age of the Earth), an order of magnitude greater in volume than that of the present-day sedimentary rocks (Fisher and Schmincke 1984). They are also immensely valuable key horizon for regional stratigraphic correlation.

The Arabian Sea and the NW Indian Ocean have active ridges and fractures with permissive volcanism in extensional settings since the Cretaceous, such as the Ninety-East Ridge, Chagos-Laccadives Ridge, Carlsberg Ridge, and Laxmi Ridge. The Carlsberg Ridge in the Arabian Sea was spreading at a rate of 5.5 cm/yr (half-spreading) at 58–49 Ma (Dyment 1998). At present, pumice covers large areas of $\sim 60,000$ km² in the central Indian Ocean Basin (Iyre and Sudhakar 1993), mainly located near morphometric features like fracture zones and sea mounts (Kalangutkar *et al.* 2011). Its presence in the western Indian basins is therefore not surprising, but has a sparse record except for the euhedral bipyramidal quartz of volcanic origin in the lignite seams in Vastan area, Gujarat, India (Sahni *et al.* 2006). Interestingly, the sediment supply (including pumice) and deposition in Vastan at $\sim 52\text{--}50$ Ma significantly post-date Deccan volcanism (~ 65 Ma), though marginal basin sediments in this part are considered to be mainly sourced from the Deccan basalts (Courtilot *et al.* 1988; Prasad *et al.* 2013).

In this work, we identify and describe, for the first time, the occurrence of few centimetres-thick, discontinuous, pumiceous lenses within the Early Eocene Vastan sequences in the western India continental margin basins. We present the field characteristics of the pumiceous deposits in the Vastan

lignite mine and their likely position in the Vastan stratigraphy, petrography and textures of the pumice clasts including the vesicle characteristics, and their bulk geochemical compositions. Combining field, petrographic, and geochemical evidences, we then discuss the origin of pumices. We also evaluate the plausible reasons how these rafts of post-Deccan marine pyroclasts survived the vagaries of marine action and finally were deposited and preserved in the lagoonal condition near the sea shore. Finally, we infer that these pumices were sourced plausibly from the Arabian Sea volcanism in the NW Indian Ocean region $\sim 52\text{--}50$ Ma ago, most likely at the spreading Carlsberg Ridge.

2. Geology, stratigraphy and palaeo-environment

The Vastan open-cast lignite mine in Gujarat, western India is located on the eastern fringe of the Tertiary hydrocarbon-bearing intra-cratonic graben or rift, called Cambay Basin (figure 1). The 20- to 150-m thick Cambay Shale is well exposed in the Vastan lignite mine and comprises of carbonaceous grey to green shales, clay-bearing marly sediments, and lignite (Sahni *et al.* 2006; Garg *et al.* 2008) with two thicker lignite seams – one at the base (Lignite 2) and the other at top (Lignite 1), and a few thinner seams that occur interspersed within the sequence (figure 2a, b). Few centimetres-thick pumice layers occur as detached lenses within the shale ~ 15 cm above the upper boundary of Lignite 1 seam (figure 3); the boundary is marked by the presence of amber with bipyramidal quartz of volcanic origin (figure 4). The strike of the deposit is NE–SW with dips of about 4° towards NW.

The amber, present in the Vastan sequence, is extremely rich in both plant and animal fossils including mammalian fauna and insects. A number of workers have attempted to decipher the nature and style of sedimentation, construct facies-based models of depositional environment, and understand the biotic affinities and biostratigraphy of the Vastan sequence (Sahni *et al.* 2006; Prasad *et al.* 2013). Although, no radiometric age data is available for the rocks of the Vastan sequence, the presence of age-diagnostic dinoflagellate cysts in lignite suggest an Early Eocene ($\sim 55\text{--}52$ Ma) age for the sequence (Garg *et al.* 2008). On the basis of the presence of Nummulites (*N. burdigalensis*), the Vastan sedimentary section was assigned basal Cuisian age ($\sim 52.5\text{--}50.5$ Ma: Puneekar and Sarswati 2010). In the main Cambay Basin, a sedimentary succession (comprising Vagadkhol Formation and Olpad Formation) underlie the Cambay Shale, but unconformably overlie the Deccan Trap basalts.

The depositional milieu of the Vastan Mine area is marked by the absence of coarse clastics and

Figure 1. Location map of Vastan in western India and the tectonic elements in the surrounding Arabian Sea. The figure is taken and redrafted from Google Earth.

represent inner shelf to lagoonal setting (Bhandari *et al.* 2005; Sahni *et al.* 2006; McCann 2010). A recent high-resolution sequence-stratigraphy study suggests a marsh-dominated coastal plain opening into a shallow bay on its seaward side in Vastan, with rising sea along the western Indian margin in response to Early Eocene warming between 55 and 52 Ma; climatic transgression may have occurred at ca. 53.7 Ma (Prasad *et al.* 2013).

3. Field aspects of pumiceous deposit

The pumiceous rocks, first recognized and collected in the northern quarry face (21°25'47"N: 73°07'30"E) by two of us (HS and RSR), are about 6–10 cm thick, occurring within the shale as detached lenses (10–15 cm in length), ~15 cm above the upper boundary of the lignite 1 deposit (figure 3),

and are laterally traceable for more than 200 m in the quarry face. The lenses are massive, but are altered and friable, dusty grey to tan in colour, and have distinct spongy/vesicular appearance, often filled with rounded to subrounded pebbles of diverse origin (figures 5a, 6). The distinct lithological attributes help in recognizing these anomalous pumiceous sedimentary lenses within the shale and shale/lignite 1 interface, but surprisingly, far from any known volcanism in the vicinity. These lenses have different texture than the abundantly present terrigenous fine-grained bedded/laminated muddy detritus (shale to mudstones) sediments derived from the Deccan Trap basalt source.

Recently, on March 2015, about 2 km away from the section mentioned above, a new 3- to 4-cm thick discontinuous pumice lens laterally traceable for more than 100 m was identified (by HS) in the southern quarry face (21°24'46"N: 73°05'53"E)

Figure 2. **(a)** Section on the NW mine face resting on the Deccan Volcanics (DV) showing position of the pumice lenses (PL), main lignite seams (Lig 1 and 2), N, *Nummulites burdigalensis burdigalensis*, foram zone marker for the SBZ10, M, level with terrestrial mammals (modified after Sahni *et al.* 2006). **(b)** Section observed on the southern mine face showing position of the pumice lens (PL) and upper lignite seams (Lignite 1).

Figure 3. Dumpster trucks mark location of Lignite 1; 15 cm above pumice lens (PL) level (see also figure 2a).

Figure 4. Q, amber nodules with excellent bipyramidal quartz crystals.

within the same Cambay shale unit (figure 1). The pumice lens occurs at ~ 5 m above the upper boundary of a lignite bed in this section (figure 2b). On the basis of similar lithological assemblages and their lithological attributes within the same Cambay Shale, the lignite lens in southern quarry face is tentatively considered equivalent to Lignite 1

deposit encountered in the earlier section. The supply and deposition of pumiceous material in Vastan was thus not a localised phenomenon in Early Eocene, rather more extensive and/or may have even been repeated in the sequence. This new pumice lens again is dusty brown with/without whitish tint to tan in colour and spongy in appearance (figure 5b). However, unlike the earlier samples, it is crudely laminated in hand samples.

4. Materials and methods

Thin section petrography was crucial in characterizing the different volcanic/pyroclastic aspects of the Vastan samples. A large number of thin sections have been studied under the advanced research-model Leica Microscope (DM 750) with in-built automated softwares for undertaking different measurements and taking large number of photographs. Since the samples are extremely altered, special care was taken during mounting and polishing of the sections.

Given the typical altered nature of the Vastan pumice, the least altered portion from the inside (core) of several samples were obtained by drilling, along with altered samples from the near surface portion, for chemical analyses. Foreign detritus present in the pumice vesicles were carefully removed by gentle crushing using agate and mortar, followed by hand picking the foreign particles. All samples were then cleaned with deionized water and ultrasonicated for 30 min. The concentrations of major oxides and selected trace elements (Sr, Zr, Ba, V, Ni, Zn) for five pumice samples were determined by XRF method using 30 mm diameter pressed-powder pellets (~ 5 g sample powder of $< 70 \mu\text{m}$ size), at the Indian Institute of Technology, Kanpur. The analyses were done using a Rigaku ZSX PRIMUS II wave-length-dispersive XRF (WD-XRF), with 4KW Rh target, operated at 60 kV and 50 mA. Calibration curves were obtained from regressions of about 20 international rock standards from the USGS and Centre de Recherches Pétrographique et Géochimiques, CRPG-CNRS, Nancy (e.g., BHVO, BE-N, AGV, etc.). Based on the repetitive analyses of BE-N and AGV, the accuracy of the XRF data for major and trace elements is estimated to be $\leq 5\%$ and $\leq 10\%$, respectively. Additionally, we have also done ICP-MS analyses of one sample each from the core and from near the surface to determine rare earth elements (REE) abundances. Samples (pumice, BHVO-2, BIR-1a, AGV, and a blank) were digested using (ultrapure $\text{HF} + \text{HNO}_3 + \text{HClO}_4$) and subsequently analyzed using a Thermo Scientific iCAP Qs ICP-MS. Several dilutions of a multi-element standard solution

Figure 5. (a) Dark brown spongy pumice lens (PL) with pinch and swell margins on the NW mine face (see also figure 2a); (b) closer view of dark brown to brown pumice lens (PL) on the southern mine face (see also figure 2b).

Figure 6. Close view of the PL of figure 5(a) in field. Note the spherical to elliptical pebbles set in vesicles.

(Merck, Germany) containing 24 trace elements were used for calibration. The three USGS rock standards, BHVO-2, BIR-1a, and AGV-2, digested

with the rock samples using the same method reproduced the elemental concentrations in good agreement with the certified values.

Table 1. Major and selected trace element compositions of Vastan pumice; compositions of hydrothermal Fe-rich clay, MORB-palagonite and Carlsberg Ridge basalts for comparison.

Sample	Group 1			Group 2			SN1	SN2	Hydrothermal Fe-rich clay (Singer and Stoffers 1987)	Palagonite (9) (Pauly <i>et al.</i> 2011)	Palagonite (4) (North Arch (26D-6)) (Pauly <i>et al.</i> 2011)	Carlsberg Ridge basalt (avg) (Ray <i>et al.</i> 2013)	
	PM-C1	PM-C2	PM-C3	S1	S2a	S2b							
Major oxides (wt%)													
SiO ₂	5.46	5.73	4.05	14.63	22.17	23.58	28.81	4.86	8.97	21.78	27.8	26.4	51.01
TiO ₂	0.70	0.70	0.22	2.20	3.29	0.87	1.39	—	—	—	3.99	4.47	1.45
Al ₂ O ₃	1.39	1.47	1.27	6.58	10.92	14.50	17.90	0.72	1.46	1.78	7.55	4.93	15.60
Fe ₂ O ₃	71.16	71.13	74.54	62.12	48.93	31.63	39.10	82.22	83.23	58.17	16.74	21.37	9.94
MnO	3.64	3.64	3.86	2.85	2.08	0.76	0.76	0.18	—	0.16	0.19	0.03	0.18
MgO	—	—	0.48	0.04	0.22	0.94	1.24	0.51	1.41	4.96	2.22	0.83	8.00
CaO	1.89	1.90	1.46	1.54	1.51	20.38	0.90	0.11	1.09	0.07	6.87	0.71	11.57
Na ₂ O	0.27	0.28	0.00	0.34	0.41	1.71	1.51	0.03	0.04	0.05	0.72	2.24	2.47
K ₂ O	—	—	—	—	—	0.05	0.09	0.11	0.17	0.11	0.16	0.83	0.13
P ₂ O ₅	0.68	0.68	0.49	0.47	0.29	0.17	0.20	—	—	—	0.70	0.00	0.14
Total	85.19	85.52	86.37	90.76	89.82			88.74	96.37	87.08	66.94	61.81	
Trace elements (ppm)													
Ba	66	43	8	—	—	—	—	—	—	—	—	38.22	15.23
Ni	39.29	125	55	228	306	—	—	—	—	—	—	—	102.89
Sr	—	580	92	502	408	85	67	—	—	—	—	267.81	108.14
V	781	245	61	743	1145	1722	2265	—	—	—	—	54.56	279.81
Zr	134	18	16	76	127	—	—	—	—	—	—	264.95	100.06
Zn	75	75	86	433	513	655	762	—	—	—	—	100.63	109.19
Co	130	134	240	338	343	—	—	—	—	—	—	18.48	43.55
Cr	57	35	126	199	182	—	—	—	—	—	—	277.6	315.2
Cu	19	19	56	92	92	—	—	—	—	—	—	174	79.21
Sc	36	34	38	35	34	5	9	—	—	—	—	8.75	36.88

Table 2. Rare earth elements (REE) compositions of Vastan pumice; compositions of MORB-palagonite and Carlsberg Ridge basalts for comparison.

Sample trace elements (ppm)	PM-C1	S2a	Palagonite (4) (North Arch (26D-6)) (Pauly <i>et al.</i> 2011)	Carlsberg Ridge basalt (avg) (Ray <i>et al.</i> 2013)
Ce	19.29	125.02	7.18	10.50
La	8.32	46.43	24.89	3.47
Pr	2.90	15.10	7.74	1.39
Nd	13.32	66.18	32.19	10.28
Sm	2.98	18.57	7.91	3.29
Eu	0.97	5.33	2.28	1.18
Gd	3.45	25.76	8.72	4.86
Tb	0.50	3.86	1.42	0.90
Dy	2.10	21.75	9.62	5.22
Ho	0.42	4.11	2.05	1.13
Er	1.01	11.29	5.63	3.55
Tm	0.17	1.52	0.79	0.61
Y	10.67	85.83	–	34.03
Yb	0.97	9.34	5.2	3.28
Lu	0.13	1.47	0.68	0.48

Additionally, we have also analyzed one more recently identified pumice sample by XRF-method. Two analyses were performed: one on the least altered part (core) and another on the more altered portion near the surface. The XRF and ICP–MS data of Vastan pumice are shown in tables 1 and 2, respectively.

5. Petrography of pumice

Because of severe alteration and diagenesis of the pumice samples, establishing the original texture, mineralogy, and chemical composition have been extremely difficult. Nevertheless, some relict primary textures are still preserved in limited samples, which form the basis of this study. Textural characterization of pumiceous deposits has indeed been a powerful method in investigating pyroclastic deposits, both primary and non-primary types, and in constraining the magmatic processes that formed the pyroclasts, which otherwise are not directly observable nor easily recognizable from other studies of volcanic-sediment deposits (e.g., Fiske 1969; Polacci 2005).

We present below characteristic petrographic features observed in the pumice samples, which suggest submarine basaltic eruption origin. Subsequent to eruption, these pumiceous rocks were rafted in the ocean water and deposited in the continental margin basin(s) near Vastan along with other non-volcanic fragments. Circular vesicles (40–65% by volume) are common in these rocks, set in brown to dark brown matrix. There are no concentric rings observed around the nuclei of small crystals, as commonly expected in

oolites. Instead, we identify vesicles (>40–60%), characterized with vesicular texture, that represent fragments of magma with bubbles. The matrix constitutes about 20–30% of the samples. Occasional crystals (<7%) with/without sparse lithic fragments (<2%) are present in the matrix. A description of crystals, vesicles, and matrix present in the Vastan pyroclasts are presented below.

5.1 Crystals

Crystals, which constitute <7% by volume of the matrix present in the Vastan pyroclasts, are sparsely distributed. Well-developed plagioclase with lamellar twinning occurs in an intersecting arrangement (figure 7a). Pyroclasts with slender plagioclase micro-laths showing horsetail morphology, enclosed by dark brown devitrified glass, are also present (figure 7b). Inclusions of black circles of about 20 μm diameter trapped on the albite twin surfaces of the plagioclase micro-laths are interpreted as gas bubbles, which suggest faster growth together with decompression. The alignment of labradorite micro-laths along their long dimension shows characteristic flow texture (figure 7b). Euhedral broken pyroxene crystals with second order interference colour are intensely fractured (figure 7c). Sparse long prismatic euhedral crystal with high refractive index (R.I.) and distinct peaks of $\text{Fe}\pm\text{Mn}$ in EDX spectra, possibly magnetite, are also present (figures 7d, 8a), sometimes in criss-cross arrangement. These Fe-rich crystals are also fractured. In the same section, smaller euhedral broken opaque crystals show distinct peaks of Ti and Fe in EDX spectra, indicating possible presence of ilmenite (figure 8b). Dark brown glass

Figure 7. (a) Intersecting plagioclase with lamellar twinning. (b) Skeletal horsetail plagioclase micro-laths in devitrified glassy material show trachytic texture. (c) Larger pyroxene (px) formed during early crystallization of basaltic magma; vesicles (V) also present. (d) Long prismatic euhedral crystal of Fe-Mn oxide, possibly magnetite. (e) Vesicles in thin sections nearly spherical and homogeneously distributed, show polymodal distribution. Vesicles rarely coalesce. (f) Vesicles (v) coalescence to form dumbbell shape with complex margins. Dark glassy rims around circular margins of vesicles; quartzo-feldspathic granitoid fragment (L) at centre. (g) Fragmentation of vesicle walls forming shard structures. (h) Angular carbonate lithic fragment with well-developed rhombic cleavage (L) of local derivation; larger vesicles (radius $\sim 400\ \mu\text{m}$) seen in background. (i) SEM image: concentric onion-peel cooling and radial fractures, but without nucleus, in vesicles.

Figure 8. (a) Energy dispersive spectroscopy (EDS)-diagram showing distinct peak of Fe for long prismatic euhedral magnetite crystal. (b) Energy dispersive spectroscopy (EDS) diagram for smaller euhedral broken crystal of ilmenite showing distinct peaks of Ti and Fe.

rinds (cryptocrystalline under high magnification) coat the crystal surfaces. Pyroxene crystals probably were in the magma prior to eruption as pyrogenic minerals (see Fisher and Schmincke 1984). We infer that plagioclase and pyroxene may represent near-liquidus phases of the magma, which was of basaltic composition.

5.2 Vesicles

Thin sections of selected pumiceous lenses were studied using optical and scanning electron microscopy (SEM) at different magnifications to determine vesicle size, shape and spatial distribution, two-dimensional bulk vesicularities (area

fraction of vesicles: 40–60%), and vesicle number densities ($>5.5/\text{mm}^2$) (figure 7c, e–f).

A foam-like appearance consisting of evenly-spaced spherical vesicles is generally observed in the samples. The frequency distribution plot shows polymodal size distribution for vesicles with radii ranging from ≤ 10 to ≥ 30 μm ; $\sim 50\%$ of vesicles with radius ≥ 15 – 20 μm represent the dominant mode (figure 9). Rarely, but significantly we also encounter very large vesicles (radius ~ 400 μm : figure 7h), characteristic of basaltic melt degassing only (see later). Following the classification of Polacci *et al.* (2003), we characterize the vesicle population as microvesicular, which is the most common juvenile component of the pumice deposit.

Figure 9. Frequency distribution shows polymodal size distribution of vesicles in Vastan pumice with radii ranging from ≤ 10 to ≥ 30 μm ; $\sim 50\%$ of vesicles with radius ≥ 15 – 20 μm represent the dominant mode. Very large vesicles (radius ~ 400 μm) rarely present.

Brown to dark brown fine-grained cryptocrystalline material often lining individual vesicles represent solidified residual melt, which moved into the early formed vesicles before consolidation of the magma. The shape of vesicles is mostly circular in outline and homogeneously distributed (figure 7e, f). However, vesicles with an elliptical shape are also present occasionally. Elongation is defined here as $(\lambda_1 - \lambda_2)/(\lambda_1 + \lambda_2)$, where λ_1 and λ_2 are the semi-long and semi-short axes of the best-fit ellipse, respectively (after Rust and Monga 2002; Rust *et al.* 2003). A spherical vesicle and an elongated (sheared) vesicle with an aspect ratio 3:1 have elongation < 0.1 and 0.5 , respectively (Moitra *et al.* 2013). The elongated single vesicles in our thin sections have average elongation of 0.13 with a maximum value of 0.26 , which suggest that vesicles may represent bubbles that have not been significantly affected by shear deformation. The bubbles with maximum elongation (0.26) have circularity, i.e., ratio of cross-sectional to surface area of ~ 0.87 , it is 1 for a circle. Although, the vesicles in our samples are mostly circular and separated from each other, in few cases these came in contact with each other, or were in the process of coalescence, resulting in a dumbbell shape with complex margins (figure 7e, f). These vesicles, however, are not affected by vesicle elongation.

We compared regularity of the coalescing vesicles to those of hypothetical vesicles using the method of Shea *et al.* (2010) (also see in Moitra *et al.* 2013), as the shape of vesicles is sensitive to complex margins of convolutedly shaped vesicles developed out of coalescence. Typically, the regularity of

coalesced vesicles (measured as $1-\Omega$; Moitra *et al.* 2013) increases with increasing complexity of vesicle shapes; sphere has $\Omega \approx 1$. The regularity of coalesced vesicles ($1-\Omega$ range from 10^{-3} to 10^{-1}) in our samples is quite high, with very rare coalesced vesicles with extremely complex margins ($1-\Omega \leq 10^\circ$) (figure 7e, f). Larger vesicles have better regularity of margins.

Glass shards are pieces of vesicle walls formed during vesicle collapse and fragmentation (Heiken and Wohletz 1991). Curved or plate-like glassy shards are also observed in our samples (figure 7g). Vesicles show concentric and radial cooling fractures that are better observed in the SEM images (figure 7i). Brown to dark brown fine-grained cryptocrystalline material lining vesicles (not shown) may represent solidified residual melt, which moved into the early formed vesicles before consolidation of magma. Partial crystallization of vesicular lava followed by complete crystallization at a higher confining pressure (as happens in under water column in marine condition) could produce the observed features (cf. Smith 1967).

5.3 Lithic fragments

Lithic fragments are very sparse in the rock. Angular carbonate fragments (figure 7h) occur rarely, which suggests derivation from local sources in the Vastan basin. Presence of sub-angular quartzose to quartzofeldspathic fragment (figure 7f) represent weathered fragment of granitoid rocks.

5.4 Matrix

The matrix constitutes $\leq 30\%$ of the rock. Under petrological microscope, the matrix in the samples are monotonously nearly isotropic to tan-altered glass. The glass in matrix vary from semi-transparent light-yellow sideromelane and more-strongly-coloured yellow to brown palagonite, expected in altered basaltic glass. Smaller euhedral broken opaque crystals (Ti-bearing), observed in SEM-EDX, are common and dispersed in the matrix. The matrix is occasionally vesiculated with lumpy/botroidal appearance. Diagenetic carbonates in the matrix show mosaic textures. Variably dark-brown coloured palagonite, unlike its light-coloured variety, may have been formed at elevated magmatic temperatures (see in Stroncik and Schmincke 2002 and references therein).

6. Geochemistry of pumice

6.1 Major elements

Although the samples show variable compositions (table 1), compositional plots show a coherent

Figure 10. Major and trace element composition of Fe-Mn rich clay material (Group 1) and palagonite (Group 2) in the Vastan pumice plot along consistent trend suggesting their genetic connection. Element compositions are plotted mostly against TiO_2 which is considered relatively immobile during alteration. (a) Fe_2O_3 is negatively correlated with TiO_2 , but (b) Fe_2O_3 is positively correlated with MnO and (d) P_2O_5 in the samples. TiO_2 , on the other hand, show positive relations with (c) Al_2O_3 , (e) Ni , (f) V , (g) Zr and (h) Cu contents in the samples implying Al, Ni and V partition in the secondary Ti-bearing phase(s) during alteration of the basaltic glass.

and consistent pattern (figure 10). The major element compositions in the three pumice core samples (Group 1) show very high Fe (Fe_2O_3 71.1–74.5 wt%) and Mn ($\text{MnO} \geq 3.64$ wt%) contents, but extremely deficient SiO_2 (4.05–5.73 wt%), Al_2O_3 (1.27–1.47 wt%), MgO (<0.5 wt%), CaO (<1.5 wt%), and Na_2O (0.27 wt%) contents. The compositions of Group 1 samples are comparable to those of Red Sea clay of hydrothermal origin in a deep sea core (cf. Singer and Stoffers 1987: see table 1) known to precipitate at hydrothermal sites. The very low Al:Fe ratios (0.01) in our samples indicate Fe^{3+} substitution for Al^{3+} in the clay mineral lattice. On a Fe_2O_3 – Al_2O_3 – MgO plot (figure 11), the samples plot very close to the Fe-rich clay (nontronite) compositions.

On the other hand, four samples taken out at or near the surface of pumice, including the newly identified pumice samples (Group 2), show larger variations. Compared to the Group 1 samples, the Group 2 samples have higher SiO_2 (14.6–28.81 wt%), Na_2O (0.4–1.7 wt%), Al_2O_3 (6.58–17.9 wt%), MnO (0.76–2.8 wt%) and MgO (0.4–1.24 wt%), but comparatively lower Fe_2O_3 (31.63–62.1 wt%) (table 1). It can be seen in table 1 that these are closely similar to the palagonite compositions (samples 7010404 and North Arch 26D-6: Pauly *et al.* 2011)

formed during basaltic glass (sideromelane)–seawater interactions. The Group 2 samples also have high Fe/Mn-ratio (>20), though Fe and Mn contents are lower compared to the Group 1 samples. Also, Group 2 plot very close to the Fe-smectites formed during palagonitization of Mid Oceanic Ridge basalt (MORB) glass (figure 11).

The palagonite formation (Group 2) from basalt has resulted in a significant loss of SiO_2 , Al_2O_3 , MgO , CaO and Na_2O contents, but gain in Fe_2O_3 , MnO and TiO_2 (table 1). On the other hand, the Fe–Mn-rich clays (Group 1) show extreme deficiency in SiO_2 , Al_2O_3 , TiO_2 , MgO , alkalies, but high enrichment in Fe_2O_3 (>71 wt%) and MnO (>3 wt%) contents. Overall, major element variations in the Vastan pumice samples (figure 10) reveal that, while Fe, Mn and P are preferentially fractionated into the Fe–Mn-rich clays (Group 1), Al, Ti, and to some extent Si, show preference for palagonite (Group 2). Low total (see table 1) and pattern of element gain/loss for the samples suggest basaltic glass–aqueous interactions during alteration.

6.2 Trace elements

Selected trace element data of Vastan pumice are presented in table 1. The trace elements also show

Figure 11. Fe_2O_3 – Al_2O_3 – MgO plot (after Chamley 1997) showing presence of Fe-rich clay material (cross; Group 1) with extremely high Fe concentrations (~71 wt%), and palagonite (triangle and square) in Vastan pumice. Group 1 samples plot close to Fe-rich Nontronite field similar to Red Sea hydrothermal clays (plus: Singer and Stoffers 1987), but palagonite samples (Group 2) have lower Fe_2O_3 contents, compositionally similar to Fe-smectite. The average Carlsberg Ridge basalt (CRB: Ray *et al.* 2013) are also plotted for comparison.

consistent patterns and are comparable to seafloor palagonite compositions (North Arch 26D-6: Pauly *et al.* 2011). Vastan pumices have very high concentrations of Ni (up to 306 ppm) and V (up to 2265 ppm) and show positive correlations with TiO_2 concentrations (figure 10e, f), implying Ni and V partitioning into the secondary Ti-bearing phase(s) during alteration of the basaltic glass. Several Vastan samples (both groups) have Ti/V (~ 50) comparable to that of MORB (20–50: Shervais 1982).

The REE concentrations in Vastan samples (table 2) are also consistent, and their ratios (e.g., Sm/Nd: 0.16–0.33 *vs.* 0.25; La/Ce: 0.28–0.38 *vs.* 0.49 for Group 1 *vs.* Group 2) are comparable to those of same palagonite compositions mentioned above (see table 1). The chondrite normalized (McDonough and Sun 1995) REE patterns in the Vastan samples show only a $3\times$ enrichment of LREE over HREE, with very weak (in core) to absence of resolvable Eu-negative anomaly (in outer part) (figure 12). Although, the REE patterns of core and surface samples are essentially parallel, REE is more partitioned into the surface with concomitant rise in TiO_2 in the palagonite samples (Group 2). This is also true for Cu and Zr indicating systematic chemical fractionation of these elements occurring during palagonitization. The Fe–Ti oxyhydroxide coating has been reported to be an important phase containing the REE in sediments (Singh and Rajamani 2001) (figure 11).

The REE patterns of our samples are also similar to the ocean floor palagonite of Pauly *et al.* (2011), except for the negative Ce-anomaly observed in the latter (figure 12). This is likely to be controlled

by the redox conditions in the ocean water that result in the oxidation of Ce (III) to insoluble Ce (IV), which is subjected to preferential scavenging over REEs (III) by iron and/or manganese oxides (Sholkovitz and Schneider 1991). The Ce oxidation may also take place in the shallow water environment through bacterial mediation in the absence of dissolved Mn (II) (Moffet 1990; Pourret *et al.* 2008). This implies that Vastan pumice may have formed at a depth in the ocean where Ce (III) oxidation was inhibited. The REE thus behaved coherently without any Ce-negative anomaly in the samples.

On the other hand, Sr, Ba, and P show preference for Fe–Mn clays (Group 1). Therefore, the Vastan pumice seems to be a composite mixture of palagonite and Fe-rich clays (nantronite?) and/or Fe-rich marine encrustations.

7. Discussion

In this section, we address: (a) magma-type/possible compositions that may have erupted, (b) why the pumice rafts did not sink in the ocean water, rather floated and travelled a long distance from its source, and (c) plausible tectono-volcanic source of this pumice, which is the Early Eocene marine volcanism.

7.1 Volcanological interpretations

Constraining the nature and location of submarine eruption from pumice clasts and their vesicles is not straight forward (Binns 1972; Jutzeler *et al.* 2014) because natural pumiceous samples represent only

Figure 12. C1-chondrite normalized plots for REE in the Vastan pumice samples (both core and surface). The C1-normalized REE plots for MORB (palagonite) and average of Carlsberg Ridge basalts (CR basalt aver) are shown for comparison. C1-chondrite data taken from McDonough and Sun (1995).

the snapshot of final state of volatiles exsolution and do not provide information about early stages of degassing. Theoretical models, therefore, are not suitable to predict bubble-size distribution and their evolution in natural samples. Interestingly, however, we find Vastan pumice vesiculated at early stages of magmatic degassing, and thus bubble-size distribution and their evolution in the samples may bring out important constraints when compared with the high temperature (1000–1250°C) experimental results on bubble growth in basaltic, andesitic, and rhyodacitic magmas (Masotta *et al.* 2014).

Dark tan colour in hand samples, low crystallinity, presence of plagioclase (labradorite) and pyroxene as near-liquidus phases, presence of Fe–Ti phases, and abundant round-bubble vesicles of varying radii (≤ 10 to ≥ 30 μm) in the samples argue for basaltic magma volcanism. The directional alignment of plagioclase micro-laths (figure 7b) indicates flow in magma (flow texture), commonly encountered in basalts. Skeletal plagioclase as described above, are indeed found in basalts, especially those erupted and cooled in marine conditions (see Lofgren 1974). This type of plagioclase morphology generally develop at moderate to high undercooling of magma and concomitant supersaturation that limits formation of perfect crystals common in marine conditions (Higgings 1995).

Although basaltic magmas are normally less charged in volatiles, round-bubble vesicular structure similar to those in our samples are observed in basalt eruption of Kilauea volcano (see figure 1 in Masangan *et al.* 1993). During magma ascent, the decline of confining pressure permits dissolved volatiles to form bubbles due to a decrease in the pressure-dependent volatile solubility. In the process, bubbles may grow and later coalesce with each other to a limited extent. Rare presence of very large bubble (radius ~ 400 μm) with smaller bubbles within, as observed in our samples, is remarkably similar in appearance to that found in experiments of basaltic melt only (see figure 7h *vs.* figure 9 in Masotta *et al.* 2014). This further confirms a basaltic magma source for the Vastan pumice. However, at this stage we are unable to confirm whether these large vesicles were formed out of ostwald ripening, a process where smaller bubbles are redissolved to increase the volume similar to a large bubble at late stage of basalt magma vesiculation (Masotta *et al.* 2014).

Melt degassing in basalts dominates, and at a faster rate, in the early stages of vesiculation, till the bubbles are grown enough to interact with each other and coalesce later (Klug *et al.* 2002; Masotta *et al.* 2014). In the light of experimental results, presence of dominant microvesicles with very limited to rare coalescence in the Vastan

pumice suggest that (a) pyroclasts represent early magmatic stage and (b) faster ascent of magma. Using the experimentally constrained relations between bubble radius (μm) *vs.* time (s) for basaltic melt (see figure 8 of Masotta *et al.* 2014), we obtain a time span of about 10^4 s (≈ 3 hr) from bubble initiation to bubble growth in the Vastan samples. Weak elongation of vesicles also implies that there was not much time before fragmentation for vesicles to be elongated by flow. In our samples, uniform bubble growth might have started, as can be seen in figure 7(d and e), in magma of low viscosity that contained less number of phenocrysts, which is common in basalts.

Since our pumiceous samples contain mostly nearly spherical vesicles and none of the thin sections analysed in this study seem to have undergone considerable vesicle collapse or vesicle elongation due to shear, vesiculation must have occurred at shallow depths, perhaps near the mouth of the volcanic vent. A foam-like appearance consisting of evenly-spaced spherical bubbles (figure 7d) might have formed when the magma approached the magma-atmosphere interface, very similar to that described in a hypothetical case of bubble formation in pumice pyroclasts (see figure 1B in Heiken and Wohletz 1991). Thus, the fragmentation might have taken place at the magma–water interface, in a phreatomagmatic-type of eruptions. Fragmentation of thin-walled spherical vesicles, just before or at the time of eruption, of highly inflated pumiceous magma, gave rise to shard structures. We attribute vesicle collapse in this case to magma fragmentation that did not undergo bubble collapse during ascent. We concur with Rotella *et al.* (2013) that the generation and dispersal of marine vesicular basaltic pumice, thus, does not necessarily argue for explosive magmatic activity for bubble formation, though explosive basaltic eruption at mid-ocean ridges is possible (Helo *et al.* 2011).

Experimental simulation suggests that bubbles in rapidly vesiculating mafic magmas attain connectivity when vesicularities reaches up to 60–70% (Rust and Cashman 2011). The Vastan pumice have lesser vesicularities (~ 40 –50%) and thereby are practically free of connective bubbles, with only a handful of vesicles rarely either in contact, or coalesce. High-viscosity fluids (e.g., rhyolites, dacites, and andesites) are not suitable in this case either, as these fluids enhance bubble coalescence and elongation due to internal shear, which decreases the drift velocity of individual bubbles, i.e., decreasing rate of rise of individual bubbles in comparison to magma ascent rate (Cashman and Sparks 2013; Masotta *et al.* 2014). Also, silicic eruptions in Indian Ocean are unknown in Early Eocene.

7.2 Petrological constraints

Experiments on basaltic melt crystallization show most of the samples start to crystallize at 1200–1260°C with plagioclase and olivine, followed by augite (Ca-rich pyroxene), and finally magnetite/ilmenite (Thy and Lofgren 1994). At this stage, it is difficult to determine the crystallization sequence of magmatic phases in the fragmental pumiceous samples because primary magmatic textures are largely destroyed and not identifiable in these fragmental samples. However, in the light of basaltic magma crystallization history, as revealed in experiments, we infer that Fe–Ti crystals (ilmenite/magnetite) in our rocks may have crystallized in the range of 1176–1189°C, irrespective of whether magnetite crystallized earlier than ilmenite or vice-versa. The first order implication is that basaltic magma liquidus was higher than ~1200°C, most probably at 1200–1250°C. The occasional large vesicles (radius 400 µm), as encountered in our samples, is also remarkably similar to that observed in basaltic melt experiments conducted at 1230°C (see figure 4 in Masotta *et al.* 2014). We therefore conclude that the basaltic magma for Vastan pumice may have started to crystallize around ~1200–1250°C.

7.3 Geochemical constraints

The geochemical results are remarkably consistent with physical and volcanological interpretations. The mixture of Fe-rich clays/encrustations+palagonite in the bulk samples indicates basaltic glass (sideromelane)–seawater interactions (Singer and Stoffers 1987; Chamley 1997; Stroncik and Schmincke 2002; Pauly *et al.* 2011). The extremely Fe-rich clay samples with negligible MgO content (Group 1) may contain Fe-rich smectites (nontronite?) and hematite and/or goethite. The Fe-rich smectites are known to precipitate at hydrothermal sites on smoker chimneys fairly close to the vent areas (cf. Chamley 1997; Von Damm 1990). These clays were formed by hydration of basaltic glass, accompanied by oxidation of Fe and Mn and loss of SiO₂, Al₂O₃, MgO and Na₂O during basalt–water interactions. Samples PM-C1 and PM-C2, with very high Fe₂O₃ (Fe 44%) and MnO (Mn 5%) concentrations and Fe/Mn (>8), are also similar to marine Fe-crusts (table 1). Enrichment in Fe and V, depletion in Al, Mg, Zr, Ti and weak REE fractionation with unresolvable negative Eu-anomaly in the Vastan pumice (Group 1) are similar indeed to the geochemical characteristics of hydrothermal clays, as reported by Shao *et al.* (2015). The very high Fe-rich content in the Group 1 samples may thus represent Fe-rich smectites + partly Fe-rich crust, related to submarine basaltic volcanism.

Loss in SiO₂, Al₂O₃, and alkali, also took place during palagonite (Group 2) formation. Mg is extremely mobile during aqueous alteration (Zhang and Smith-Duque 2014 and references therein). Thus, low MgO content in the samples is consistent with aqueous alteration. The variable compositions of palagonite may be attributable to the compositionally-heterogeneous nature of the original basaltic glass, elemental mobility during alteration, nature of altering fluid (fluid composition and pH), and the rate and intensity of palagonitization (Stroncik and Schmincke 2002).

As mentioned above, alteration of basaltic glass to clays may partly be biologically mediated as well, which can also mobilize Fe and Mn (Banerjee and Iyer 2003; Zhang and Smith Duque 2014). Positive correlation observed between two major life-sustaining elements, i.e., Fe₂O₃ and P₂O₅ (figure 10d) is in qualitative agreement with this possibility. Phosphorous is also known to sorb strongly onto hydrothermal iron oxide precipitates in seawater (Jones *et al.* 2015). However, the preferential concentrations of REE in the outer part (surface) of pumice with higher TiO₂ content compared to its core (table 2) may indicate their association with secondary Ti-minerals.

Overall compositional similarities of the pumice samples (Group 2) encountered at different levels within the Vastan Shale signify repetitive supply and deposition of similar pumice detritus, expected in such a setting.

7.4 Plausible source in the western Arabian Sea

The tectonic framework of the western continental margin of India was established by the end of Cretaceous, having a direct link to the seafloor spreading and tectonomagmatic evolution of the Arabian Sea (Naini and Talwani 1982; Biswas 1987). The Arabian Sea region has several prominent features, such as several deep ocean basins by submarine plateaus, aseismic Laxmi and Chagos-Laccadive Ridges, seismically active Carlsberg and Sheba Ridges, the regionally extending Owen fracture zone, and the central Indian Ocean basin (figure 1). Pumice rafts in the central Indian Ocean may not represent our samples, because they are largely high silicic compositions (rhyolite to dacite) believed to have sourced from large rhyolitic calderas active in Sumatra/Indonesia (Kalangutkar *et al.* 2011; Pattan *et al.* 2013).

Two major ridges, the Laxmi and Chagos-Laccadive Ridges, divide the Arabian Sea region further into two broad crustal provinces, namely the western and eastern Arabian basins. The western Arabian basin is underlain by oceanic crust (Moho at ~11 km) with well detectable magnetic

anomalies consistent with extensional tectonic setting (Naini and Talwani 1982). On the other hand, the crustal structure in the eastern part is ambiguous, with no clearly detectable normal oceanic crust. Rather, the presence of continental crust with insignificant oceanic crust is more likely in the eastern part (Moho at 20–22 km: Naini and Talwani 1982; Bhattacharya *et al.* 1994). Because spreading oceanic centres are sites of MOPB volcanism, the tectonic framework in the western part of the Arabian Sea is obviously more conducive for permissive basaltic volcanism compared to its eastern counterpart.

Since Eocene, the Carlsberg Ridge has been characterized by slow spreading at 10–40 mm/yr, full-spreading rate (Iyre and Ray 2003; Merkouriev and Sotchevanova 2003). Thus, the spreading along the Carlsberg Ridge, a major tectonomagmatic feature in the western Arabian Sea, seems a more plausible source of submerged Early Eocene basaltic volcanism in the Arabian Sea (see also Banerjee and Iyer 2003).

We are not aware of any published chemical compositions of palagonitized basaltic glass from the Carlsberg Ridge system that can be readily compared with our samples. Further the major element compositions of average Carlsberg Ridge basalts (table 1; see also figure 9) are practically closely similar to global MORB. The compositions of altered MORB-derived palagonite (table 1; Pauly *et al.* 2011) are essentially comparable to our samples (Group 2) too, except for the Ce-negative anomaly. We therefore hypothesize that the origin of the Vastan pumice is related to the basaltic volcanism in the western Arabian Sea, plausibly in the Carlsberg Ridge. However, the lack of Ce-negative anomaly in the REE patterns (figure 12) in pumice may indicate that the submarine basaltic volcanism took place at a depth where Ce (III) oxidation was inhibited.

7.5 Role of rafting

Pumice with long and interconnected vesicles is more likely to be easily waterlogged, and hence can sink rapidly. On the other hand, rounded and non-connected vesicles keep the pumice afloat for a longer duration (Binns 1972; Fiske 1969), and can spread widely. This implies two points. First, the Vastan pumice are not sourced from the Deccan Basalt province, as continental weathering and reworking would destroy them completely beyond recognition, and produce fine muddy detritus. As mentioned earlier, the muddy detritus of Deccan basalt province indeed constitute a significant part of the Vastan Shale (Prasad *et al.* 2013). Second, it is conceivable that the pumice must have been sourced not far from the site of deposition,

which indicates the adjacent Arabian Sea region to be the most likely source. Palaeogeography maps (see Rose *et al.* 2014) indicate that the Carlsberg Ridge system was nearer to the western Indian continental margin (approximately ~500 km) at about 52–50 Ma; present distance >2000 km. We are of the view that (a) short-distance transport, (b) circular non-connected vesicles in pumice coupled with more confined inner shelf to lagoonal depositional condition and (c) quick burial were key to the remarkable preservation of the pumice in the area. It is possible that the pumice rafts may have been deposited at or near the same stratigraphic level elsewhere along the belt.

Marine pumice-rafts are immensely valuable as potential nutrient-supplier for mega- and micro-organism and also as floating substrate for bio-dispersal (Brasier *et al.* 2013). Given the close spatial and temporal association of pumiceous layers and fossiliferous horizons in Vastan, we speculate pumice formed at the Carlsberg Ridge region could have also been involved in transporting organisms during sea-rafting.

8. Conclusions

The main conclusions of this study are as follows.

- Vastan pumice have spherical polymodal micro-vesicles (>40–60%), matrix (20–30%), crystals (<5–7%), and lithic fragments (<2%). The vesicles have radius ranging from ≤ 10 to ≥ 30 μm with ≥ 15 –20 μm representing the dominant mode. Vesicles are non-coalescing, show weak elongation, and have vesicle number densities $>5.5/\text{mm}^2$.
- Bulk pumice represents a composite mixture of hydrothermal Fe-rich clay-like and palagonite material. Palagonite shows significant loss of SiO_2 , Al_2O_3 , MgO , but gain of TiO_2 , REE, Ni, V, Zr, Cu. The Fe-rich clays have very low SiO_2 , Al_2O_3 , TiO_2 , MgO , alkalies, but very-high to high Fe_2O_3 , MnO, P, Sr and Ba contents. Secondary Ti-minerals may have controlled distribution of trace elements including REE.
- Early Eocene (~52–50 Ma) pumiceous lenses in the Vastan Mine represent a contemporaneous marine MORB-type volcanism in the western Arabian Sea, most plausibly along the Carlsberg Ridge. The basaltic magma started to crystallize around ~1250–1200°C. Bubble initiation to growth in the fast ascending magma may have taken in ~3 hr. The Vastan pumice thus serves as a pointer to active spreading centres and transport directions during the opening of the Arabian Sea and resultant submarine mafic volcanism.
- Circular non-connected vesicles, coupled with inner shelf to more confined lagoonal depositional

condition and quick burial were key to remarkable preservation of Vastan pumice.

- Accretion of MORB-type marine pumice rafts may be important globally in influencing bulk compositions of continental margins and microbial dispersals.

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