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Utilisation of High Energy Propellant Waste in Manufacturing of Fired Clay Bricks to Enhance the Acoustic Properties

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ABSTRACT

The disposal and waste management of solid high energy propellant (HEP) is a considerate conservational problem. HEP waste is currently disposed in open or confined burning which may cause environmental hazards. In this paper, we examined and discussed results on recycling of HEP waste into fired clay bricks baked in different orientation. HEP modified bricks with 1.5%, 3% and 5 wt. % HEP waste content were manufactured and tested, and then compared against virgin clay bricks without HEP content. The effect of directional orientation of bricks baked with varying HEP content on acoustic properties were experimented and discussed. The sound transmission loss decreases with increase in HEP waste due to formation of independently closed directional pores. The transmission loss of horizontally baked during firing of bricks is nearly 5dB lower than vertically baked bricks. Results of the experimental studies indicate that HEP waste can be utilised in fired clay bricks and different orientation baking further enhances the acoustic properties.

Keywords: Waste management; High energy propellant; Bricks; Sound transmission loss

NOMENCLATURE

HTPB	Polybutadiene hydroxyl terminated binder
AP	Ammonium perchlorate
XRD	X-ray diffractometer
SEM	Scanning electron microscopy

1. INTRODUCTION

Solid rocket propellant comprising ammonium perchlorate (oxidizer), poly-butadiene (binder) and aluminium (fuel) are extensively used in propulsion systems of missiles and launch vehicles. They offer superior performance and energy level and strain capability compare to conventional propellants. Composite propellants of nearly few thousand tonnes per annum are manufactured for various missile and space programs in India by various Government agencies. Considering a conservative evaluation, nearly 15% wastage is assumed during production which is nearly 150 tons of propellant waste. The propellant waste includes in-process waste, left-out and end-trimmed portion of propellants, rejected and unutilised propellant lots. The amount of life expired propellant in India is estimated to be around 2500 tonnes. Cost of the estimated composite propellant is ₹ 3000/kg and energy content of composite propellant is above 1 kCal/g. A huge amount of cost and energy is wasted because of unutilised and rejected lots of propellants. As the generation of waste is inevitable and as the mandate for a clean, safe environment grows, the clash of these two conflicts must be dealt with best recycling methodology. Reduction of propellant waste and better waste management will certainly remain a top priority for defence and space agencies. The emphasis of the study is on solid propellants cast in missile and rocket motors. To compound the problem as time passes by, they get expired and beneficial for intended use. The researchers focussed on formulating new ways in utilising and recycling the waste in various systems.

Currently, these composite propellant wastes are disposed by open or confined burning[1]. The issues concerned with current disposal methods of composite propellant are (i) environmental and safety hazards, (ii) production and waste disposal cost, (iii) time consuming, (iv) logistics and legal hurdles, (v) pits and earthen damage and (v) inherent loss of propellant energy. Various waste discarding practices like air and salt oxidation, biodegradation, alkaline pressure hydrolysis and biochemical process[2-4] were discussed as alternatives for open burning method. Conversion of propellant waste in to liquid fertilizer has little succeeded due to agricultural benefits[5]. All these methods are purely depended on energetic material composition and requires huge infrastructure to treat them chemically and convert into usable products. And the same chemical/biological method will not be available commonly for waste disposal of all energetic materials. Hence, alternative disposal method is needed for waste management of high energy materials.

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The disposal process needs to be safe, technically feasible, low cost and common for all energetic materials. The competent use of HEP waste for industrious purpose and consumption in bricks processing is the efficient waste management methodology for the solid propellant waste. This helps in enhancement of functional properties of the bricks.

Fired clay bricks are generally used as a construction material because of their exceptional properties. The composition of material, firing temperature and added additives determine the functional parameters of bricks[6]. Currently, incorporating waste as building material is getting importance and benefiting construction sector by improving the functional properties. Recycling of various waste materials like corn cub, rice husk, wine waste, glass powder, marble powder, shea waste, fly ash, ciggerate buds, granite waste, sugarcane baggage[7-15] were utilised in manufacturing of bricks. Each additive has its own process and functional advantages like light weight, increased water absorption and energy efficient. Nowadays lot of importance has been given for energy and sound efficient buildings. Pores of micro level are generated in the bricks using various industrial and bio-waste additives. The best ways are improving the thermal and acoustic performance of the bricks by creating optimal level of porosity with appropriate waste additives in manufacturing of bricks. Recycling of waste in clay bricks not only has functional advantages but also performance enhancement is observed in certain cases.

As compared to conventional pores, materials with long aligned pores exhibit special features like light weight, sound absorption and vibration damping. Sound absorption studies were carried out for porous materials with directional pores and its effect on functional properties[16]. Similar study on acoustic properties and its effect on directional orientation of bricks during firing were reported in this paper.

Recently, reducing the energy consumption during the firing of bricks by adding high calorific energy waste is explored by few researchers[13]. However, during firing of bricks over long period it will still cause environmental problems, which can be restricted to some extent by reducing the firing time with addition of high calorific energy waste. Studies on varying the firing temperature and its role on functional properties were carried out earlier[17]. Whereas reusing of high energy propellant waste in brick manufacturing were not studied. It has massive benefit of reducing the energy required for firing of bricks. Currently energy efficacy is considered along with cost and quality as important parameter in brick manufacturing. The amount of energy generated by adding HEP waste (1 wt%) in bricks is between 150-200 kJ per brick and the energy required for baking a virgin brick is of 2 to 3 MJ per brick and varies with baking method. Utilising this recycling methodology of HEP waste in baking of bricks will minimise the required firing energy and result in homogeneous baking, and mechanically more sound bricks[18].

The core intent of the study is to utilise recycling of high energy propellant waste in processing of bricks. Acoustic properties of bricks baked in different concentration and different thickness were compared with standard bricks. Studies on effect of directional orientation of bricks on acoustic properties during firing were discussed broadly.

2. MATERIALS AND METHODS

2.1 Processing of Bricks

Standard composite propellant consists of hydroxyl terminated polybutadiene (HTPB) – ammonium perchlorate (AP) – aluminium (Al) metal powder collected as process waste, from DRDO propellant processing centre, Nashik, India. Propellant waste was obtained during process and rejection lots and grinded in the form of dust powder. The average particle size of the propellant waste powder is found to be in the range of 200-250 µm. The soil used in the study was collected from brick kilns, Nasik, India and their characteristics are given in the reference[19].

The complete work flow diagram is shown in Fig. 1. Three batches for preparation of bricks were prepared using different quantity of high energy propellant waste in each. These three mixes were categorised by the amount of HEP waste weight percentage and named as 1.5 HEP, 3 HEP and 5 HEP. The quantity of different ingredients in the mix are soil ~60 %, fly ash ~26%, HEP waste ~4 wt% and water ~ 10%. The homogeneous mixing of soil, fly ash, HEP waste, and water were carried out in sigma blade mixer with single blade. The duration of mixing is 80 min and speed is 60 rpm. The mixing process was carried out initially with soil, fly ash and water for 20 min and subsequently after weathering, HEP waste added and mixing was carried out for 60 min. The mixed slurry was used in moulding of bricks in to different shapes and sizes as shown in Fig. 2: 52×25×20 mm (rectangular blocks) for compressive test studies, and 30 × 5 mm, 30 × 10 mm, 30 × 15 mm and 30 × 20 mm (30 mm diameter cylindrical blocks with different height of 5, 10, 15 and 20 mm) for acoustic studies. The bricks were compacted manually for optimum density using the moulds with desired quantity of masses as per the standards. The moulded bricks were initially heated in oven for

Figure 1. Propellant recycling in processing of bricks.

Figure 2. Fired clay brick samples.

48 h at 100 °C and subsequently heated at around 400 °C for moisture removal and propellant decomposition, respectively. The preheated brick samples were fired in electric furnace between 1000-1100 °C at the rate of 6 °C/min and dwelled for few hours. Experiments were conducted by placing the brick samples horizontally and vertically in the furnace during firing, for angling the pores in horizontal and vertical direction.

2.2 Combustion Products of High Energy Propellant Waste used in Bricks

Studying the thermal decomposition and combustion characteristics of energetic materials is a big challenge and it depends on pressure and temperature sensitivities of the burning rate, propellant surface condition, energy and temperature distribution. It is a physiochemical process results from the ingredient reactants present in the propellant. The propellant waste used here is composite propellant with polybutadiene hydroxyl terminated binder (HTPB), ammonium perchlorate (AP) and aluminium. The composition of combustion products is determined using thermochemical calculation using EXPLO5 Software. The isobaric combustion products composition is given in Table 1.

In the current experiment in batch making of bricks, the propellant waste is added in small quantity. Whereas the firing of bricks is a slow process, where bricks are initially slow heated at 100 °C for 48 h and subsequently heated up to 400 °C slowly for few hours for decomposition of propellant. The combustion process involved here is altogether different than open burning/rocket propulsion. The environmental hazards of high energy propellant waste combustion products are studied earlier by few researchers[20-24].

2.2 Investigation

Mineralogical phases of fired clay bricks were studied using an X-ray diffractometer (Model, Bruker D8 Advance) with for brick samples with various quantities of waste additives. XRD peaks were recorded in the range of 20˚ to 70˚ at a step size of

Table 1. Composition of major combustion products of High energy propellant waste

Products	mol%	mass %
H_2	32.406	2.561
CO	25.324	27.806
HCl	12.618	18.03
H_2O	8.562	6.04
Al_20_3	7.916	31.64
N_2	7.368	8.09
H	3.051	0.12
AlCl	0.788	1.93
CO_2	0.788	1.35
Cl	0.755	1.05
$AlCl_2$	0.225	0.86

0.05˚. Archimedes method was used to determine the amount of porosity. Porosity, pore distribution, and pore size were quantified using scanning electron microscopy (SEM) images. For every sample batch, five samples were used for image analysis to obtain average porosity values. The morphology, microstructure, and pore size distribution were studied by SEM (Model, Zeiss, Merlin). The compressive strength was determined from the known values of fracture load (N) and brick area (mm^2).

The sound transmission loss is an important parameter for designing sound absorbing porous materials for civil sector. Impedance tube experimental setup is vital for investigation of acoustic transmission loss for relatively smaller specimen. The experimental apparatus includes long rigid metallic tube, microphones, Gaussian white noise source, variable gain power amplifier, speaker and data acquisition system. The working principle of the impedance tube is based on transfer function method[25]. The acoustic measurements were carried out in 30 mm tube made of heavy steel frame to minimise the vibrations. The tube can be divided in two segments: a loud speaker and a sound termination segment. Specimen with a diameter ~30 mm under investigation is gripped between two segments (Fig. 3). Each section has provision of two microphones which can be flush mounted before measurement of transmission loss. Edges are tightly wrapped with adhesives to lessen the transmission loss between the gaps. The impedance tube was designed for 1000 - 5000 Hz with a spectral resolution of 2 Hz (BSWA Tech). Sound source of class II calibrator at standard frequency and power level of 1000 Hz and 114 dB, respectively were used for calibration of microphones prior to each experiment.

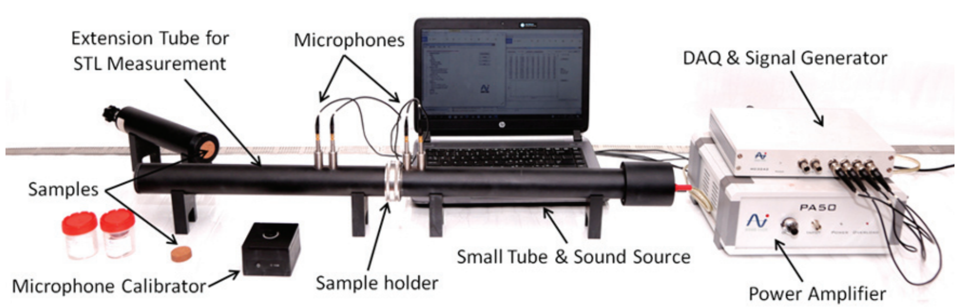


Figure 3. Impedance tube - Acoustic measurement setup.

The sensitivity values of installed microphones were in the range of 53 mV/Pa and 63 mV/Pa. Background disturbances were also verified by switching off the acoustic source. An acoustic driver was used to excite the acoustic source and the mounted microphones recorded the sound spectrum on incident and transmitted face of brick samples. Measured background noise was 65 - 70 dB at the test place and amplifier gain was tuned between 90 and 100 dB to attain a better signal to noise ratio (SNR). Because of the sound pressure, the excitation levels in acoustic tube studies are in the limit. Sound transmission loss of the two varieties of brick samples, one fired keeping horizontally the samples and other fired positioning vertically the samples in the furnace. Transmission loss results with varying HEP waste and sample thickness were studied in detail.

3. RESULTS AND DISCUSSION

The important properties of waste propellant dust powder and soil are listed in Table 2. The compressive strength, water absorption and acoustic properties of bricks are tabulated in Table 3.

Compressive strength and water absorption studies were carried out in rectangular blocks as per the standards and the same had been reported in our earlier work[18].

The distinct mineralogical content of the processed HEP modified bricks samples after firing are carefully indexed during XRD analysis (Fig. 4). The XRD patterns of 1.5 %, 3 % and 5% HEP waste modified bricks are shown in Fig. 4. The studies confirm that XRD peaks of all the HEP waste modified bricks are similar and no secondary phases are observed. Presence of hematite material is identified by the formation of red colour in bricks. The analysis indicates the existence of clay minerals (quartz, hematite, feldspar, and mullite) and confirms the incidence of vitrification process during firing. The presence of mullite peaks indicates the sound densification of bricks occurred during firing.

Figures 5 (a-d) shows the microscopic images of horizontally and vertically baked propellant modified bricks respectively, fired in both horizontal and vertical orientation. The fractured surface morphology of the horizontally and vertically fired bricks is discussed. It is very evident the porosity is created by addition of HEP waste. The samples show a comparatively dense structure with small irregular and spherical pores (10 – 50 µm). In horizontally baked propellant bricks, higher porosity content is observed in horizontal direction compare to vertical direction. Whereas, in vertically placed and baked sample the porosity content is higher in vertical direction. It shows the direction of baking of bricks plays a dominant role in creation and orientation of porosity using HEP waste. Segregation and aggregation of pores are observed in horizontal and vertically baked samples, respectively in principal axis direction of baking.

The development of advanced bricks with high sound transmission loss is desirable and it's conceivable by introducing porosity in the bricks. The increase in the variation of HEP waste increases porosity. The sound transmission loss of porous bricks is associated with attenuation of sound energy over intricate HEP modified brick material structure.

Table 2. Soil and Propellant waste powder physical properties

Soil Physical properties	Values	Propellant physical properties	Values
Average particle size, µm	~ 25	Density, g/cc	1.7-1.8
Water plasticity, %	20-22	Elongation, %	40
Water absorption content, %	19	Specific impulse, s	245
Max. dry density, kgm^{-3}	1950	Calorific value, cal./g	1100-1200

Table 3. Compressive strength, Water absorption and Sound transmission loss of bricks

HEP waste	Compressive strength, N/mm^2	Water absorption (%)	Sound transmission loss, dB (at 1.6 kHz)
0 %	4.3	22.7	60
1.5 %	4.8	20.8	65
3.0 %	4.0	22.4	61
5.0 %	4.2	20.2	54

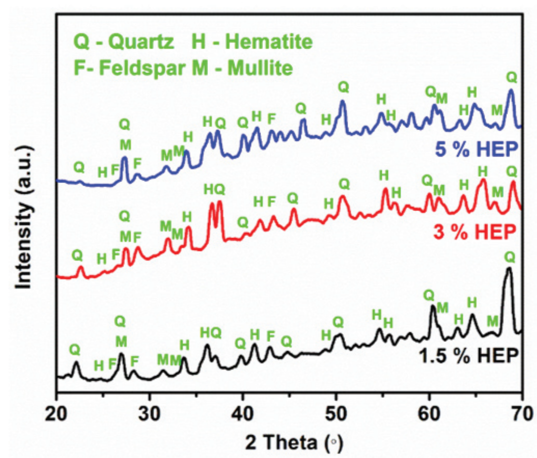


Figure 4. XRD studies on variation of HEP waste in bricks.

The effect of variation of HEP waste on sound transmission loss of horizontally and vertically oriented and baked bricks with the uniform thickness and frequency is studied in Fig. 6. Whereas, increase in amount of HEP waste decreases sound transmission loss and the same has been observed in both horizontally and vertically baked samples. The results indicate the sturdy dependency of sound transmission loss on porosity of the brick samples. Sound transmission in porous bricks mainly gets affected due to diminution of sound energy by viscous resistance of air during sound propagation in the bricks. With increase in HEP waste, the coefficient of resonant sound absorption also increases and that may also be attributed to decrease in the sound transmission loss[26-27]. The increase of HEP waste introduces numerous independent closed pores and that results in decrease of sound transmission loss.

Sound transmission loss studies of horizontally and vertically baked bricks with the uniform thickness and frequency is studied as shown in Fig. 7. It is observed that transmission loss of vertically baked bricks increase compared to that of horizontally baked ones. The same behaviour has been seen in different amount of propellant modified bricks.

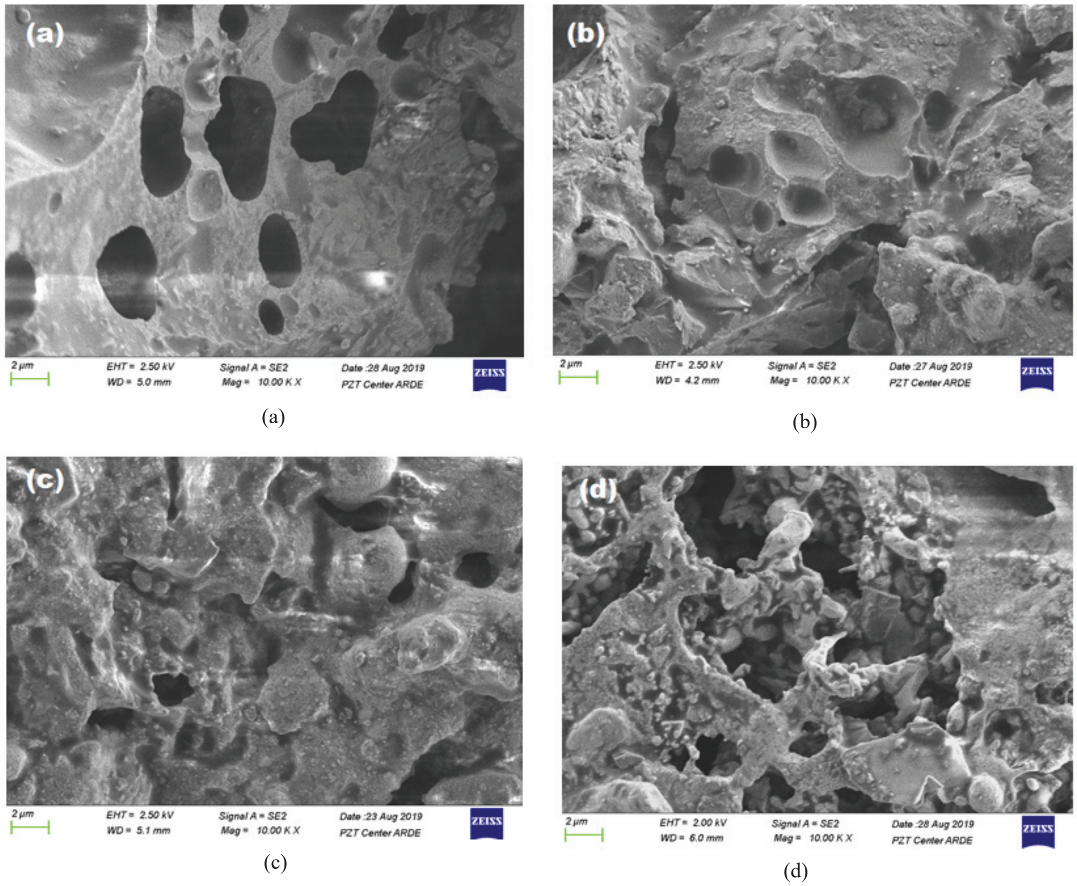


Figure 5. (a-b) SEM of Horizontally baked [horizontal (a) and vertically (b) oriented images] and (c-d) Vertically baked [horizontal (c) and vertically (d) oriented images] propellant modified bricks.

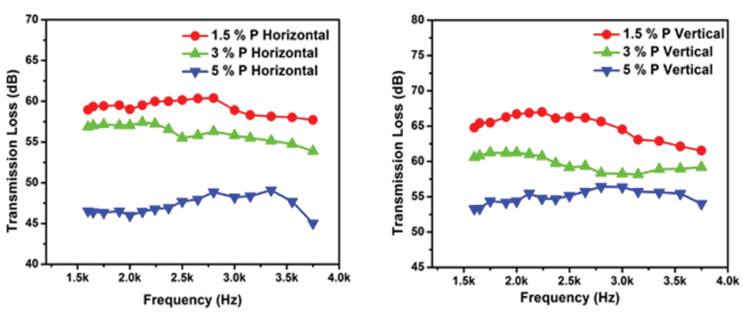


Figure 6. Variation of Sound transmission loss with frequency for horizontally and vertically baked samples.

Transmission loss of porous bricks increases significantly with presence of pores because of multiple scattering and diffraction of sound waves occurs at air-brick interface[28]. The sound transmission loss is related to incident angle of sound on porous surface. In vertically baked bricks, the direction of incident sound wave and pore orientation are parallel and during the transmission of sound waves it may come across many pore scatterers and interface boundaries which may reduce the sound transmission significantly. Whereas, in horizontally baked bricks, the sound waves and pores are not parallel oriented and that may not affect sound transmission compared to vertically baked ones. It

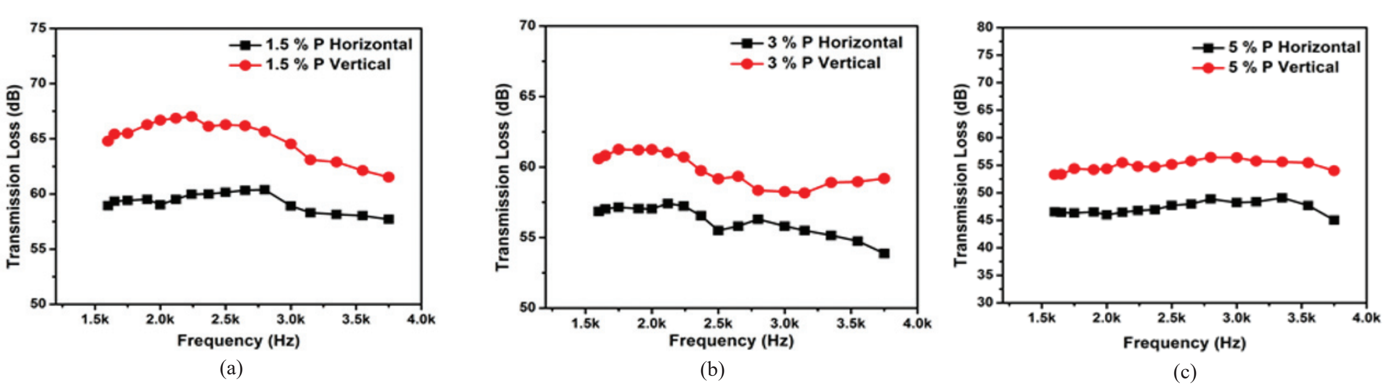
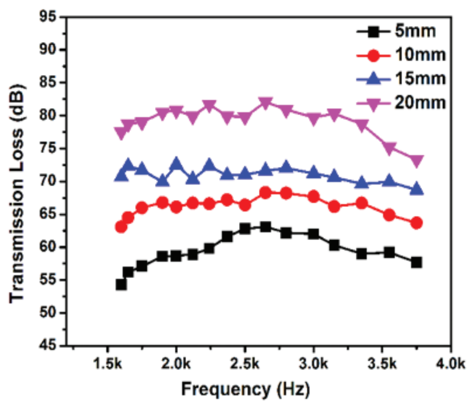


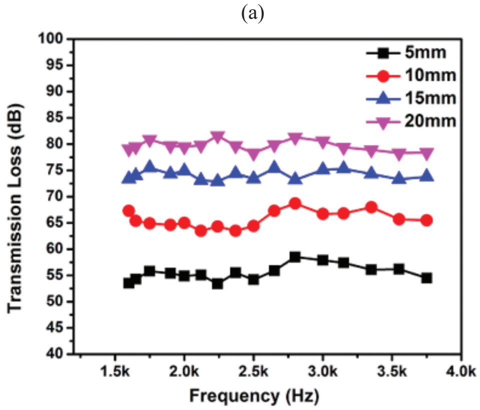
Figure 7. Sound transmission loss comparison of horizontally and vertically baked samples with frequency (a) 1.5 %, (b) 3 %, and (c) 5%.

may also be attributed to domination of viscous forces over the inertial forces in the low frequency range and sound propagation direction.

Sound transmission loss studies of horizontally and vertically baked bricks in the low frequency range with varying thickness are shown in Fig. 8. Sound transmission loss increases with increase in thickness of the bricks and the same has been observed for all horizontally and vertically baked samples. It is well known that low frequency sound wave means higher wavelength, and the transmission loss will be higher due to intensified acoustic energy absorption with raise in thickness of the bricks. The increment in acoustic damping with thickness of bricks is due to viscous inertial damping mechanism of double porosity materials. The obtained average sound transmission loss is equally comparable with reported literature[29-31].



(a)



(b)

Figure 8. Thickness dependent Sound transmission loss studies for (a) horizontally and (b) vertically baked 1.5 % HEP samples.

4. CONCLUSIONS

This paper discusses about the addition of HEP propellant waste in processing of fireclay bricks for recycling of propellant waste in brick industries. The key benefits obtained from incorporating the optimal propellant waste in bricks is light weight, porosity, and increase in compressive strength and sound transmission loss.

(a) Utilising the HEP waste, it is feasible to fabricate light weight bricks which are preferable for loadbearing walls. The obtained sound transmission loss improves sound insulation of bricks and considered to be best for construction of internal walls.

(b) Presence of high calorific value in propellant waste, the firing energy required for firing of bricks reduced significantly. With addition of 1.5 wt % of HEP, a large quantity of propellant waste can be recycled. Also, the fuel requirement of firing of bricks can be trimmed down nearly 10 %.

(c) The microstructure study clearly brings out the effect of direction of firing of bricks and the same had been observed in pore distribution.

(d) In vertically fired bricks, the direction of incident sound wave and pore orientation are parallel and during the transmission of sound waves the sound transmission loss increases significantly due to pore scattering and interface boundaries.

Summarising the above studies, it is proposed that 1 to 1.5 wt % HEP waste can be utilised in brick manufacturing industry and by orientation of bricks during firing changes the pore structure and its orientation. The environmental effects of the current disposal technology are not confirmed. This can be utilised for enhancing the sound insulated properties of bricks used for dividing walls or partitions as per the BIS standard specification (clause 6.2.6).

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