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Shallow water soundscape off-Goa Grande Island

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ABSTRACT

Soundscape monitoring is an effective tool to characterize Marine habitats. The passive acoustic studies lead to an understanding of the biodiversity of the ecosystem. Analyses of the soundscape data acquired at 30m water depth are carried out along with the kind of anthropogenic sound. An algorithm to segment the recorded data is described. We identified that the recorded fish sound from Grande Island is similar to the species Condon nobilis (also known as a Barred grunt) of the Haemulidae family, which is commonly available in the western Atlantic. However, such species are not reported from the present area. Four species types of the Haemulidae family are reported from the Grande Island area using gillnet fishing. The available information and acquired sound data suggest that fish sound data belongs to the Haemulidae family though more investigation is needed to identify the species.

1. INTRODUCTION

Hydroacoustic is the science of sound waves in the water that has become an important tool for underwater remote sensing (Balk, 2001; Shabangu *et al.,* 2014). Hydroacoustic can be broadly classified as two disciplines: *(i)* active and *(ii)* passive acoustics. For an active acoustic system, acoustic pulses are transmitted into the water for producing backscatter echoes. By examining the received echoes, it is possible to estimate the range and in certain cases detecting the presence and bearing of an underwater target (Urick, 1983). Active acoustic systems are widely used for many oceanographic applications (Mann *et al.,* 2008). However, the transmission of sound levels in the ocean for a prolonged duration may cause longrange effects on aquatic animal health (Popper and Hawkins, 2012). Active acoustic activities (for *e.g.,* in marine protected areas) are now being subject to formal permission as emerged recently (Tyack *et al.,* 2015). Therefore, passive acoustic technique, a method for detecting and monitoring acoustic signals in an underwater environment is advancing as a vital tool for ocean soundscape studies.

The passive acoustic system transmits no signal, and it is designed to detect acoustic signals emanating from the original sources, including natural processes in the ocean, underwater noise sources of biological origin such as marine mammals (Southhall *et. al.,* 2007), crustaceans or fish (Tavolga, 1971) and anthropogenic noise sources (Ainslie, 2012). By analyzing passive acoustic recordings, it is possible to discriminate and identify different animal species and to calculate the relative number of animals present within the measurement range. These key pieces of information can be complemented by ocean productivity or yearly migratory passage of animals such as great whales. A new application of passive

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acoustics involves awareness of environmental issues, which has spurred the development of passive acoustic techniques (Nystuen *et al.,* 2004). Progress in the field of passive acoustics has attracted researchers to investigate physical and biological processes such as oceanic features, seafloor habitats, and associated processes (Dahl *et al.,* 2007). There is a growing consensus that anthropogenic sound levels in oceans are increasing that can have adverse effects on marine life (Tyack, 2008).

Hitherto, most of the passive acoustic experiments such as propagation modeling and related geoacoustic inversion studies have been carried out in deeper waters (Gervaise *et al.,* 2007). However, the focus is needed for shallow water studies such as physical and biological characterization of a littoral environment (Pace and Jensen, 2002), especially in the reef and off reef regions (Bertucci *et al.,* 2016). Understanding the underwater environment is possible through ambient sound field measurement and "soundscape" studies (Pijanowski *et. al.,* 2011). The term "soundscape" has been used in many disciplines to describe the relationship between the waterscape (or landscape) and the relative composition of sound present.

Most of the fishes and invertebrates use sound for vital life functions. Based on a review of 115 primary studies encompassing various human-produced underwater noise sources, 66 species of fish and 36 species of invertebrates reveal noise impacts on development, including body malformations, higher egg or immature mortality, developmental delays, delays in metamorphosing and settling, and slower growth rates (Weilgart, 2018). Anatomical impacts from noise involve massive internal injuries, cellular damage, hearing loss, and even mortality (Hastings and Popper, 1996; Hawkins and Poppers, 2017). Ecological functions of invertebrates such as water filtration, mixing sediment layers, and bio-irrigation, which are key to nutrient cycling on the seabed, were adversely affected by noise. Once the population biology and ecology are impacted, it will have succeeding consequences on fisheries and even food security for humans.

Studies on population dynamics and related ecosystem function of non-migratory fishes and invertebrates are relatively easy to accomplish as compared to migratory marine mammal species. Many fish species rely on vocal signaling during their activities and produce sounds using sonic muscles that vibrate the swimbladder or bony elements (stridulation) (Fine and Parmentier, 2015; Permentier *et al.,* 2016). Fishes use sound to attract mates and defend their territory (Vasconcelos *et al.,* 2010). In shallow water, the ambient sound field generally consists of various types of sound sources such as fish sounds (biophonies), wind and flow sounds (geophony), and boat sounds (anthrophony) (McWilliam and Hawkins, 2013). The spatial structure of the sound field is dependent on the nature of the waveguide comprising the multipath sound propagation between the sea surface and the seabed (Jensen *et al.,* 2011). Therefore, the characteristics of any signal received at the recording location can be affected by the variability of environmental parameters (*i.e.,* sound speed and absorption) in the medium. If these propagation features are characterized, it is possible to use the recorded soundscape and fish sound as an acoustic metric for studying ecosystem function (Rountree *et al.,* 2006).

In this context, the analysis presented here expounds passive acoustic (fish sound) data recorded using an autonomous wideband hydrophone system with an intention to understand shallow-water biodiversity of the study area (Au and Lammers, 2016). In general, the temporal and spectral characteristics of passive acoustic recordings such as "oscillogram", "spectrogram", and peak sound level of the "power spectral density" (PSD) are used for fish sound identification (Fish and Mowbray, 1970; Erbe *et al.,* 2015; McCauley and Cato, 2000; Mahanty *et al.,* 2015). The power spectrum encompasses several dominant frequencies, which presumably represent major oscillation modes in the fish sound, but the amplitudes of these modes vary in a complex manner (Wilden *et al.,* 1998; Chakraborty *et al.,* 2014; Chanda *et al.,* 2020a; Chanda *et al.,* 2020b). Here, we present soundscape of the Grande Island off Goa location having water depth of 30m.

2. METHODOLOGY

2.1 Materials and methods

Here, investigations making use of passive acoustic data is carried out. For this purpose, the Song

Meter acoustic system for a marine application is extensively used for fish sound data acquisition. Present work illustrates how the passive acoustic data were acquired employing Song Meter (SM2M+). Acquisition of ancillary data such as wind, current, water temperature, sound velocity profiler data as well as surface sediment data is also a component of data acquisition. Here, the technical aspect of the Song Meter system is discussed.

2.2 Song meter

The Song Meter acoustic system (https://www.wildlifeacoustics.com) is a cost-effective, weatherproof marine recorder that can be used for underwater acoustic monitoring of fish. It has also been effectively used during long-term bioacoustics monitoring of dolphins, whales and other marine life including fish as well as and anthropogenic noise in an underwater environment. Song Meter systems, SM2M+ is used for present data acquisition activities. This recorder (SM2M+), is submersibles having a 16-bit analog to digital converter designed for short- or long-term deployment in fresh or saltwater. The unit is designed to allow quick refurbishment of the device along shipside for immediate redeployment. The batteries and SD flashcards can be easily swapped and the housing resealed for redeployment. The device can be anchored and recovered via tether, diver or by optional acoustic release. These systems are self-buoyant submersible that uses a thick-walled PVC housing rated for deployment up to a depth of 150 m. The core electronic motherboard accommodates 32 D cell batteries which are installed on both sides of the board. Dimension wise, the system (SM2M+) is identically cylindrical shaped with a height 79.4 cm and 16.5 cm diameter, they can be fitted with a hydrophone with a length of 2.5 cm and 1.9 cm diameter. The systems weigh around 9.5 kg in the air without batteries, and the buoyancy in saltwater is 5.5 kg.

The SM2M+ system consists of a single hydrophone having a frequency bandwidth of 2 Hz - 48 kHz. The system records in audio (WAV) format files for predefined sampling interval. The sensitivity of the hydrophone is calibrated to 0.1 dB resolution. The SM2M+ submersible is powered through 32 D cells alkaline batteries. The recorder can accept 1.5V alkaline batteries, 1.2V NiMH batteries or 3V-3.3V lithium batteries. A board contains protection diodes that must be configured for the appropriate cell voltage. The SM2M+ is normally configured for 1.5 or 1.2 V cells. In this configuration the batteries are wired in parallel groups of 4 in series. Two AA batteries run the SM2M+ clock. The system has the battery life and memory capacity to record for hundreds of hours. The Song Meter systems were calibrated at ESSO-National Institute of Ocean Technology (NIOT) calibration facility (http://www.niot.res.in/ATF/). Operational deployment of SM2M+ and SM3M including schematic diagram of the mooring system is displayed (Fig. 1) along with the deployment photographs of the Song Meter (SM2M+).

2.3 Study area

In this work, passive acoustic data were acquired from new regions of the Grande Island from WCI (Western Continental Shelf of India). The area is located at relatively deeper than the previous study areas (Chanda *et al.,* 2020a; Chanda *et al.,* 2020b; Chanda *et al.,* 2020c). Passive acoustic data along with the ancillary data were acquired from Grande Island, acquisitions of fish sound data at spot locations. The data recorded (8-12 May 2015) from the deeper part at 30m water depth (Location 4: 15°18.544' N 73°41.667' E) are used for fish sound analyses. Fish identification studies utilizing fish sound analyses are presented here.

2.4 Spectral methods

In this section spectral techniques such as spectrogram for visualization of segmented fish sound data is made. Similarly, power spectral density is applied to estimate the frequency peak of the fish calls and for the identification of fish calls. Here, these two techniques are extensively used.

Spectrogram : It provides the time localized frequency information for situations in which frequency components of a signal vary over time. The spectrogram is a visualization of time series to understand the frequency pattern of the recorded signal. The spectrogram is a linear time-frequency representation of the pre-windowing of the fish sound signal, and calculating its Fourier transform. This transform is

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of SM2M+ and SM3M: (a) schematic diagram of the mooring system is
photographs of the Song Meter (SM2M+ and SM3M). (b-f). The Song
the shore to finalize the data acquisition timings. The equipment is
menter where acoustic **Fig. 1.** Operational deployment of SM2M+ and SM3M: **(a)** schematic diagram of the mooring system is given along with the deployment photographs of the Song Meter (SM2M+ and SM3M). **(b-f).** The Song Meter system is programmed at the shore to finalize the data acquisition timings. The equipment is synchronized with the current meter where acoustic Doppler technique is used. This is necessary to avoid recording acoustic signal emanating from ADCP based current meter. U shaped moorings having positively buoyant Song Meter submersible (SM2M+) tied to a 40 kg dead weight, which is lying on the seafloor, is employed here. The same deadweight is tied to another deadweight which is lying on the seafloor by a twenty-meter long rope. 2-3 glass floats where each float weighs around 20 kg to another mooring where beacon lights are attached to the floats to maintain the lights above the surface are used. For the Song Meter system, beacon light is important from the safety and navigational aspects.

known as a Short Time-Fourier Transform and referred to as STFT *(t, f)* where t is the time variable and f the frequency. A quadratic form related to the Short Time Fourier Transform can be obtained by taking the square of this transform. The spectrograms provide the spectral energy density of the signal in the time-frequency domain. The spectrogram of a signal *x(t)* is referred to as SPECT *(t, f)* (Padovese *et al.,* 2009)

$$
SPECT(t, f) = \left| \int x(\tau) h^*(\tau - t) e^{-2j\pi f \tau} d\tau \right|^2 \tag{1}
$$

where $h(t)$ is a sliding window weight, and the superscript $*$ denotes conjugate. Matlab (www. mathwork.com) was employed.

Power Spectral Density (PSD) : Power spectral density is the measure of signal power content versus frequency. The power spectral density is typically used to characterize the peak frequency of the signal. The power spectrum is defined as the square of the amplitude of the Fourier transform of a time series and can thus be regarded as an expression of the variance of the underlying process. Power spectral density function (PSD) shows the strength of the variations (energy) as a function of frequency. In other words, it

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shows at which frequencies variations are strong and at which frequencies variations are weak. The unit of PSD is energy per frequency (width) and energy can be obtained within a specific frequency range by integrating PSD within that frequency range. Computation of PSD is done directly by the method called FFT or computing autocorrelation function and then transforming it.

2.5 Spectral analyses

Spectral analyses of 1 to 2 minutes of data acquired at the 15-minute interval were used to calculate power spectral density (PSD). Concatenated PSDs were generated on the time-frequency axis for location soundscape. Such soundscape plots indicating the duration of data acquisition, and intensity depicted by the color bar are presented. Thereafter, the recorded sounds were analyzed using an algorithm developed for the purpose (Fig. 2), waveforms, spectrograms and power spectral density (PSD) applied to the segmented signals of individual fish calls (Fig. 3). Spectrograms visualize the time-frequency content of the signal, and are commonly used to analyze animal vocalizations. More details of animal vocalization have been covered in this chapter. PSD has been used to estimate peak frequency. For estimation of the PSD and spectrogram, Matlab (www. mathwork.com) related functions were used. Furthermore, temporal call parameters such as call duration, the number of pulses and inter-cell separation of the fish calls were made use of to identify the fish sound. In order to corroborate the fish sounds, available data of fish sound files accessible on the websites *(i)* Discovery of Sound in the Sea (https://dosits.org/galleries/audiogallery/#fish), *(ii)* The Fish Base project (http://www.fishbase.org/) and *(iii)* Mc Cauley library (https:/ /m.soundcloud.com/abc-science/sounds-of-science-teraponsspawning) were utilized. Besides, the seminal work on biological underwater sounds for fish sound identification (Fish and Mowbray, 1970) was extensively made use of.

3. RESULTS AND DISCUSSION

For identification of the fish species, calculations for estimation of the spectrograms and PSDs of an individual call are made. For spectrogram, the 'pwelch' function is used. For species identification, temporal and spectral parameters of the fish calls are determined (Fish and Mowbray, 1970). The inter-call interval (from the end of one call to start of next call), single call duration and the number of pulses per bout were estimated. The spectral parameters such as PSD of single call are also calculated using 'pwelch' function, and the frequency peaks are estimated.

 The procedure to determine fish sound parameters requires noise reduction, segmentation and classification (Chanda *et al.,* 2020a). Therefore, to understand the nature of the biological sound present, the spectrogram (b in Fig. 2), of the raw 60 sec data is generated. A large spurt of signals is observed close to the lower frequencies. The PSD of the entire data has been depicted [Fig. 2 (c)]. Thereafter, an application of the noise reduction method is employed (Zimmer, 2011). The extraction of noise-free fish sound is imperative before data processing. In order to identify the start and stop point of the individual fish call, an enhancement of the contrast between signal and background noise in the data stream is required. Therefore, noise level reduction is carried out by multiplying the time series with ratios of its corresponding absolute to the maximum absolute value (d in Fig. 2) (Haris *et al.,* 2014). In the next step, Hilbert transform based envelope detection procedure is applied. Data segmentation is implemented in two steps. At first the coarse segmenting each call is visually completed. Next, a cumulative sum is employed across the individual data to determine the exact start and stop time. A sharp rise in the slope of the cumulative sum marks the beginning of a sound event. The flat or saturated part of the curve corresponds to the end of a sound event (e $\&$ f in Fig. 2). After segmenting the signals, to identify the fish family/species, the spectrogram of each call signal is generated (g in Fig. 2). Each fish family/species produces a sound that will have a peak frequency which is distinct, and can be used to distinguish the species. To determine the peak frequency, the PSD plots of all the call signals, (h in Fig. 2), are compared with the peak frequencies of the segmented call signal and those given in the referenced works. Thereafter, the determination of temporal parameters such as call duration, number of pulses within the calls, and inter-call durations are made. Detailed temporal and spectral parameters are shown (Fig. 3):

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Fig. 2. Fish sound segmentation technique flow chart employed in this work (adopted from Chanda *et al.,* 2020a).

3.1 Soundscape

The term 'soundscape' has been used by a variety of disciplines to describe the relationship between the landscape (or waterscape) and the relative composition of all sounds present therein (Pijanowski *et al.,* 2011; McWilliam and Hawkins, 2013). In shallow water, the ambient sound field generally consists of various types of sound sources such as biophony, anthrophony as well as the geophony. Here, the analysis of fish sounds (biophonies) and abiotic sounds such as wind and flow sound (geophony) and sounds of the boat (anthrophony) are carried out. The spatial structure of the sound field is dependent on the nature of the waveguide which forms due to the multipath propagation between the sea surface and seabed (Jensen *et al.,* 2011). Therefore, the characteristics of any signal received at a recorder's location can be affected by the variability in environmental parameters. While these propagation features are acknowledged, this research aims to quantify the soundscape and fish acoustic signals as received at the recorders to serve as a representation of what others may receive from their environment in their given locations. The study locations are well-known for tidal-stream influence such as seawater inflow, freshwater runoff, and salinity variations mainly during the southwest monsoon seasons (Manikandan *et al.,* 2016; Sreekanth *et al.,* 2015). Moreover, the variability in the soundscape arises from the bathymetric Shallow water soundscape off- Goa Grande Island

Fig. 3. Temporal and spatial parameters of the fish sound data.

relief, an active shipping channel, frequent small boat transits and biological sounds. The study objectives will include identification of fish species using their vocalizations, and characterization of recorded biophony to understand their relationships using passive acoustic data from ecologically important shallow water regions off Goa. from West Coast of India (WCI).

3.2 Fish sound characteristics

The present location is situated towards the deeper end at 30 m water depth, away from the coral reef system near Grande Island. SM2M+ - a passive acoustic data acquisition system was moored midway in the water column. The passive acoustic data acquisition was carried out from 07-12 May 2015 from the present location. The analyses of the concatenated PSD data reveal three types of sounds. Fish sound (indicated as 2 in Fig. 4a), the anthropogenic sound (boat sound indicated as 1), and another kind of sound probably from metal chains used by boats to anchor during the period starting from 07:00 h (on 11 May 2015) to 03:00 h (of 12 May 2015) (indicated as 3). The wind data reveal high-speed winds due to the premonsoon session, especially during the period 11-12 May 2015, having an average value of 4.20 m/s [0.5 (min.) to 9.25 (max.) m/s]. The analysis of time series data indicates a daily fish chorus from the early hours of dawn (02:30-06:00 h) (Fig. 4a). The grunting fish sound signatures are symptomatic that are also observed in the data. The waveform, spectrogram, and peak PSD of a single fish call have been depicted (Fig. 4b-d). The PSD of the sound data substantiates the presence of the fish sound (Location 4) near Grande Island. The peak level of the PSD for fish chorus sounds is found to be within the $(95.0\pm 2.6$ dB re1µPa²/ Hz) (Fig. 4d), and the peak frequency of the PSDs of the fish chorus sounds is noted (289.06±74.93 Hz). We determined temporal parameters of each segmented calls, and average value of the 1726 calls are found to be (call duration: $0.85.0\pm0.12$ sec), (no. of pulses/call: $10.20.0\pm1.82$ sec), and (inter call duration: $2.36\pm$ 1.08 sec). The fish sound signals were identified as the grunting sound of the Haemulidae family using the waveforms (Fish and Mowbray, 1970). The temporal call parameters such as call duration, the number of pulses per call, and inter-call duration determined from waveforms along with the spectral peak of

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Fig. 4. Concatenated power spectral density (PSD) in dB re 1µPa²/Hz concerning the time in hr at an interval of 15 min, b) waveform (1 µ Pa) versus time (s), c) spectrogram and *c*) Power spectral density in (dB re $1\mu Pa^2/Hz$) for Haemulidae family of Grunter fish which is similar to Barred Grunt *(Conodon Nobilis)* (given arrows are discussed in the text).

the PSD are also estimated. The single call parameters, including peak frequency, corroborate with the species *Conodon nobilis* (Pombo *et al.,* 2014; Fish and Mowbray, 1970) having a family name: Haemulidae. This fish uses stridulation mechanisms to produce sound (pharyngeal teeth), which is then amplified using a swim bladder (http://www.fishbase.org/). These sounds are associated with feeding. Both males and females produce sounds when the fish is distressed.

 Interestingly four fish species of Haemulidae families are reported within the Zuari estuary (close to Grande Island). They are: *Plectorhincus Chubi, Plectorhincus gibbosus, Pomadasys guoraca,* and *Pomadasys furcatus* (Sreekanth *et al.,* 2018). No records of their sounds are available to date. Here we have recorded the sound, which is very much similar to the *Condon nobilis* (also known as a Barred grunt) of Haemulidae families from Caraguatatuba Bight from southeastern Brazil (Pombo *et al.,* 2014) in the Western Atlantic Ocean (fishbase.org). Fish and Mowbray (1970) have provided vocalization details for grunter sound

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acquired from many parts of the world including *condon nobilis* species of Haemulidae family. Unfortunately, such records are unavailable for the Arabian Sea and adjacent coasts. Therefore, we can surmise that sound data characterized and presented from Grande Island belongs to Haemulidae family, however, confirmation of species out of four species needs more research.

4. CONCLUSION

In this work, we highlighted the importance of a passive acoustic survey to record fish vocalization and related identification. Analyses of the soundscape records acquired at 30m water depth are carried out along with the anthropogenic sound. An algorithm to segment the recorded data is described. We identified that the recorded fish sound from Grande Island is similar to the *Condon nobilis* (also known as a Barred grunt) of the Haemulidae family (Fish and Mowbray, 1970). However, such sounds are not stated from this area, though four species of the Haemulidae family is reported using gillnet fishing.

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Density ratio for inner and outer regions of a Mridangam head

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ABSTRACT

Some of the drums in India are made musical by changing a homogeneous membrane to a heterogeneous one. In this paper we obtain the best density ratio required for the outer and inner membranes to produce harmonics in a Mridangam.

1. INTRODUCTION

Ancient people experienced rhythm from their heartbeat, the sound of rain falling on leaves and the flow of water in the river. The pits on the ground covered with a bark could have been the oldest form of drums. One of the oldest drums mentioned in Vedas was the Bhumi dhundhubhi which consists of a pit on the ground covered with an ox skin¹. The instruments that consists of a pot covered with animal skin, such as Bheri, are common in ancient times². As the time passed, the drums were constructed with hollow barrels of wood covered with animal skin on both sides3. Dhol and Mridanga are the most important among them and their variants are common in different parts of India. Dhol type drums are largely used in festivals and ceremonies. Pakhawaj, Khol in North India, Maddalam in Kerala are drums identical in many aspects with Mridangam. Big drums were difficult to be carried over for long distances and hence small hour glass type drums were created, such as Udukku, Damaru and many other varieties can seen in different places of India. The tuning of drums was not easy, since moisture, temperature etc. affect the pitch but the introduction of multiple layers of animal skin helped to tune the drums to the desired pitch. The drums were not only used for music production but were also used for communication, rituals, and wars⁴. In Natya sastra, drums were classified according to their position used to play, shape and structure. B.C. Deva gives a diverse and systematic classification of drums⁵. Accordingly there are three categories of drums namely struck, rub and pluck types. A representation of the classification is given in Fig. 1.

Plucked drums are not practically seen. Indian drums like Mridangam and Maddalam come under the vessel type with bi-facial and cylindrical features. Tabla is a twin drum under mono facial type. All these drums have a loaded drum head. The stretching because of the loading, helped them to produce musical tones. It is because of this special character we classify them separately under pitched drums. In this paper, we study the characteristics of the pitched drum Mridangam in detail.

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Density ratio for inner and outer regions of a Mridangam head

Fig. 1. Classification of Indian drums

2. THE CONSTRUCTION OF THE DRUM HEAD

One or more layers of some animal skin is commonly used to make ordinary drums with a drum

head of particular thickness and they produce sound without discriminate pitch. In India, traditionally many homogeneous drum heads are converted to non homogeneous drum heads by fixing a fine paste on the drum head that make their pitch definite. Mridangam, Tabla and Maddalam are some among the drums that possess this special non homogeneous membrane. In Mridangam, there are two heads-left and right. The left head does not have a permanent paste added to the head and it does not produce pitched tones. The right head produces pitched tones. To make the right drum head, 3 layers of goat skin are sandwitched between layers of cow skin. A circular portion at the centre of the topmost layer is removed and a mixture made with Puranakeedam stone powder, rice and glue is pasted several times. The process takes about 2-3 hour. A pasted head of Mridangam is shown in Fig. 2.

Fig. 2. A Mridangam head

3. MODES OF MRIDANGAM

Mridangam produces five pitched sounds. To tune the drum, the stroke Num is used. The frequencies present in Nam stroke of Mridangam tuned to *d* sharp pitch in third octave are shown in Fig. 3. The obtained frequencies and their ratios are given in Table 1.

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Fig. 3. Frequencies in Mridangam

| | . . $\tilde{}$ | | | |
|----------|------------------------------|-------|--|--|
| Peak | Frequency (Hz) | Ratio | | |
| | 168.935 | 1.09 | | |
| γ | 309.148 | 2.00 | | |
| 3 | 459.03 | 2.96 | | |
| 4 | 599.242 | 3.87 | | |
| | 770.881 | 4.98 | | |

Table 1. Frequency ratios of the stroke of Mridangam.

4. MODES OF MRIDANGAM

¹Outside the circular region, along its boundary, there exists an annular region made of animal skin. Hence a Mridangam drum head can be divided into an inner region and an outer region that makes the drum head heterogeneous. Raman⁶, Ghosh⁷ and Rao⁸ studied the vibration of the heterogeneous membrane by choosing power-law variation for mass density. But the modes and their ratio experimentally obtained doesn't match with the power-law variations in density proposed by Raman, Ghosh and Rao. We will give a brief description of the three earlier works in the following subsections. For a circular membrane, the wave equation is given by $\frac{\partial^2 \psi}{\partial r^2} + \frac{1}{r} \frac{\partial^2 \psi}{\partial \theta^2} = \frac{T \partial^2 \psi}{\partial t^2}$ 100.253

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4.1 Studies by ghosh

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$$
(1)

¹ Some parts of the studies in this were presented at the 3rd National Conference on Frontiers in Modern Physics (NCFMP2021) held at Adamas University, Kolkata on 26-27 November 2021, *Emerging Trends in Physical Sciences (ETPS-2021)* held at ICFAI University, Tripura on 27 September - 01 October 2021 and *26th International Symposium Frontiers of Research in speech and Music (FRSM-2021)* on 11-12 February 2022 held at IIIT, Pune, Maharashtra.

Density ratio for inner and outer regions of a Mridangam head

Let Ψ = *coskt* $z(r)$ *cosn* θ and $c = \sqrt{\frac{T}{r}}$. Here Ψ is the transverse displac $\frac{1}{\rho o}$. Here *Ψ* is the transverse displacement, *k* is the wave number, *c* is the velocity of sound through membrane, *t* is the time, *z(r)* is the radial component of transverse displacement, n is an integer and θ is the angular component of transverse displacement. Let the mass density be $\rho = \frac{\rho_o}{r}$ and by using $s = 2k\sqrt{r}$ Eq. (1) changes to Density ratio for inner and outer regions of a Mridangam head
 $\cos k t \, z(r) \cos n\theta$ and $c = \sqrt{\frac{T}{p}}$. Here *Ψ* is the transverse displacement, *k* is the wave number,

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Let $\Psi = \cosh t z(r) \cos \theta$ and $c = \sqrt{\frac{r}{p}}$. Here Ψ is the transverse displacement, *k* is the wave number, *c*
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cyllar component of transverse displacement. Let *z* and outer regions of a Mridangam head
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ular component of transverse displacement. Let the mass

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(2)
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$$
\frac{\partial^2 z}{\partial s^2} + \frac{1}{s} \frac{\partial z}{\partial s} + \left(1 - \frac{4n^2}{s^2}\right)z = 0
$$
\n(2)

$$
z = AJ_{2n}(s) + BY_{2n}(s)
$$

At the centre the second part of the above equation does not give a finite solution and hence

$$
z = AJ_{2n}(s)
$$

$$
z = AJ_{2n}(2k\sqrt{r})
$$

At the boundary $r = a$ and the vibration is zero. Hence the solution is

$$
J_{2n}(2k\sqrt{r}) = 0\tag{3}
$$

From the roots of Eq. (3), the frequency ratios are found. The obtained values are shown in the Table 2. The roots does not form harmonics.

4.2 Raman's studies

Raman⁹ and Rao⁸ also studied density variation in a circular membrane. A power-law variation in the mass density as a function of radius was used by them. The mass density decreases from the centre to the outer edge of the black loaded region. Raman substituted the complete solution of the form ation and its solution is
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4.2 Raman's studies
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Raman⁹ and Rao⁸ also studied density variation in a circular membrane. A power-law variation

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r = R(r) \cos n\theta \cos(\, pt - \varepsilon)
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$$
\frac{d^2R(r)}{dr^2} + \frac{1}{r}\frac{dR(r)}{dr} + \left[\frac{p^2\rho}{T} - \frac{n^2}{r^2}\right]R(r) = 10\tag{4}
$$

0 The solution of Eq. (5) is *m m*

as was used by them. The mass density decreases from the centre
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se in the wave equation given in Eq. (1) and obtained

$$
\frac{d}{dt} + \frac{1}{r} \frac{dR(r)}{dr} + \left[\frac{p^2 \rho}{T} - \frac{n^2}{r^2} \right] R(r) = 10
$$
(4)
of form $\rho = r^{2m-2}$ and using $\frac{p^2}{T} = \lambda^2$ Eq. (4) was modified as

$$
\frac{d}{dt} \left(\frac{dr}{dt} + \frac{1}{r} \left[\lambda^2 r^{2m-2} - \frac{n^2}{r^2} \right] R(r) = 0
$$
(5)

$$
R(r) = AJ_{\frac{n}{m}} \left(\frac{\lambda}{mr} r^m \right) + BY_{\frac{n}{m}} \left(\frac{\lambda}{mr} r^m \right)
$$
(6)
ons Eq. (6) changes to

$$
J_{\frac{n}{m}} \left(\frac{\lambda}{mr} r^m \right) = 0
$$
(7)
the boundary as

$$
J_n(ka) = 0
$$
(8)
r membrane and its roots does not follow the integer ratio.
the boundary as

$$
J_{2n}(2\lambda\sqrt{a}) = 0
$$
(9)
various modes are same as for Eq. (3) given in Table 2. The roots

On applying the boundary conditions Eq. (6) changes to

$$
J_{\frac{n}{m}}\left(\frac{\lambda}{m}r^{m}\right) = 0\tag{7}
$$

When $m = 1$, Eq. (7) is modified at the boundary as

$$
C_n(ka) = 0 \tag{8}
$$

This is the equation for an ordinary membrane and its roots does not follow the integer ratio.

When $m = \frac{1}{2}$, Eq. (7) is modified at the boundary as

$$
J_{2n}\left(2\lambda\sqrt{a}\right) = 0\tag{9}
$$

The Journal of Acoustical Society of India 15 $\rho = r^{2m-2}$ and using $\frac{p^2}{T} = \lambda^2$ Eq. (4) was modified as
 $\frac{(r)}{h} + \left[\lambda^2 r^{2m-2} - \frac{n^2}{r^2} \right] R(r) = 0$ (5)
 $J_n \left(\frac{\lambda}{m} r^m \right) + B Y_n \left(\frac{\lambda}{m} r^n \right)$ (6)

(6) changes to

(6) changes to
 $J_n \left(\frac{\lambda}{m} r^m \right) = 0$ (7)
 For Eq. (9), the frequency ratio of various modes are same as for Eq. (3) given in Table 2. The roots

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| | Table 2. Frequency ratios of modes of vibration. | |
|--|--|------|
| Mode | Frequency ratio | |
| (0,1) | 1.0000 | |
| (1,1) | 2.1355 | |
| (0,2) | 2.2954 | |
| (2,1) | 3.1554 | |
| (1,2) | 3.5001 | |
| (0,3) | 3.5984 | |
| where $\gamma = 2 - 2m$ and substituting this mass density in Eq. (4) and using $\frac{p^2}{T} = \lambda^2$, a differential equation was obtained as | $\frac{d^2R(r)}{dr^2} + \frac{1}{r}\frac{dR(r)}{dr} + \left[\lambda^2 r^{-(2-2m)} - \frac{n^2}{r^2} \right] R(r) = 0$ | (10) |
| Eq. (10) has same the solution given by Eq. (6) . The second part of the solution becomes infinite at centre and hence the second term from the solution is neglected. So | | |
| | $J\frac{n}{m}\left(\frac{\lambda}{m}r^m\right) = R(r)$ | (11) |
| At the boundary, the membrane is fixed at $R(r) = a$. Then | | |
| | $J\frac{n}{m}\left(\frac{\lambda}{m}a^m\right)=0$ | (12) |
| To study the consequences of loading on the drum head, K. N. Rao assumed the density at the | | |

Table 2. Frequency ratios of modes of vibration.

4.3 Studies by K.N. Rao

$$
\frac{d^2R(r)}{dr^2} + \frac{1}{r}\frac{dR(r)}{dr} + \left[\lambda^2 r^{-(2-2m)} - \frac{n^2}{r^2}\right]R(r) = 0\tag{10}
$$

$$
J\frac{n}{m}\left(\frac{\lambda}{m}r^m\right) = R(r) \tag{11}
$$

$$
J\frac{n}{m}\left(\frac{\lambda}{m}a^m\right) = 0\tag{12}
$$

requencies of vibration on a heterogeneous membrane.

Raman, K. N. Rao used mass density of the form
 $\rho = r^{-r}$

msity in Eq. (4) and using $\frac{p^2}{T} = \lambda^2$, a differential equation
 $\left(\frac{r}{r}\right)^{\frac{1}{r}} + \left[\lambda^2 r^{-(2-2m)} - \frac{n$ uencies of vibration on a heterogeneous membrane.

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ty in Eq. (4) and using $\frac{p^2}{T} = \lambda^2$, a differential equation
 $\left[\lambda^2 r^{-(2-2m)} - \frac{n^2}{r^2}\right] R(r) = 0$ (10)

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ty in Eq. (4) and using $\frac{p^2}{T} = \lambda^2$, a differential equation
 $\left[\lambda^2 r^{-(2-2m)} - \frac{n^2}{r^2}\right] R(r) = 0$ (10)

(6) To study the consequences of loading on the drum head, K. N. Rao assumed the density at the boundary to be a constant. To produce a valid finite solution at the origin, the power γ in the density function must be less than zero. To achieve this, *m* must take values between 0 and 1. For a normalised At the boundary, the membrane is fixed at $R(r) = a$. Then
 $J\frac{n}{m} \left(\frac{\lambda}{m}r^m\right) = R(r)$

To study the consequences of loading on the drum head, K. N. Rao ass

boundary to be a constant. To produce a valid finite solution at membrane $R = a = 1$. When $m = 1$, $\rho = r^{2x-2} = 1$ and this is the mass density for ordinary membrane. For mass density in Eq. (4) and using $\frac{p^2}{T} = \lambda^2$, a differential equation
 $\frac{p^2}{r} + \frac{1}{r} \frac{dR(r)}{dr} + \left[\lambda^2 r^{-(2-2m)} - \frac{n^2}{r^2} \right] R(r) = 0$ (10)

ven by Eq. (6). The second part of the solution becomes infinite at

n t **and** $\frac{d^2R(r)}{dr^2} + \frac{1}{r} \frac{dR(r)}{dr} + \left[\lambda^2 r^{r/2-3n/2} - \frac{n^2}{r^2} \right] R(r) = 0$ **(10)

Eq. (10) has same the solution five part of Eq. (6). The second part of the solution becomes infinite at

centre and hence the second te** 6). The second part of the solution becomes infinite at

in is neglected. So
 $\frac{1}{n}r^m = R(r)$ (11)
 $= a$. Then
 $\frac{\lambda}{m}a^m = 0$ (12)

e drum head, K. N. Rao assumed the density at the

inte solution at the origin, the po 6). The second part of the solution becomes infinite at

in is neglected. So
 $\frac{1}{n}r^n = R(r)$ (11)
 $= a$. Then
 $\frac{A}{m}a^m$ = 0 (12)

e drum head, K. N. Rao assumed the density at the

in the outloon at the origin, the p

(12) is modified as

$$
J_0\left(\frac{\lambda}{m}\right) = 0\tag{13}
$$

When $m = 1$, Eq. (13) represents, the eigen value equation of an ordinary circular membrane. When $m = 0$, the density function goes infinity at the centre of the membrane and the value $m = 0$ is excluded.

| m | First root | Second root | Ratio | | | |
|-----|-------------------|--------------------|--------|--|--|--|
| 0.2 | 0.480965 | 1.10402 | 2.2954 | | | |
| 0.4 | 0.96193 | 2.20803 | 2.2954 | | | |
| 0.6 | .44287 | 3.1198 | 2.2954 | | | |
| | | | | | | |

Table 3. Frequency ratio for different m values.

Hence *m* can take values between one and zero. The frequency ratio of (0, 2) mode with different values of *m* between 1 and 0 are given in Table 3. It is seen that the frequency ratio remains the same with different *m*.

5. BI-DENSITY MEMBRANE-THEORY

The composite membrane model¹⁰ with two regions of different sities can produce an integer ratio for frequencies of all the modes $\langle d_2 \rangle$ densities can produce an integer ratio for frequencies of all the modes for Tabla. We are going to find at what density ratio this will happen for Mridangam. In the composite membrane model, the drum head is divided into an inner region and an outer region with radii *p, q* and mass densities $\rm{d_{1}}$ and $\rm{d_{2}}$ as shown in Fig. 4. densities can produce an integer ratio for frequencies of all the modes
for Tabla. We are going to find at what density ratio this will happen
for Mridangam. In the composite membrane model, the drum head is
divided into

Then, $D = \frac{d_1}{d_2}$, the ratio of mass densities and $R = \frac{p}{q}$, the ratio of **Fig. 4.** The represent radii of inner and outer regions on the drum head. For inner and outer

Fig. 4. The representation of two regions on the drum head

 2 2 2 2 2 2 2 2 2

$$
\frac{\partial^2 \psi_2}{\partial r^2} + \frac{1}{r} \frac{\partial \psi_2}{\partial r} + \frac{1}{r^2} \frac{\partial^2 \psi_2}{\partial \theta^2} = \frac{1}{c_2^2} \frac{\partial^2 \psi_2}{\partial t^2}
$$
(15)

Then, $D = \frac{1}{d_2}$, the ratio or mass densities and κ :

radii of inner and outer regions on the drum head. For

regions, the wave equations are
 $\frac{\partial^2 \psi_1}{\partial r^2} + \frac{1}{r} \frac{\partial \psi_1}{\partial r} + \frac{1}{r^2}$

where $c_1 = \sqrt{\frac{T}{d_$ **Driverse and the wave equations are**
 $c_1 = \sqrt{\frac{T}{q_1}}$, $c_2 = \sqrt{\frac{T}{q_2}}$. At boundary α is defined the circuities of all the circuities of all the modes

in the composite membrane model, the drum head is

in the an in continuous, since the entire membrane vibrates when the drum head is struck. To have a finite solution everywhere on the drum head, the function must be differentiable everywhere. Then, the derivative of $\frac{\partial^2 \psi_2}{\partial r^2} + \frac{\partial \psi_2}{\partial r} + \frac{1}{r} \frac{\partial^2 \psi_2}{\partial r^2} = \frac{1}{c_2^2} \frac{\partial^2 \psi_2}{\partial t^2}$

where $c_1 = \sqrt{\frac{T}{d_1}}$, $c_2 = \sqrt{\frac{T}{d_2}}$. At boundary $\psi_2(q) = 0$. At the circumference of the inner region, t

continuous, since th gion with radii p, q and

and $R = \frac{p}{q}$, the ratio of **Fig. 4.** The representation of

ead. For inner and outer two regions on the drum head
 $\frac{\partial \psi_1}{\partial r} + \frac{1}{r^2} \frac{\partial^2 \psi_1}{\partial \theta^2} = \frac{1}{c_1^2} \frac{\partial^2 \psi_2}{\partial t^2}$ (14) **and** $R = \frac{p}{q}$, the ratio of **Fig. 4.** The representation of head. For inner and outer two regions on the drum head $\frac{1}{r} \frac{\partial w_1}{\partial r} + \frac{1}{r^2} \frac{\partial^2 w_1}{\partial \theta^2} = \frac{1}{c_1^2} \frac{\partial^2 w_2}{\partial t^2}$ (14)
 $\frac{1}{r} \frac{\partial w_3}{\partial r$ and $R = \frac{p}{q}$, the ratio of **Fig. 4.** The representation of

ead. For inner and outer two regions on the drum head
 $\frac{\partial w_1}{\partial r} + \frac{1}{r^2} \frac{\partial^2 w_1}{\partial \theta^2} = \frac{1}{c_1^2} \frac{\partial^2 w_2}{\partial t^2}$ (14)
 $\frac{\partial w_2}{\partial r} + \frac{1}{r^2} \frac{\partial$ region with radii p, q and

es and $R = \frac{p}{q}$, the ratio of Fig. 4. The representation of

two regions on the drum head
 $+\frac{1}{r}\frac{\partial \psi_1}{\partial r} + \frac{1}{r^2}\frac{\partial^2 \psi_1}{\partial \theta^2} = \frac{1}{c_1^2}\frac{\partial^2 \psi_2}{\partial t^2}$ (14)
 $+\frac{1}{r}\frac{\partial \psi_2}{$ s and $R = \frac{p}{q}$, the ratio of Fig. 4. The representation of

thead. For inner and outer two regions on the drum head
 $\frac{1}{r} \frac{\partial \psi_1}{\partial r} + \frac{1}{r^2} \frac{\partial^2 \psi_1}{\partial \theta^2} = \frac{1}{c_1^2} \frac{\partial^2 \psi_2}{\partial t^2}$ (14)
 $\frac{1}{r} \frac{\partial \psi_$ $\frac{\partial^2 y}{\partial r^2} + \frac{\partial^2}{r} \frac{\partial r}{\partial r^2} + \frac{\partial^2 y}{\partial r^2} = \frac{1}{c_1^2} \frac{\partial^2 y}{\partial t^2}$ (14)
 $\frac{\partial^2 y_2}{\partial r^2} + \frac{1}{r} \frac{\partial y_2}{\partial r} + \frac{1}{r^2} \frac{\partial^2 y_2}{\partial \theta^2} = \frac{1}{c_2^2} \frac{\partial^2 y_2}{\partial t^2}$ (15)

lary $\psi_2(q) = 0$. At the circumfere $\frac{\partial^2 w_2}{\partial r^2} + \frac{1}{r} \frac{\partial w_2}{\partial r} + \frac{1}{r^2} \frac{\partial^2 w_2}{\partial \theta^2} = \frac{1}{c_2^2} \frac{\partial^2 w_2}{\partial t^2}$ (15)
 y $\psi_2(q) = 0$. At the circumference of the inner region, the function is

ane vibrates when the drum head is struck. To

$$
\frac{\psi_1(p) = \psi_2(p)}{dp} = \frac{d\psi_2(p)}{dr}
$$

These are the conditions used to solve the wave equations of two regions. Using these conditions Ramakrishna and Sonthi arrived at an eigen value equation that give integer ratio for frequencies of different modes. The model also produced the frequency ratio of degenerate modes observed in Tabla everywhere on the drum head, the function must be differentiable everywhere. Then, the derivative of

the function must exist on the entire membrane. Hence
 $\psi_1(p) = \psi_2(p)$
 $\frac{d\psi_1(p)}{dr} = \frac{d\psi_2(p)}{dr}$

These are the co

$$
D\frac{J_{n-1}(DRx)}{J_n(DRx)} = \frac{Y_n(x)J_{n-1}(Rx) - J_n(x)Y_{n-1}(Rx)}{Y_n(x)J_n(Rx) - J_n(x)Y_n(Rx)}
$$
(16)

where $J_n(Rx)$ is Bessel function of first order and $Y_n(Rx)$ is the modified Bessel function of first order.

6. OBSERVATIONS

A commonly played Mridangam also have a pasted inner region and an outer region like Tabla. A practical Mridangam has a radius of 4 cm for its pasted region and it has 9 cm radius for its outer region on the drum head. Hence our *R* is 0.44. Using the mass density ratio $D = 3.125$ suggested in reference¹⁰ we get frequency ratios as given in Table 4. The ratios showed much deviation from expected integer values.

In reference¹⁰ Ramakrishna and Sonthi identified $R = 0.4$ in commonly used Tabla. By keeping $R =$ 0.4, different values of *D* are given to obtain the frequency ratio of different modes. The frequency ratio of modes of vibration calculated by fixing *R* and choosing different values of *D* in Eq. (16) are given in

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| Mode | Frequency ratio | |
|-------|------------------------|--|
| (0,1) | 0.95 | |
| (1,1) | 1.81 | |
| (0,2) | 2.85 | |
| (2,1) | 2.72 | |
| (1,2) | 3.84 | |
| (3,1) | 3.64 | |
| (0,3) | 4.73 | |
| (2,2) | 4.84 | |
| (4,1) | 4.54 | |

Table 4. Frequency ratios for $R = 0.44$ and $D = 3.125$

Table 5. The frequency ratio of the lowest mode is obtained around 1 at *D* = 3. An increment of 0.1 for the value of *D* made the value closer to one. A further increment of 0.05 is made to *D* and the ratio of lowest mode turned out as less than 1. Hence for further calculation, the mean value between 3.1 and 3.15 is chosen and frequency ratios are found. The process is repeated and an appropriate ratio is obtained at $D = 3.140625$. By choosing the different values of D it is seen that as the mass density ratio increases, the frequency ratio decreases. Frequency ratio with good harmonic relationship is obtained at *D* = 3.140625. The instrument makers produce a black pasted region on the drum head in such a way that radius ratio and mass density ratio is achieved in this range.

| Mode | Frequency ratio for $R = 0.4$ | | | | | | |
|-------|-------------------------------|---------|-----------|------------|--------------|-------------|----------|
| | $D=3$ | $D=3.1$ | $D=3.125$ | $D=3.1375$ | $D=3.140625$ | $D=3.14375$ | $D=3.15$ |
| (0,1) | 1.04 | 1.01 | 1.00 | 1.00 | 1.00 | 1.00 | 0.99 |
| (1,1) | 2.02 | 1.96 | 1.94 | 1.94 | 1.94 | 1.93 | 1.93 |
| (0,2) | 3.17 | 3.09 | 3.07 | 3.06 | 3.05 | 3.05 | 3.05 |
| (2,1) | 3.07 | 2.98 | 2.96 | 2.95 | 2.95 | 2.95 | 2.94 |
| (1,2) | 4.24 | 4.14 | 4.12 | 4.11 | 4.10 | 4.10 | 4.09 |
| (3,1) | 4.14 | 4.02 | 3.99 | 3.97 | 3.97 | 3.97 | 3.96 |
| (0,3) | 4.93 | 4.86 | 4.85 | 4.84 | 4.84 | 4.83 | 4.83 |
| (2,2) | 5.30 | 5.20 | 5.17 | 5.16 | 5.16 | 5.15 | 5.15 |
| (4,1) | 5.19 | 5.03 | 4.99 | 4.97 | 4.97 | 4.96 | 4.96 |

Table 5. Frequency ratios of modes.

According to Raman, five sustained musical tones are produced by Mridangam. From Table 5 it is seen that fundamental mode (0, 1) and first overtone mode (1, 1) have independent frequency ratios. Modes (0, 2), (2, 1) have ratios around 3.00. They vibrate with nearby frequency and they are degenerate modes. Hence the vibrations of these modes create a single harmonic tone. Similar ratios are seen for (1, 2) and (3, 1) modes and they are also degenerate modes of vibration. So, single tone is produced by (1, 2) and $(3, 1)$ modes. The vibration of three modes $(0, 3)$, $(2, 2)$ and $(4, 1)$ produce ratios around 5.00 and they also forms a set of degenerate modes that produce a single tone. Hence all the tones identified by Raman in his experiment are found from the model.

The drum Mridangam originated in ancient times and hence its construction must have changed many times with many modifications. The word Mridangam itself indicates part made with mud and the descriptions in literature like Natyasastra shows that the central loaded region on the drum head was

earlier made with mud¹¹⁻¹². Subsequently, loading with stone and rice emerged and is now common in use.

As the construction evolved with time, mostly trial and error methods, no standardisation in the parameters or process can be seen. Hence the exact value of either mass density ratio or radius ratio is not followed by the makers. In 2006, S. Gaudet *et al*. mathematically tried to answer the possibility of other configurations for Tabla13. They found new configuration of Tabla at $D = 2.9$ and $k = 0.38$. We calculated the frequency ratios of Tabla with the values given by Gaudet et al. and the obtained ratios are tabulated in Table 6. In this new configuration unlike one predicted by Ramakrishna and Sonthi, the

Table 6. Frequency ratios for $R = 0.38$ and $D = 2.9$

fifth harmonic tone is produced by modes $(1, 3)$, $(2, 2)$ and $(4, 1)$. The ratio of $(0, 3)$ is shifted near to 6.00 which indicates that the mode contributes a new tone. Hence in this new configuration, Tabla can generate 6 harmonic tones.

By keeping $R = 0.38$ we analysed the possible values of *D* for Mridangam. Using Eq. (16) the obtained values are given in Table 7. Good harmonic relationship for frequency ratio is obtained at $D =$ 2.8515625 by keeping the maximum deviation from integer ratio as 0.1.

A further improved frequency ratios are obtained by changing *R* to 0.384 at $D = 2.835$. The obtained ratios are shown in Table 8. The obtained value of R indicates that the pasted region span across 38.4 percent of the radius of the drum head in Mridangam.

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7. CONCLUSIONS

In Mridangam, the mass density and radii of two regions on the membrane influences the ratios produced by the drum. In South Indian music two variants of Mridangam are commonly played - male and female. Male Mridangam is a low pitched Mridangam whereas female Mridangam is played at high pitch. Generally, pitch and the radius of Mridangam are inversely related. Hence the first configuration with $R = 0.4$ and $D = 3.140625$ represents a male variant. The second configuration with $R = 0.384$ and *D* = 2.835 represent a female type.

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Marine propeler noise control by ducted isolation – A study

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ABSTRACT

The present work aims at a numerical study of controlling the propeller noise by using the method of isolation. In this method four types of ducts are modeled around 6 bladed propellers and are analyzed. Cylindrical, Profile, Convergent and Convergent-divergent ducts are studied. Unsteady non-cavitating noise of ducted propellers at its rotating speed and vehicle speed is predicted. Numerical simulations are carried out using a finite volume code FLUENT. The output of CFD results is used for Acoustic analysis. The methodology adopted in CFD analysis is large eddy simulation (LES) and in acoustical analysis it is Ffowcs William's-Hawkings (FW-H) formulation. Noise spectrum is predicted over the frequency range of 0-2 kHz. From this numerical study on these propellers, duct configuration for least noise and better performance of propeller is identified.

1. INTRODUCTION

Propeller noise mainly depends on Propeller geometry, Propeller wake inflow, and Propeller isolation. Due to the pressure difference in rear and aft end of the propeller blade the noise is generated. Reducing pressure oscillations on the propeller is one of the most effective ways of reducing the radiated noise. The parameters which influence the noise levels produced by the propeller are number of blades, pitch angle, blade area, diameter of blade, skew angle, Trailing edge geometry and Propeller blade finishing fineness. This noise can also be reduced by designing a duct around the propeller and this method is known as propeller isolation. In this study, an existing six bladed propeller is selected. Four types of ducts are designed around the propellers which are cylindrical duct, profile duct, convergent duct and convergent-divergent duct. The prediction of non-cavitation noise of propeller is carried out by using FW-H equation and eddy viscosity model of large eddy simulation (LES) in computational fluid dynamics Fluent software. The study resulted in understanding the effect of ducts on propeller noise.

2. LITERATURE REVIEW

Over the years Propeller noise control has been studied by various researchers. In this section some of the studies reported by researches on topic are presented.

K. P. Santhosh Babu *et al.* **(1) :** Marine ducted propeller is a rotating duct fan that is used on tugs and trawlers which creates a greater propulsive thrust force to drive over a water medium in heavy

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working condition in harbor. This paper progresses with comprehensive information of marine ducted propeller having blade made of aluminum alloy and duct of alloy steel designed and analyzed with various blade formations of 4 and 5 separately. Main aim is to check the performance of each blade individually to show which blade performs better with maximum velocity rate under stream line motion in water at dynamic condition. Ducted propeller is modeled in solid works. Hydrostatic and hydrodynamic analyses of each blade are performed with ANSYS workbench

R. Bontempo *et al.* **(2) :** This paper depicts the investigation of the flow around a ducted propeller ducted with a so-called accelerating duct. In this paper, axial momentum theory and nonlinear actuator disk methods are used. The straightforward application of the first approach reveals that if the duct and rotor thrusts are concordant, then a beneficial effect on the propulsive efficiency can be readily obtained by enclosing a propeller in an accelerating duct. When the more advanced nonlinear actuator disk method is applied to verify the outcomes of the axial momentum theory additional information on the performance of the device are obtained. Moreover, the non-linear actuator disk method is also employed to investigate, through experimental design techniques, the effect of the key geometrical parameters of the duct on the efficiency and robustness of this kind of propulsive system. In particular, it has been found that a propulsive efficiency gain can be achieved through a duct thickness, camber and chord increase, and through an inci-dence decrease.

Mehdi Chamanara *et al.* **(3) :** In their study, the effect of the duct angle and propeller location on the hydrodynamic characteristics of the ducted propeller using Reynolds-Averaged Navier Stokes (RANS) method is reported. A Kaplan type propeller is selected with a 19A duct. The ducted propeller is analyzed by three turbulence models including the k- ε standard, k- ω SST and Reynolds stress model (RSM). The numerical results are compared with experimental data. The effects of the duct angle and the location of the propeller inside the propeller are presented and discussed.

Tadeusz Koronowicz *et al.* **(4) :** This article describes new computer system which is specific for the design of ducted propellers. This system concerns first of all the procedures for the design calculation of ducted propellers and for the analysis of the ducted propeller operation in the nonuniform velocity field behind the ship hull. The comparative analysis of computation results for different types of ducts is also presented.

João Manuel Baltazar *et al.* **(5) :** In their work, a comparison between the results obtained by a panel code with a Reynolds-averaged Navier-Stokes (RANS) code is made to obtain a better in-sight on the viscous effects of the ducted propeller. They also studied the limitations of the inviscid flow model, especially near bollard pull conditions or low advance ratios, which are important in the design stage. From the comparison, several modeling aspects are studied for improvement of the inviscid (potential) flow solution. Finally, the experimental open-water data is compared with the panel method and RANS solutions. A strong influence of the blade wake pitch, especially near the blade tip, on the ducted propeller force predictions is seen. A reduction of the pitch of the gap strip is proposed for improvement of the performance prediction at low advance ratios.

3. BRIEF DESCRIPTION OF PROPELLER

The dimensions of the reference propeller configuration studied are mentioned in Table 1.

Propeller blade is made of Forged Aluminium Al-24345 alloy and its Material Specifications are depicted in Table 2.

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Table 2. Material Specifications.

4. MODELLING OF DUCTS

Modeling of the propeller was carried out using CATIA V5 R20. The model is generated by using propeller geometry of each section of the blade at various radii *i.e.* main geometrical characteristics of propeller geometry of cylinder blade at different sections.

The dimensions of the four types of ducts namely Cylindrical, Profile, Convergent and Convergentdivergents are given in Table 3.

Model of the 6-blade reference propeller is shown in Fig. 1. Solid models of 4 types of ducts are shown from Fig. 2 to 5.

Fig. 1. Reference propeller

Fig. 2. Cylindrical duct **Fig. 3.** Profile duct

Fig. 4. Convergent duct **Fig. 5.** Convergent-divergent duct

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5. ANALYSIS

The modeling approach presented in this paper is similar to the approach by various authors who had contributed their research in the field of propeller duct designs. The aim was to assess the importance of ducts to reduce the induced noise during the operation of propeller. This can lead to a reliable and versatile simulation setup which may be used for tests of different propeller ducts and the selection of best duct with low sound pressure level.

5.1 Meshing

The type of mesh used for this analysis is as follows

Type of Element : Tetrahedral, Mesh type: Fine, Mesh Size: 11 mm.

Number of Nodes and Elements of all 4 types of ducts are as shown in Table 4.

Optimum mesh size of 11 mm is arrived through iterative study by using various mesh sizes.

The meshed model of cylindrical ducted Propeller is shown in Fig. 6.

| S. no | Type of Duct | Number of Nodes | Number of Elements |
|-------|---------------------------|------------------------|---------------------------|
| | Cylindrical Duct | 48306 | 239855 |
| 2 | Profile Duct | 47154 | 236178 |
| | Convergent Duct | 47793 | 243450 |
| 4 | Convergent-Divergent Duct | 45880 | 234328 |

Table 4. Number of Nodes and Elements of all 4 types of ducts.

Fig. 6. Meshed model of cylindrical duct

5.2 CFD analysis

Numerical simulations have been carried out with a finite volume module called FLUENT. In the CFD analysis, LES turbulence model is used. Surfaces which can rotate relatively are defined as moving wall and are dependent on the fluid around them. Fluid Zone has to be considered as a cylinder extending either side of the bladed propeller. Cylinder walls are categorized as stationary wall and the inlet and outlet are defined as velocity inlet (7.08 m/sec) and outflow. Fluid zone in the inner volume is defined as moving mesh at 780 rpm in x-direction.

With this configuration, CFD analysis is carried out on all the four kinds of ducted propellers using large eddy simulation (LES).

Contours of Total Pressure (pascal) E S $72e + 05$ $63e + 05$ $\star^{\uparrow}_{\downarrow}$ $.53e + 05$ $.44e + 05$ $^{\circ}$ $.35e + 05$ $26e + 05$ $^{\circledR}$ 16e+05 $07e + 05$ $.77e + 04$ P $8.84e + 04$ $91e+04$ $.99e + 04$ $^{\circ}$ 6.06e+04 $5.13e + 04$ ¥ $4.20e + 04$ $3.27e + 04$ σ ā $.34e + 04$ $1.42e+04$
4.88e+03 ४ $-4.40e + 03$ OQ. $-1.37e + 04$

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Fig. 7. Total pressure distribution of Cylindrical ducted propeller

Results of CFD analysis of cylindrical ducted propeller is shown in Fig. 7.

6. ACOUSTIC ANALYSIS

The output of CFD results is carried further for Acoustic analysis. Acoustic analysis was done in ANSYS using Ffowcs Williams-Hawkings (FW-H) formulation. Noise spectrum was predicted over the frequency range of 0-2 kHz. Noise spectra for cylindrical ducted propeller is shown in Fig. 8.

6.1 Comparison of Sound pressure level (SPL) of ducted propellers

Sound Pressure level (SPL) of all 6 bladed propeller with four kinds of ducts are given in Table 5.

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| | Table 5. Comparison of SPL of ducted propellers with Reference propeller. | | | | | | |
|--|--|--------------------------------|-----------------------------------|---|--|--|--|
| Reference Propeller SPL(dB) | Cylindrical Duct SPL(dB) | Profile Duct SPL(dB) | Convergent Duct SPL(dB) | Convergent divergent Duct SPL (dB) | | | |
| 130 | 122.5 | 127 | 125.6 | 128 | | | |
| | but cylindrical ducted propeller provide least noise among studied configurations. | | | It can be seen from table 5, compared to reference propeller, all ducted propellers provide lesser noise | | | |
| | 7. CALCULATION OF THRUST AND TORQUE COEFFICIENTS | | | | | | |
| | from the CFD analysis is shown in Table 6. Operating conditions are : | | | Thrust and Torque values of Reference and 04 varieties of ducted Propellers which were obtained | | | |
| | Density of sea water (ρ) =1029 kg/m ³ | | | | | | |
| | Diameter of propeller (D) =0.389 m | | | | | | |
| | The Thrust and Torque coefficient were calculated from the following equations. | | | | | | |
| Thrust coefficient, $K_T = \frac{T}{\rho n^2 D^4}$ | | | | | | | |
| Torque coefficient, $K_Q = \frac{Q}{\rho n^2 D^5}$ | | | | | | | |
| | propeller. A Comparison of Thrust coefficient and Torque coefficients is shown in Table 7. | | | Using the above, Thrust and Torque coefficients are calculated for ducted propeller and reference | | | |
| | | | | From the above table, Thrust and Torque coefficients are increased for cylindrical ducted propeller compared to reference propeller. Even though Convergent and Convergent Divergent ducted propellers have higher Thrust coefficient, lower noise is from Cylindrical ducted propeller while satisfying required | | | |

Table 5. Comparison of SPL of ducted propellers with Reference propeller.

7. CALCULATION OF THRUST AND TORQUE COEFFICIENTS

From the above table, Thrust and Torque coefficients are increased for cylindrical ducted propeller compared to reference propeller. Even though Convergent and Convergent Divergent ducted propellers have higher Thrust coefficient, lower noise is from Cylindrical ducted propeller while satisfying required Thrust and torque.

7. CONCLUSION

The non-cavitating underwater noise of ducted propellers has been studied through Numerical approaches. It is seen that ducting provides reduction in noise. Further, it is seen that propeller with cylindrical duct generates lesser sound than profile, Convergent-divergent and convergent ducted

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propeller. Thrust and Torque coefficients are better for cylindrical ducted propeller compared to reference propeller thereby implying that cylindrical ducted propeller is optimum choice.

8. ACKNOWLEDGEMENTS

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Marine propeller noise control using ducts with stator

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ABSTRACT

To evade detection by enemies, the underwater-radiated noise of marine vehicles must be reduced. The propeller, machinery, and flow cause this noise. Many researchers devised ways for reducing noise from each of these sources. According to the literature, a reasonable knowledge of machinery noise control has been established, but propeller noise and flow noise management still require further study. Because flow noise contributes so little to underwater radiated noise, propeller noise reduction readily emerges as a worthwhile subject. Propeller noise is controlled in principle by geometry modifications, wake modification and isolation.

The present work aims at a numerical study of controlling the propeller noise by using the method of isolation. In this method, four types of ducts and a stator are modeled around 6 bladed propeller and are analyzed. First type of duct is a cylindrical, second type is profile, third type is convergent duct and fourth type is convergent-divergent duct. Unsteady non-cavitating noise of ducted propellers with stators at its rotating speed and vehicle speed is predicted and studied. Modelling of propellers is carried out using CATIA V5. In order to obtain better results, the numerical simulations are carried out using a finite volume code FLUENT. The methodology adopted in CFD analysis is large eddy simulation (LES) and in acoustical analysis is Ffowcs William's-Hawkings (FW-H) formulation. The output of CFD results is used for Acoustic analysis. Noise spectrum is predicted over the frequency range of 0-2 kHz for 6 bladed propellers of four types of ducts with stator. From this numerical study on these propellers, duct with stator configuration for least noise and better performance of propeller is identified

1. INTRODUCTION

Propeller noise of Marine vehicles like Torpedo, Submarine and Warships is to be controlled from stealth point of view. The radiated noise from the propeller depends on Propeller geometry, Propeller wake inflow, and Propeller isolation. Due to the pressure difference in rear and aft end of the propeller blade the noise is generated. Reducing pressure oscillations on the propeller is one of the most effective ways of reducing the radiated noise. The parameters which influence the noise levels produced by the propeller are changing the number of blades, changing the Pitch angle, change in blade area, changing the diameter of blade, change in Skew angle, Trailing edge geometrical modifications and Propeller blade finishing fineness. This noise can also be reduced by designing a duct around the propeller and this method is known as propeller isolation. In this study, a six bladed propeller of existing propeller is taken as

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reference, is used and four types of ducts with stator are designed around the propeller which are cylindrical duct with stator, profile duct with stator, convergent duct with stator and convergent-divergent duct with stator. The diameter of propeller is 0.389m and hub to propeller diameter ratio is 0.25. Prediction of non-cavitation noise of propeller was carried out by numerical method at rotating speed of 780 rpm at vehicle speed of 7.08 m/s. The prediction of non-cavitation noise of propeller was done by using FW-H equation and eddy viscosity model of large eddy simulation (LES) in computational fluid dynamics Fluent software. The study resulted in understanding the effect of ducts on decrease and increase of propeller noise.

2. LITERATURE REVIEW

Over the years Propeller noise control has been studied by various researchers. In this section some of the studies reported by researches on topic are presented.

K. P. Santhosh Babu *et al.* **(1) :** Marine ducted propellers are rotating duct fan that are used on tugs and trawlers which creates a greater propulsive thrust force to drive over a water medium on heavy working condition in harbor. This paper progresses with comprehensive information of marine ducted propeller having blade made of aluminum alloy and duct of alloy steel is designed and analyzed with various blade formations of 4 and 5 separately. Main aim is to check the performances of each blade individually to show which blade performs efficiently better with maximum velocity rate under stream line motion on water at dynamic condition. Ducted propeller is modeled in solid works. Hydrostatic and hydrodynamic analyses of each blade are performed with ANSYS workbench

R. Bontempo *et al.* **(2) :** This paper depicts the investigation of the flow around a ducted propeller ducted with a so-called accelerating duct.Main aim is, both the axial momentum theory and a nonlinear actuator disk method are used. The straightforward application of the first approach reveals that if the duct and rotor thrusts are concordant, then a beneficial effect on the propulsive efficiency can be readily obtained by enclosing a propeller in an accelerating duct. When the more advanced nonlinear actuator disk method is applied to verify the outcomes of the axial momentum theory additional information on the performance of the device are obtained. Moreover, the nonlinear actuator disk method is also employed to investigate, through experimental design techniques, the effect of the key geometrical parameters of the duct onto the efficiency and robustness of this kind of propulsive system. In particular, it has been found that a propulsive efficiency gain can be achieved through a duct thickness, camber and chord increase, and through an incidence decrease.

Mehdi Chamanara *et al.* **(3) :** In this study, the effect of the duct angle and propeller location on the hydrodynamic characteristics of the ducted propeller using Reynolds-Averaged Navier Stokes (RANS) method is reported. A Kaplan type propeller is selected with a 19A duct. The ducted propeller is analyzed by three turbulence models including the k- ε standard, k- ω SST and Reynolds stress model (RSM). The numerical results are compared with experimental data. The effects of the duct angle and the location of the propeller inside the propeller are presented and discussed.

Tadeusz Koronowicz *et al.* **(4) :** The computer system for the completed design of the ducted ship propellers has some common blocks andprocedures. This article describes only these blocks and procedures which are specificfor the design of ducted propellers. These new blocks concern first of all the procedures for the designcalculation of ducted propellers and for the analysis of the ducted propeller operation in the non-uniformvelocity field behind the ship hull. The com-parative analysis of computation results for different types ofducts is also presented.

João Manuel Baltazar *et al.* **(5) :** Inthis work, a comparison between the results obtained by a panel code with a Reynolds-averaged Navier-Stokes (RANS) code is made to obtain a better in-sight on the viscous effects of the ducted propeller. In this paper also studied on the limitations of the inviscid flow model, especially nearbollard pull conditions or low advance ratios, which are important in the design stage. The analysis is carried out for propeller Ka470 operating inside duct 19A. From the comparison, several modeling aspects are studied for improvement of the inviscid (potential) flow solution. Finally,

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the experimental open water data is compared with the panel method and RANS solutions. A strong influence of the blade wake pitch, especially near the blade tip, on the ducted propeller force predictions is seen. A reduction of the pitch of the gap strip is proposed for improvement of the performance prediction at low advance ratios.

Negin Donyavizadeh (6) : In this study, hydrodynamic performance of the linear jet propulsion system is numerically investigated. Accordingly, the Ansys CFX software is utilized and RANS equations are solved using the SST turbulent model. The results of the proposed numerical model, in the form of thrust and torque coefficient as well as efficiency, are compared with available experimental data for a ducted propeller, and good compliance is achieved. Considering the importance of stator cross section on the performance of the linear jet propulsion system, the influence of thickness and camber size of the stator on linear jet propulsion systems are examined. Based on the numerical findings, it is determined that at constant advance ratio, with increasing thickness of stator, the efficiency increases. It is also observed that as the span length increases, the maximum and minimum of the pressure coefficient increase for different thicknesses. Furthermore, it is seen that positive and negative pressure coefficients decrease with an increase in foil thickness.

3. NUMERICAL MODEL OF PROPELLER

3.1 Modelling of ducts

Modeling of propeller was carried out using CATIA V5 R20. Propeller modeling is carried out by using propeller geometry of each section of the blade at various radii i.e. main geometrical characteristics of propeller geometry of cylinder blade at different sections were used. In this paper four kinds of ducts with stators are modeled around the 6 blade reference propeller is as shown in fig. 1. Solid model of 4 types of ducts with 7 blade stator are shown in below fig. 2 to fig. 6. The dimensions for the stator and four types of ducts namely cylindrical, profile, convergent, convergent-divergent with stators are given in table 1 and 2.

Fig. 1. Reference propeller **Fig. 2.** Stator

Fig. 3. Cylindrical duct **Fig. 4.** Profile duct

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Fig. 5. Convergent duct **Fig. 6.** Convergent-divergent duct

3.2 Dimensions of Stator

3.3 Dimensions of Ducts

| Duct | Inner radius (mm) | Outer radius (mm) | Length (mm) | Thickness (mm) | | | | |
|----------------------|----------------------|----------------------|----------------|--------------------------|--|--|--|--|
| Cylindrical | 195.5 | 195.5 | 300 | | | | | |
| Profile | 195.5 | 165 | 300 | 12 | | | | |
| Convergent | 230 | 165 | 300 | 12 | | | | |
| Convergent-divergent | 230 | 240 | 300 | | | | | |

Table 2. Dimensions of Ducts.

4. CFD ANALYSIS

The aim was to assess the importance of kind of ducts with stator to reduce the induced noise during the operation of propeller. This can lead to a reliable and versatile simulation setup which may be used for tests of different propeller ducts with stator and the selection of best duct with low sound pressure level.

To get better results the numerical simulations have been carried out with a finite volume module called FLUENT. In the CFD analysis, LES module turbulence model is used. Surfaces which can rotate relatively are defined as moving wall and are dependent on the fluid around them. Fluid Zone has to be considered as a cylinder extending either side of the bladed propeller. Cylinder walls are categorized as stationary wall and the inlet and outlet are defined as velocity inlet (7.08 m/sec) and outflow. Fluid zone in the inner volume is defined as moving mesh at 780 rpm in x-direction.

With this configuration CFD analysis is carried out on all the four kinds of ducted propellers with stator using large eddy simulation (LES).

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| | | Table 3. Details of non-cavitating flow. | |
|--|--|--|--|
|--|--|--|--|

4.1 Meshing

ANSYS ICEM CFD meshing software has advanced CAD readers and repair tools to mesh the complex geometries. ICEM CFD numerical code is used for meshing in order to obtain grid independence.

The type of mesh used for this analysis is as follows Elementtype : Unstructured tetrahedral

Order : Quadratic

The meshed models of all 6 bladed marine ducted propellers with stators are shown in the below figures 7 to 11.

Fig. 7. Meshed cylindrical duct **Fig. 8.** Meshed profile duct

Fig. 9. Meshed convergent duct **Fig. 10.** Meshed convergent-divergent duct

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Fig. 11. Cut plane of inner and outer domain

4.2 Calculation of Thrust and Torque Coefficients from ANSYS

The force component *i.e.,* is directly obtained from the CFD analysis is considered as Thrust.

Operating conditions required for solving Thrust coefficient and Torque coefficient :

Density of seawater (ρ) : 1029 kg/m³ Diameter of propeller (D) : 0.389 m Speed of the propeller (n) : 13 rps

Thrust and Torque values of Reference and 04 varieties of ducted Propellers with stators which were obtained from the CFD analysis is shown in Table 4

| Parameter | Reference Propeller | Cylindrical duct with stator | Profile duct with stator | Convergent duct with stator | Convergent Divergent duct with stator | |
|---------------|-------------------------------|------------------------------------|--------------------------------|-----------------------------------|---|--|
| Thrust (N) | 1385.73 | 2585 | 1888 | 6325 | 3099 | |
| $Torque(N-m)$ | 51.89 | | 84 | 76 | 72 | |

Table 4. Values of thrust and torque of ducted propellers with Reference propeller.

4.3 Comparison of Thrust and Torque coefficients

Using the above, Thrust and Torque coefficients are calculated for ducted propeller with stators and reference propeller. A Comparison of Thrust coefficient and Torque coefficients is shown in Table 5.

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From the above table, Thrust and Torque coefficients are increased for cylindrical ducted propeller with stator compared to reference propeller. Even though Convergent and Convergent Divergent ducted propellers with stator have higher Thrust coefficient, lower noise is from cylindrical ducted propeller while satisfying required Thrust and torque.

5. ACOUSTIC ANALYSIS

The output of CFD results is carried further for Acoustic analysis. Acoustic analysis was done in ANSYS using Ffowcs Williams-Hawkings (FW-H) formulation. The sound pressure level was predicted over the frequency range of 0-2 kHz for 6 bladed propeller with four different kinds of ducts with stator. Receiver is placed at 1m. Reference pressure is taken as 1e-06. Noise spectra for cylindrical ducted propeller with stator is shown in Fig. 12.

Fig. 12. Frequency vs sound pressure level graph of cylindrical ducted propeller with stator

5.1 Comparison of Sound pressure level (SPL) of ducted propellers

Sound Pressure level (SPL) of all 6 bladed propeller with four kinds of ducts with stators are given in Table 6.

It can be seen from table 6, compared to reference propeller, all ducted propellers provide lesser noise but cylindrical ducted propeller with stator provide least noise among studied configurations.

6. CONCLUSION

The Non-Cavitating noise of underwater marine propeller has been carried out by numerical method. The numerical investigation of turbulent flow is carried out using Large Eddy Simulation (LES) approach in CFD analysis and using Ffowcs Williams -Hawkings (FW-H) in computational acoustic analysis to find Marine propeller noise control using ducts with stator

overall Sound Pressure Level (SPL) of a propeller with duct and stator at rotational speed of 780 rpm and flow velocity at 7.08 m/s. From the analysis, it is seen that propeller with cylindrical duct with stator generates less noise of 112.22 dB than Profile duct with stator, Convergent duct with stator and Convergent divergent duct with stator. Thrust and Torque coefficients are better for cylindrical ducted propeller compared to reference propeller thereby implying that cylindrical ducted propeller with stator is optimum choice.

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Evaluation and analysis of Kirkwood-Buff integrals of 1, 4-dioxane + aromatic hydrocarbon binary mixtures using inversion procedure and regular solution theory from ultrasonic speed and density data

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ABSTRACT

The Kirkwood-Buff (K-B) theory can be used to characterize the intermolecular interactions in liquid mixtures. The interactions are characterized by the K-B parameters, G_{AA} , G_{BB} and G_{AB} , which reflect interaction between like-like and like-unlike species in the mixture. The experimental ultrasonic speed and density data of 1, 4-dioxane + benzene/toluene/*o*-xylene/*m*-xylene/*p* xylene/mesitylene binary mixtures at 298.15 K have been used to calculate the K-B integrals by using the inverse procedure and regular solution theory. This procedure utilizes experimental thermodynamic properties of mixtures, *viz.,* partial molar volume, isothermal compressibility, excess free energy and partial vapour pressure. The K-B parameter, G_{AB} obtained using this procedure indicated that the correlation/interaction between 1, 4-dioxane and aromatic hydrocarbon molecules follow the order: benzene > toluene > *p*-xylene > *m*-xylene > *o*-xylene > mesitylene, which is found in good agreement with the trends exhibited by the excess properties of these mixtures obtained experimentally.

1. INTRODUCTION

The physicochemical and thermodynamic properties of liquid mixtures provide valuable information regarding intermolecular interactions between the component molecules¹⁻⁴, which is of significance in many theoretical and applied areas of research and such proprties are regularly required in designing many chemical and industrial processes⁵⁻⁸. Although there exists a large amount of experimental data on the physicochemical, thermodynamic, transport, acoustic and spectroscopic properties of liquid systems in the literature⁹⁻¹⁸, but in comparison, there exists very few theoretical reports on the elucidation of molecular structure and valuation of thermodynamic properties of liquid mixtures using the existing theories of solutions. The researchers in this area have focused their interest significantly on the molecular structure along with some representative macroscopic properties that assists to characterize it. The theoretical prediction of physicochemical properties of liquid systems comprises an interdisciplinary interest and

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proved to be an excellent qualitative and quantitative tool for explaining the molecular structure and nature and extent of interaction in liquid systems.

Kirkwood & Buff¹⁹ theory of solutions relate the radial distribution functions of various molecular species in a mixture to the derivatives of their thermodynamic properties. This is a general statistical mechanical theory of solutions, which is valid both classically and quantum mechanically and is applicable to all types of intermolecular interactions. Also, it is one of the most recognized theories of solutions that directly relate the thermodynamic quantities with the solution structure without any assumptions. K-B theory, being a powerful liquid state theory, has not received considerable attention in thermodynamic literature, as it should get. Only few examples of practical applications²⁰⁻³⁸ to aqueous binary mixtures can be found in which this theory has been to be used for aqueous-alcohol20-28 binary mixtures, aqueous solutions containing electrolytes^{29,30} and aqueous solutions containing amino acids^{31,32}. To the best of our knowledge, very few studies have been made in the literature on non-aqueous binary mixtures $33-38$. The main objective of the present study is to apply the K-B theory to binary solvent systems of varied nature in order to extract new information on the interactions between the species existing in these systems on molecular level. In this paper, the K-B theory has been extended to binary mixtures of 1, 4-dioxane with benzene, toluene, *o*-xylene, *m*-xylene, *p*-xylene and mesitylene at 298.15 K and at atmospheric pressure. Some new routes for predicting various parameters/terms involved in K-B theory have also been incorporated and effectively used. The experimental data required for the calculations have been taken from our previous studies^{39,40}. antities with the solution structure without any assumptions K-B
theory, has not received considerable attention in thermodynamic
examples of practical applications^{20,28}⁸ to aqueous binary mixtures
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types of practical applications²⁰⁻³⁸ to aqueous binary mixtures,
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2. THEORY

The solution theory proposed by Kirkwood and Buff¹⁹ contains the theory of the grand ensembles on the one hand to relate compositional fluctuations to the derivatives of the chemical potentials of the components and then relate them to the integrals of the radial distribution functions of the several type of molecular pairs of species present in the solution. The K-B theory defines thermodynamic properties of a solution over the whole concentration range using the values of G_{AB} , expressed as K-B integrals, given by ous parameters/ terms involved in K-B theory have also
experimental data required for the calculations have been
the experimental data required for the calculations have been
fluctuations to the derivatives of the chemica

$$
G_{AB} = \int_{0}^{\infty} [g_{AB}(r) - 1] 4\pi r^2 dr \tag{1}
$$

where g_{AB} (r) is the angle averaged pair correlation function. The integral extends over the whole range of intermolecular distances between the pair of molecules of species A and B. The interpretation of these parameters is best obtained by considering the product of number density, ρ and G_{AB} , *i.e.*, $\rho_A G_{AB}$ or $\rho_A[g_{AB}(r)$ – 1] $4\pi r^2 dr$, where ρ_A is the number density of the A species²⁰. The quantity G_{AB} conveys information of the average affinity of the A molecules towards B molecules and *vice-versa*. In the present study we have incorporated a new method, in the inversion procedure proposed by Ben-Naim²⁰, for the computation of partial vapour pressures of the mixtures for which these vapour pressure data are not available. The composition dependence of *GAB* values provide valuable insight into the molecular structure and nature of interactions in the multi-component liquid mixture. sed by Kirkwood and Buff¹⁹ contains the theory of the grand ensembles on
estitional fluctuations to the derivatives of the chemical potentials of the
heren to the integrals of the radial distribution functions of the sc mpositional fluctuations to the drivatives of the chemical potentials of the term to the integrals of the radial distribution functions of the several type
ties present in the solution. The K-B theory defines thermodynami

2.1 The inversion procedure to compute G_{AB} 's

The K-B equations19,20 for the binary mixture of species A and B can be written as :

$$
\eta = \rho_A + \rho_B + \rho_A \rho_B (G_{AA} + G_{BB} - 2G_{AB})
$$
\n⁽²⁾

$$
\xi = 1 + \rho_A G_{AA} + \rho_B G_{BB} + \rho_A \rho_B (G_{AA} G_{BB} - G_{AB}^2)
$$
\n(3)

where η and ζ are constants, ρ_A and $\rho_{\!B}$ are the number densities of A and B, respectively. The calculations of various terms involved in equations (2) and (3) were done by using the relations given in the literature^{20,31}. The derivatives of the chemical potentials were obtained using the vapour pressure data,

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assuming that the vapour above the mixture at room temperature as an ideal gas. Thus, for component A, we can write

$$
\mu_{\mathbf{A}} = \mu_{\mathbf{A}}^{\circ} + kT \ln P_{\mathbf{A}} \tag{4}
$$

where P_A is the partial pressure of component A over the given mixture of A and B. If x_A is the mole fraction of A in the mixture, then we get the relation

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\nure at room temperature as an ideal gas. Thus, for component
\n
$$
\mu_A = \mu_A^0 + kT \ln P_A
$$
\n(4)
\nonent A over the given mixture of A and B. If x_A is the mole
\nne relation
\n
$$
x_A \left(\frac{\partial p_A}{\partial x_A} \right) = \frac{\rho^*}{\eta}
$$
\n(5)
\n2 is calculated from the partial molar volumes, $\overline{V}_{m,A}$ and $\overline{V}_{m,B}$
\nin the mixtures.
\n
$$
+\rho_B = (x_A \overline{V}_{m,A} + x_B \overline{V}_{m,B})^{-1}
$$
\n(6)
\nmixtures are calculated from the experimental density data by

The number density, ρ^* of the mixture is calculated from the partial molar volumes, $\overline{V}_{m,A}$ and $\overline{V}_{m,B}$ of the components A and B, respectively, in the mixtures.

$$
\rho^* = \rho_A + \rho_B = (x_A \overline{V}_{m,A} + x_B \overline{V}_{m,B})^{-1}
$$
(6)

The values of $\overline{V}_{m,A}$ and $\overline{V}_{m,B}$ in the mixtures are calculated from the experimental density data by using the procedure described elsewhere⁴¹.

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the mixture at room temperature as an ideal gas. Thus, for component
 $\mu_A = \mu_A^o + kT \ln P_A$ (4)

of component A over the given mixture of A and B. If x_A is the mole

we get the relation
 $x_A \left(\frac{\partial p_A}{\partial x_A} \$ Nain and Chand
 $\mu_A = \mu_A^o + kT \ln P_A$ (4)

component A over the given mixture of A and B. If x_A is the mole

get the relation
 $x_A \left(\frac{\partial p_A}{\partial x_A} \right) = \frac{\rho^*}{\eta}$ (5)

sixture is calculated from the partial molar volumes, $\$ From equation (5) we can obtain η from the data on partial vapour pressures of either A or B in the entire composition range. The partial vapour pressures are calculated from the activity coefficients, which are related to the excess Gibbs free energy of the mixture. The excess free energy of mixtures is given as Nain and Chand

re at room temperature as an ideal gas. Thus, for component

re at room temperature as an ideal gas. Thus, for component

4)

enert A over the given mixture of A and B. If x_A is the mole

relation
 $x_A \left$ Fig. $\pi / K_A = H_A + KI \ln P_A$
 $H_A = H_A + KI \ln P_A$

The component A over the given mixture of A and B. If x_A is the mole

in we get the relation
 $x_A \left(\frac{\partial p_A}{\partial x_A} \right) = \frac{\rho^*}{\eta}$ (5)

the mixture is calculated from the partial mol

$$
G^{\mathcal{E}} = H^{\mathcal{E}} - T S^{\mathcal{E}} \tag{7}
$$

The excess enthalpies, H^E and excess entropies, $-S^E$ are calculated from the internal pressures, π _i and free volumes, V_f of the mixtures by using the modified relations from regular solution theory proposed by Hildebrand *et al.*42,43.

$$
H^{\mathcal{E}} = \pi_i V_{\mathcal{m}} - \left[x_A \pi_{i,\mathcal{A}} V_{\mathcal{m},\mathcal{A}} + x_{\mathcal{B}} \pi_{i,\mathcal{B}} V_{\mathcal{m},\mathcal{B}} \right]
$$
(8)

$$
-S^{E} = R\left[x_{A}\ln V_{f,A} + x_{B}\ln V_{f,B} - \ln V_{f,m}\right]
$$
\n(9)

where π _i of the mixtures are calculated using the thermodynamic equation of state

$$
x_{A} \left(\frac{\partial p_{A}}{\partial x_{A}} \right) = \frac{\rho^{*}}{\eta}
$$
 (5)
of the mixture is calculated from the partial molar volumes, $\overline{V}_{m,A}$ and $\overline{V}_{m,B}$
respectively, in the mixtures.
 $\rho^{*} = \rho_{A} + \rho_{B} = (x_{A} \overline{V}_{m,A} + x_{B} \overline{V}_{m,B})^{-1}$ (6)
 $\overline{V}_{m,B}$ in the mixtures are calculated from the experimental density data by
d elsewhere⁴¹.
n obtain η from the data on partial vapour pressures of either A or B in the
partial vapor pressures are calculated from the activity coefficients, which
is free energy of the mixture. The excess free energy of mixtures is given as
 $G^{E} = H^{E} - T S^{E}$ (7)
 E and excess entropies, $-S^{E}$ are calculated from the internal pressures, π_{i} and
ures by using the modified relations from regular solution theory proposed
 $H^{E} = \pi_{i} V_{m} - [x_{A} \pi_{i,A} V_{m,A} + x_{B} \pi_{i,B} V_{m,B}]$ (8)
 $-S^{E} = R [x_{A} \ln V_{f,A} + x_{B} \ln V_{f,B} - \ln V_{f,m}]$ (9)
calculated using the thermodynamic equation of state
 $\pi_{i} = (\frac{\partial E}{\partial V})_{T} = T (\frac{\partial P}{\partial T})_{V} - P = T (\frac{\alpha_{p}}{k_{T}}) - P$ (10)
ansivity of the mixture evaluated from temperature dependence of density
lation
 $\alpha_{p} = (-1/\rho)(\partial \rho / \partial T)_{p}$ (11)
the thermal pressure coefficient multiplied by absolute temperature, i.e.,
the external pressure P becomes negligible in comparison^{44,45}, therefore it

where α_p is the isobaric expansivity of the mixture evaluated from temperature dependence of density data³⁹ using the following relation

$$
\alpha_{\rm p} = (-1/\,\rho)(\partial \rho/\partial T)_{\rm p} \tag{11}
$$

he mixtures are calculated from the experimental density data by

re⁴¹.

from the data on partial vapour pressures of either A or B in the

pour pressures are calculated from the activity coefficients, which

gy of the For most of the liquids, the thermal pressure coefficient multiplied by absolute temperature, *i.e.,* $T(a_p/k_T)$ is very high so that the external pressure P becomes negligible in comparison^{44,45}, therefore it may be neglected in the equation (10) in the present calculations. The V_f of the mixtures are calculated from the relation $42,43$. pies, -5^k are calculated from the internal pressures, π_i and
modified relations from regular solution theory proposed
 $x_A \pi_{i,A} V_{m,A} + x_B \pi_{i,B} V_{m,B}$ (8)
 $V_{f,A} + x_B \ln V_{f,B} - \ln V_{f,m}$ (9)
the thermodynamic equation of stat

$$
V_{\rm f} = \frac{RT}{(P + \pi_i)}\tag{12}
$$

since *P* is very small as compared to π_i , it has been neglected in the equation (12) in the present calculations. These systems can be considered as 'regular' mixtures, as the *SE* values obtained using equation (9) for these mixtures are very low ($S^E \approx 0$), for which G^E is given by

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$$
G^{\mathcal{E}} = x_{\mathcal{A}} x_{\mathcal{B}} N w \tag{13}
$$

where *w* is a constant computed from equation (13), which may depend on temperature but is independent of composition. The activity coefficients in a regular mixture are given by

$$
\ln \gamma_i = (1 - x_i)^2 w / kT \tag{14}
$$

tegrals of 1, 4-dioxane + aromatic hydrocarbon binary mixtures
 $G^E = x_A x_B Nw$ (13)

n (13), which may depend on temperature but is independent

regular mixture are given by
 $x_i = (1-x_i)^2 w / kT$ (14)

apponents in the mixtures If integrals of 1, 4-dioxane + aromatic hydrocarbon binary mixtures
 $G^E = x_A x_B N w$ (13)

ation (13), which may depend on temperature but is independent

n a regular mixture are given by
 $\ln \gamma_i = (1 - x_i)^2 w / kT$ (14)

component The partial vapour pressures of the components in the mixtures were calculated using the activity coefficients. Using the mole fraction dependence of partial vapour pressure of either A or B, η over the entire composition range can be obtained from equation (5). The vapour pressures of the liquids have been taken from the literature⁴⁶⁻⁴⁸. The isothermal compressibility*, k_T,* is calculated using the well-known thermodynamic relation regrals of 1, 4-dioxane + aromatic hydrocarbon binary mixtures
 $G^E = x_A x_B N_W$ (13)

in (13), which may depend on temperature but is independent

regular mixture are given by
 $T_i = (1 - x_i)^2 w / kT$ (14)

ponents in the mixtures thegrals of 1, 4-dioxane + aromatic hydrocarbon binary mixtures
 $G^E = x_A x_B Nw$ (13)

on (13), which may depend on temperature but is independent

regular mixture are given by
 $\gamma_i = (1 - x_i)^2 w / kT$ (14)

edence of partial vapo rals of 1, 4-dioxane + aromatic hydrocarbon binary mixtures
 $E = x_A x_B Nw$ (13)

(13), which may depend on temperature but is independent

gular mixture are given by
 $=(1-x_i)^2 w / kT$ (14)

onents in the mixtures were calculated *C*^E = $x_A x_B Nw$ (13)
 C^E = $x_A x_B Nw$ (13)

ation (13), which may depend on temperature but is independent
 n a regular mixture are given by
 $\ln \gamma_i = (1 - x_i)^3 w / kT$ (14)

components in the mixtures were calculated usin -Buff integrals of 1, 4-dioxane + aromatic hydrocarbon binary mixtures
 $G^E = x_A x_B Nw$ (13)

equation (13), which may depend on temperature but is independent

ts in a regular mixture are given by
 $\ln \gamma_i = (1 - x_i)^2 w / kT$ (14)
 In $\gamma_{\rm f} = (1 - x_{\rm f})^2 w / kT$ (14)

ne components in the mixtures were calculated using the activity

energednese of partial vapour pressures of either A or B, n over the

ened from equation (5). The vapour pressures of th

$$
k_{\rm T} = k_{\rm s} + \frac{T V \alpha_{\rm T}^2}{C_{\rm p}} \tag{15}
$$

where k_s is isentropic compressibility and C_p is the heat capacity. The C_p values for pure liquids have been taken from literature⁴⁹ and the mixtures have been calculated by using the relation

$$
C_{\rm p} = x_{\rm A} C_{\rm p,A} + x_{\rm B} C_{\rm p,B} \tag{16}
$$

Using the values of η , ζ , $\overline{V}_{m,A}$ and $\overline{V}_{m,B}$, the G_{AB} is calculated using the relation

$$
\overline{V}_{\text{m,A}} \overline{V}_{\text{m,B}} = (\xi - \eta G_{\text{AB}}) / \eta^2
$$
\n(17)

Once G_{AB} is obtained, the values of G_{AA} and G_{BB} can be easily calculated using the relations given in the literature20,31.

Another quantity proposed by Ben-Naim, A_{AB} , which is a measure of the "degree of similarity" between the two components of the mixture²⁰, has also been calculated by using the equation given below

$$
\Delta_{AB} = G_{AA} + G_{BB} - 2G_{AB} \tag{18}
$$

The condition A_{AB} = 0 signifies symmetrical ideal solutions. The magnitude of A_{AB} can be used to specify the extent of deviation from ideal behaviour of solution.

3. RESULTS AND DISCUSSION

The values of K-B parameters for the above-mentioned binary systems have been calculated as a function of mole fraction, x_A of component A (1,4-dioxane) at 298.15 K using the above procedure. The values of various parameters of pure liquids required for the calculations are listed in Table 1. The values of K-B parameters, $viz.$, G_{AA} , G_{BB} , G_{AB} and Δ_{AB} along with some other parameters for the binary systems

| Liquid | ρ | \boldsymbol{u} | \boldsymbol{p} | $k_{\rm s}$ | $k_{\rm T}$ | $\alpha_{\rm p}$ | $C_{\rm p}$ |
|-------------|------------------|------------------|------------------------|---|--|------------------|-----------------------------|
| | $(kg \, m^{-3})$ | $(m s^{-1})$ | $(N \, \text{m}^{-2})$ | $(10^{-10} \text{ m}^2 \text{ N}^{-1})$ | $(10^{-10} \text{ m}^2 \text{ N}^{-1})$ (10^{-3} K^{-1}) | | \mod^{-1} \mathbf{J} |
| 1,4-Dioxane | 1027.95 | 1344.7 | 9288.1 | 5.380 | 6.231 | 0.706 | 149.8 |
| Benzene | 1027.95 | 1299.5 | 12685.8 | 6.778 | 10.393 | 1.362 | 136.8 |
| Toluene | 862.36 | 1307.3 | 3794.5 | 6.785 | 9.209 | 1.094 | 157.3 |
| o -Xylene | 871.54 | 1347.5 | 880.0 | 6.290 | 8.130 | 0.973 | 186.1 |
| m -Xylene | 881.00 | 1323.6 | 1113.7 | 6.637 | 8.635 | 0.997 | 183.0 |
| p -Xylene | 891.55 | 1307.4 | 1186.7 | 6.828 | 8.925 | 1.015 | 181.5 |
| Mesitylene | 902.55 | 1339.8 | 322.3 | 6.467 | 8.108 | 0.909 | 209.3 |

Table 1. Values of ρ , u, p, ks, k_p , α_p , and C_p for pure liquids at 298.15 K used in the calculation of K-B integrals.

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investigated as function of mole fraction, $x_{\rm A}$ of 1,4-dioxane for these mixtures are listed in Table 2. The variations of G_{AA} , G_{BB} , G_{AB} and Δ_{AB} against mole fraction, x_A of 1,4-dioxane for each mixture are shown graphically in Figs. 1-6. The low values excess entropy (S $\texttt{E}\approx\,$ 0) obtained for these systems (Table 2) clearly indicate that these mixtures can be considered as regular solutions ⁴³.

Table 2. Values of ρ , u, ρ^* , H^E , TS^E , $\ln(P_A)$, η , G_{AB} , G_{AA} , G_{BB} and Δ_{AB} for 1,4-dioxane + aromatic hydrocarbon binary mixtures at 298.15 K

| x_A | ρ (kg m^{-3}) (m s ⁻¹) (10 ⁴ m ⁻³)(J mol ⁻¹)(J mol ⁻¹) | \boldsymbol{u} | ρ^* | $H^{\rm E}$ | $TS^{\rm E}$ | $ln(P_A)$ | η (10^4 m^{-3}) | G_{AB} | G_{AA} | $G_{\rm BB}$ $(10^{-4} \text{ m}^3 \text{ mol}^{-1})$ | \mathbf{A}_{AB} |
|--------|--|------------------|----------|-------------|--------------------------|-----------|-----------------------------------|----------|----------|--|----------------------------|
| | | | | | | | | | | | |
| | | | | | $1,4-Dioxane + benzene$ | | | | | | |
| 0.0713 | 884.38 | 1304.0 1.1220 | | 498.9 | -38.7 | 6.237 | 2.6755 | -2.044 | -0.708 | 14.039 | 17.419 |
| 0.1403 | 894.85 | 1308.1 | 1.1254 | 929.2 | -72.4 | 6.934 | 1.4863 | -1.136 | -0.796 | 0.578 | 2.054 |
| 0.2123 | 905.82 | 1312.5 | 1.1290 | 1332.1 | -104.3 | 7.368 | 1.0872 | -0.831 | -0.863 | -1.124 | -0.324 |
| 0.2809 | 916.30 | 1316.5 | 1.1324 | 1656.0 | -130.3 | 7.667 | 0.9141 | -0.699 | -0.924 | -1.384 | -0.909 |
| 0.3528 | 927.31 | 1320.5 | 1.1359 | 1927.1 | -152.6 | 7.915 | 0.8245 | -0.631 | -0.989 | -1.365 | -1.092 |
| 0.4195 | 937.55 | 1324.0 | 1.1391 | 2109.2 | -168.2 | 8.107 | 0.7901 | -0.604 | -1.049 | -1.281 | -1.122 |
| 0.5037 | 950.51 | 1328.2 | 1.1432 | 2238.0 | -180.3 | 8.313 | 0.7976 | -0.610 | -1.117 | -1.161 | -1.057 |
| 0.5800 | 962.30 | 1332.0 | 1.1469 | 2256.7 | -183.8 | 8.476 | 0.8560 | -0.655 | -1.144 | -1.056 | -0.890 |
| 0.6504 | 973.21 | 1335.1 | 1.1502 | 2168.2 | -178.7 | 8.610 | 0.9740 | -0.745 | -1.069 | -0.966 | -0.545 |
| 0.7197 | 983.99 | 1337.5 | 1.1535 | 1964.7 | -164.2 | 8.730 | 1.2059 | -0.922 | -0.686 | -0.882 | 0.276 |
| 0.7822 | 993.74 1339.8 | | 1.1565 | 1703.7 | -144.4 | 8.831 | 1.6607 | -1.270 | 0.643 | -0.802 | 2.381 |
| 0.8561 | 1005.30 1342.0 1.1600 | | | 1267.6 | -109.4 | 8.941 | 3.6479 | -2.789 | 10.716 | -0.648 | 15.646 |
| | | | | | 1,4-Dioxane + toluene | | | | | | |
| 0.0688 | 862.36 | 1308.4 0.9487 | | 144.8 | -15.0 | 6.201 | 2.3260 | -2.133 | -0.452 | 13.129 | 16.943 |
| 0.1373 | | 871.54 1309.5 | 0.9619 | 267.6 | -28.5 | 6.911 | 1.2863 | -1.179 | -0.727 | -0.399 | 1.231 |
| 0.2112 | 881.00 | 1310.9 | 0.9766 | 383.3 | -41.6 | 7.362 | 0.9386 | -0.860 | -0.856 | -1.808 | -0.945 |
| 0.2858 | 891.55 | 1312.4 | 0.9920 | 474.8 | -52.6 | 7.686 | 0.7884 | -0.722 | -0.944 | -1.867 | -1.367 |
| 0.3552 | 902.55 | 1313.9 | 1.0067 | 536.4 | -60.7 | 7.923 | 0.7241 | -0.663 | -1.011 | -1.725 | -1.411 |
| 0.4361 | 913.11 | 1315.9 | 1.0244 | 582.0 | -67.4 | 8.150 | 0.7022 | -0.643 | -1.075 | -1.535 | -1.324 |
| 0.5064 | 925.82 | 1317.9 | 1.0403 | 597.6 | -70.8 | 8.319 | 0.7206 | -0.660 | -1.107 | -1.386 | -1.174 |
| 0.5819 | 937.25 | 1320.4 | 1.0579 | 589.3 | -71.5 | 8.479 | 0.7838 | -0.718 | -1.085 | -1.251 | -0.901 |
| 0.6506 | 949.92 | 1323.2 | 1.0744 | 563.8 | -69.6 | 8.610 | 0.8994 | -0.823 | -0.927 | -1.152 | -0.432 |
| 0.7212 | 961.80 | 1326.3 | 1.0920 | 505.0 | -63.8 | 8.733 | 1.1302 | -1.034 | -0.353 | -1.075 | 0.641 |
| 0.7903 | 974.37 | 1329.8 | 1.1097 | 420.7 | -54.4 | 8.843 | 1.6448 | -1.505 | 1.746 | -1.038 | 3.718 |
| 0.8640 | 987.06 | 1334.2 | 1.1291 | 301.9 | -40.1 | 8.953 | 3.8978 | -3.566 | 17.020 | -1.167 | 22.986 |
| | | | | | $1,4-Dioxane + o-xylene$ | | | | | | |
| 0.0672 | 882.50 | 1341.5 | 0.8408 | -123.7 | 5.8 | 6.177 | 2.0854 | -2.189 | -0.239 | 12.057 | 16.195 |
| 0.1391 | 890.29 | 1335.1 | 0.8589 | -266.4 | 13.3 | 6.925 | 1.1259 | -1.180 | -0.686 | -1.405 | 0.270 |
| 0.2103 | 898.42 | 1329.6 | 0.8776 | -379.7 | 19.6 | 7.358 | 0.8391 | -0.879 | -0.853 | -2.372 | -1.468 |
| 0.2853 | 907.43 | 1324.8 | 0.8983 | -467.6 | 24.9 | 7.684 | 0.7100 | -0.743 | -0.958 | -2.257 | -1.730 |
| 0.3543 | 916.23 | 1321.0 | 0.9183 | -535.8 | 29.4 | 7.920 | 0.6579 | -0.688 | -1.027 | -2.020 | -1.672 |
| 0.4293 | 926.36 | 1317.9 | 0.9413 | -582.9 | 32.9 | 8.133 | 0.6446 | -0.673 | -1.077 | -1.776 | -1.507 |
| | | | | | | | | | | | |

Conted........

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Figure 1(a) indicates the behavior of 1,4-dioxane-1,4-dioxane affinity over the entire composition range. Table 2 and Fig. 1(a) indicate that G_{AA} values are negative and decreases initially with increase in mole fraction till $x_A \approx 0.58$ and then increases after this mole fraction and finally become positive, suggesting decreasing correlation between 1,4-dioxane molecules and this correlation increases sharply after $x_A \approx$ 0.72. Fig. 1(b) indicates that G_{BB} values are positive and decrease sharply with increase in mole fraction till $x_A \approx 0.21$, become negative after this concentration, and then increases slightly after this point, suggesting decreasing correlation between benzene molecules on increasing the amount of 1,4-dioxane in the mixture, which leads to disruption of the orientational order 50 present in benzene. Fig. 1(c) indicates that that G_{AB} values are negative and increase initially and exhibit a maximum at $x_A \approx 0.58$ and then decrease with increase in mole fraction of 1,4-dioxane, suggesting increasing correlation between 1,4 dioxane and benzene molecules due to the electron donor-acceptor type interactions⁵¹ between electronegative oxygen atoms of 1,4-dioxane (as donor) and the π -electrons of ring of benzene molecules (as acceptor) at near equimolar concentration range $0.3 < x_A < 0.72$. Further, on increasing the amount of 1,4-dioxane in the mixture after $x_A \approx 0.72$, G_{AB} then decreases sharply with further increase in mole fraction of 1,4-dioxane. This behaviour is in agreement with the conclusions drawn in our earlier study^{39,40}, wherein $V_{\rm m}^{\rm E}$ and $\kappa_{\rm s}^{\rm E}$ vs. *x*_A curves exhibited a minimum at *x*_A \approx 0.55, indicating maximum interaction between 1,4-dioxane and benzene molecules around this composition. Figure 1(d) indicates that Δ_{AB} values are positive and decrease initially to negative values exhibiting a minimum at $x_A \approx 0.35$ and then again increase to positive values, indicating the dissimilarity between unlike molecules in the mixtures and negative deviations from the ideal behaviour²⁰.

Fig. 1. Plots of $G_{AA'}G_{BB'}G_{AB'}$ and Δ_{AB} against mole fraction, x_A of 1,4-dioxane for 1,4-dioxane + benzene mixtures at 298.15 K.

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Figures 2-6 indicate that similar trends in G_{AA} , G_{BB} , G_{AB} and Δ_{AB} values (as above for 1,4-dioxane + benzene) against mole fraction, *x*¹ are exhibited by 1,4-dioxane + toluene/*o*-xylene/*m*-xylene/*p*-xylene/ mesitylene mixtures also due to same reasons as discussed above for 1,4-dioxane + benzene mixtures. A close perusal of Table 2 indicates that the magnitude of G_{AB} varies in the order: benzene > toluene > *p*xylene > *m*-xylene > *o*-xylene > mesitylene. This suggests that the affinity or interactions between the 1,4 dioxane and these aromatic hydrocarbon molecules follows the order: benzene > toluene > *p*-xylene > *m* xylene > *o-*xylene > mesitylene. This may be due to the fact that the number of $\rm CH_{3}$ groups substituted on the ring increase from benzene (with no substituted $\rm CH_{3}$ group) to mesitylene (with three substituted CH₃ groups). Because methyl group (CH₃) being an electron-releasing group, would enhance the electron density of the benzene ring of the aromatic molecules, thereby decreasing the electron-accepting tendency of the aromatic ring, as we move from benzene to mesitylene, resulting in decreased donor-acceptor interaction between unlike molecules with increase in number of methyl group ($\rm CH_{3})$ in the aromatic hydrocarbon molecule. The results and conclusions from K-B integrals are in good agreement with the conclusions drawn regarding the intermolecular interactions from the trends exhibited by the excess functions of these mixtures^{39,40}.

Fig. 2. Plots of $G_{AA'}G_{BB'}G_{AB'}$ and Δ_{AB} against mole fraction, x_A of 1,4-dioxane for $1,4$ -dioxane + toluene mixtures at 298.15 K.

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Fig. 3. Plots of $G_{AA'}G_{BB'}G_{AB'}$ and Δ_{AB} against mole fraction, x_A of 1,4-dioxane for 1,4-dioxane + *o*-xylene mixtures at 298.15 K.

Fig. 4. Plots of $G_{AA'}G_{BB'}G_{AB'}$ and Δ_{AB} against mole fraction, x_A of 1,4-dioxane for 1,4-dioxane for 1,4-dioxane + *m*-xylene mixtures at 298.15 K.

Fig. 5. Plots of $G_{AA'}G_{BB'}G_{AB'}$ and Δ_{AB} against mole fraction, x_A of 1,4-dioxane for 1,4-dioxane + *p*-xylene mixtures at 298.15 K.

1,4-dioxane + mesitylene mixtures at 298.15 K.

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