

Study on High Energy Propellant Waste in the Processing of Fired Clay Bricks

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ABSTRACT

Utilisation of propellant waste in fabrication of bricks is not only used as efficient waste disposal method but also to get better functional properties. In the present study, high energy propellant (HEP) waste additive mixed with soil and fly ash in different proportions during manufacturing of bricks has been investigated experimentally. X-ray diffraction (XRD) studies were carried out to confirm the brick formation and the effect of HEP waste. Ceramic bricks were fabricated with HEP waste additive in proper proportions i.e. 0.5 wt %, 1.0 wt %, 1.5 wt %, 2.0 wt %, 2.5 wt %, 3 wt %, 3.5 wt %, and 4 wt % and then evaluated for water absorption capability and compressive strength. Compressive strength of 6.7 N/mm², and Water absorption of 22 % have been observed from modified fired bricks impregnated with HEM waste additive. Scanning electron microscopy (SEM) studies were carried out to analyze the effect of HEP waste additive on pore formation and distribution in the bricks. Further, the heat resulting from decomposition of propellants can cause a decrease in the energy required of baking process. The process of manufacturing of bricks with HEP waste additive is first of its kind till date.

Keywords: Clay bricks; High energy propellant; Structural Materials; Additives; Water absorption; Waste disposal; Compressive strength

1. INTRODUCTION

Clay bricks are one of the most important building elements used for civil construction in large quantities. History of fired clay bricks starts from hand making to the bull trenched kilns, and batch to the continuous viable process of brick manufacturing¹. Even though, manufacturing of bricks with commonly used materials like soil and fly ash is economical and feasible, functional property enhancements possible through waste additives, which will otherwise cause environmental hazards in disposal activities must be exploited to the full extent. Literature review shows that many researchers studied the organic and inorganic additives like polymer materials, wine waste, glass waste, rice husk, sugarcane bagasses, wheat straw, corn cob, cotton husk, groundnut shell, coal waste, olive mill waste, shea waste, foundry sand, ceramics and paper waste²⁻²⁰ were added as additives in the processing of bricks. Effect of these additives on water absorption, mechanical and thermal properties were also studied and discussed by many researchers. However, the recent research areas are focused on additives for reduction in energy consumption during the processing of bricks and energy efficient buildings. Usually additives used in the bricks burn out during firing process and create porosity to regulate the moisture and water absorption content²¹.

The materials which releases high pressure and temperature on decomposition of high velocity gaseous products in short time either by detonation or deflagration are called as high energy materials. Numerous researchers across the globe have been studying the waste disposal methods of high energy materials²²⁻²³ with minimum energy and impact on environment. High energy propellants (HEP) are one of the categories of high energy materials. HEP wastes are generally disposed in large quantities in open area and ignited from a remote site. The current disposal method is not environmental friendly and also hazardous for human health²⁴. Different methods have been studied for disposal such as incineration, molten salt and wet air oxidation, biodegradation and chemical process²⁵⁻²⁶. Disposal of HEP waste is a challenging task. Some of the problems associated with disposal of high energy materials waste include infertile land, safety hazards, air quality degradation and toxic combustion products. Large quantity (in tonnes) of HEP waste is created during the processing of large rocket mortars, lot rejection and expiry of service life. The processing waste can be efficiently used for productive purpose rather than disposing it off resulting in severe environmental issues, huge transportation and handling costs²⁷. The process propellant waste generated is of around 150 to 200 tonnes at different propellant processing centres of DRDO, ISRO and industries. Though 0.5 % of HEP waste can be disposed in the proposed process, considering the extensive requirement of

bricks in country, substantial portion of waste propellant (few tonnes) can be disposed off in the process of manufacturing of bricks.

The key open burning method used currently for waste disposal of propellant is not environmental friendly and safe. There remains a huge demand for ease waste disposal of propellant waste. Very limited work had been carried out in waste disposal and life cycle management of propellant. The waste propellant act as combustible ingredient by providing required energy for firing of bricks and also act as pore cavity forming agent in the bricks, which controls the density and weight of the bricks. Use of HEP waste in manufacturing of bricks can be one of the novel waste disposal techniques of high energy materials. The addition of high energy combustible material may lead to localized and uniform heating of bricks. This may lead to reduction in weight of bricks, optimum water absorption and improvement in mechanical properties. Current study leads to eco-friendly disposal of combustible material on one hand and enhancing the functional properties of the bricks on the other hand.

Current study is concentrated on use of HEP waste as partial substitute to soil and fly ash in manufacturing of bricks. In addition, microstructure, water absorption and compressive strength characteristics of the modified bricks with different proportions of HEP waste additive. XRD and SEM techniques were used to examine the phase formation microstructure of the bricks and to optimize the quantity of HEP waste in order to improve the functional performance of bricks.

2. MATERIALS AND METHODS

2.1 Materials and Sample Preparation

The HEP waste is standard composite propellant consists of hydroxyl terminated poly butadiene, HTPB (10-15%), ammonium perchlorate, AP (60-70 %) and aluminium powder, Al (15-20 %) obtained from process waste was used in dust form. Propellant dust was generated during the machining stage of the propellant grains and remote grinding of propellant waste obtained through the process and rejection lots (Fig. 1.). The source of HEP is from DRDO propellant processing centre, located at Nasik. The characteristics of composite propellant are (i) calorific value: 1000-1200 cal/g; (ii) Density: 1.7-1.8 g/cc; (iii) % Elongation: 40%, (iv) Specific impulse: ~ 245 s. The particle size distribution



Figure 1. Propellant dust powder.

is measured using Mi scattering theory by laser diffraction method using Horiba, LA 960 laser particle size analyzer. The d_{50} of the particle size is 145 μm and the particle size distribution varies from 22 μm to 320 μm . The soil used in the manufacturing of bricks was taken from brick kilns, Nasik, India. The characterization of clay is given in this reference under soils developed on interfluvies under sub-humid zone in Nasik²⁸. The mix proportion of various materials in the bricks is soil ~ 60 %, fly ash ~ 26%, HEP waste ~ 4 % and water ~ 10 %. Soil quantity varies according to the addition of propellant.

Sigma blade mixer with single blade is used for homogeneous mixing of the propellant, soil and fly ash. 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 soil dough with fly ash was blended with propellant dust (0 to 4 wt %) and distilled water (20 to 25 %) to obtain desired level of plasticity. Homogeneity of mix is ensured with the milling cycle to avoid localization of propellant dust powder. The achieved level of water plasticity obtained in this soil dough is in the range of 22 to 24 and the same is obtained through Atterberg plastic limit. The HEP waste content added in the samples was 0 to 4% in steps of 0.5% by wt. designates as 0B, 0.5B, 1B, 1.5B, 2B, 2.5B, 3B, 3.5B and 4B, respectively (Fig. 2). Batch of 10 samples were processed at each propellant composition. The bricks were then moulded in test brick moulds of approximate size 52x25x20 mm (Fig. 3). The brick samples were oven dried for 48 h at 100 °C to remove excess moisture and avoid cracks during firing. The propellant composition decomposes at around 350 °C to 400 °C. The dried samples were then heated slowly in electric furnace at 1100-1150 °C and dwelled for 6 h and cooled down to ambient temperature.



Figure 2. Different batches of bricks with HEP waste bricks.

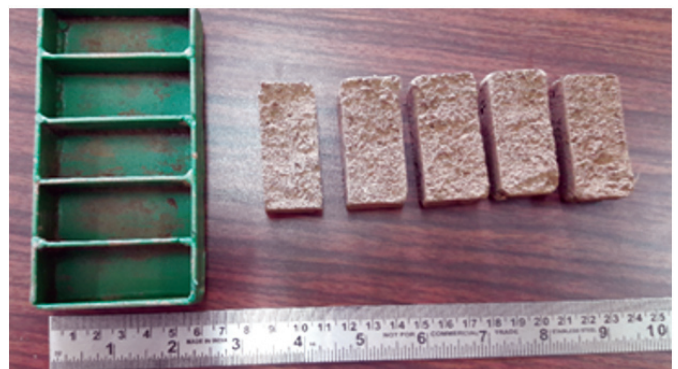


Figure 3. Moulds and green bricks.

2.2 Analysis

The structural phases in the bricks were investigated using XRD (Model, Bruker D8 Advance) with copper k_{α} radiation for samples fired with and without propellant additives. Samples were scanned at an angle from 20° to 70° at a scanning step of 0.05° . The porosity was calculated from apparent density values measured by Archimedes method. Water absorption test was carried out to obtain the durability of bricks during weathering conditions. The bricks were heated at 110°C in oven to remove the moisture content, cooled down to room temperature and then its weight (m_1) was measured. Subsequently, the samples were immersed in clean water at room temperature for 24 h. The samples were then removed from water and traces of water in the samples were wiped off using dry cloth and then its weight (m_2) was measured. The water absorption of samples was calculated as:

$$\text{Water absorption (W \%)} = \frac{(m_2 - m_1)}{m_1} \times 100 \quad (1)$$

The compressive strength of the brick samples was calculated by dividing the maximum load at failure (N) to average area of the brick samples (mm^2). In the current studies, the porosity is measured by using the formula:

$$\text{Porosity (P)} = \frac{(1 - \rho)}{\rho_0} \times 100 \text{ (\%)} \quad (2)$$

ρ_0 is bulk density of the brick and ρ is the density of propellant modified porous brick²⁹. Also the amount of porosity is verified by image analysis of scanning electron microscopy images. The pore sizes are measured directly from scanning electron images using image analysis software by linear intercept method. This method is normally used for grain size measurement and the same is used for pore size measurement. For every percentage of HEP waste, 4 samples are tested for SEM studies and pore size measurements. The densification, microstructure, pore size and porosity of the samples were investigated by scanning electron microscope (Zeiss, Merlin).

3. RESULTS AND DISCUSSION

XRD patterns of brick samples with 4 % HEP waste and without HEP waste fired at 1100°C are shown in Fig. 4. The XRD patterns reveal that both the fired bricks are almost similar in the structure except some extra peaks of alumina observed in bricks with HEP waste. The alumina peaks may be due to presence of aluminum particles in HEP waste, which may get oxidized at higher temperature to form alumina. It is also confirmed that addition of HEM waste not resulted in any of the additional phases. It is observed that Quartz, hematite, feldspar and mullite minerals are present in the fired bricks and their corresponding patterns are identified. The red color of the fired bricks may be due to the presence of hematite mineral. The presence of other minerals confirms that occurrence of vitrification process during firing which is responsible for densification of the bricks⁴.

SEM images of fired samples with variation of HEP waste (O B, 0.5 B, 1 B, 1.5 B, 2 B, 2.5 B, 3 B, 3.5 B and 4 B) are shown in Fig. 5. The surface morphology of the fractured surface of the fired bricks is studied in detail. It is very evident

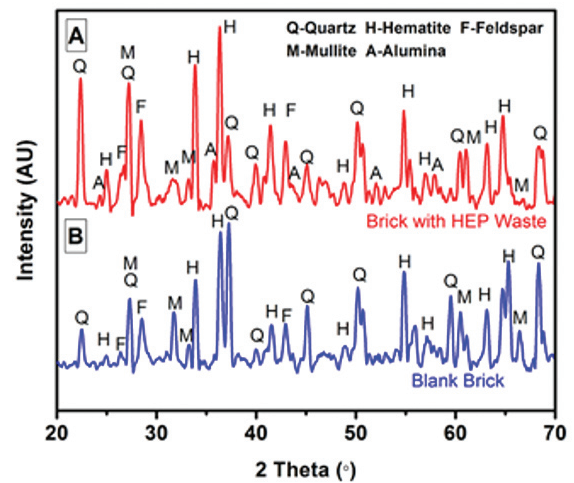


Figure 4. XRD patterns of fired bricks (a) with 4 % HEP waste and (b) without HEP waste (blank).

that addition of HEP waste additives acted as pore forming agents and created porosity in the samples. Increase in HEM waste additives increases the porosity in the fabricated fired bricks. Higher amount of porosity is observed in the 3.5B and 4B samples, due to addition of larger quantity of HEP waste additives. It is also important to note that the dense microstructure of bricks in the area other than porosity. The pores are uniformly dispersed indicating the homogeneity of the mixing and wide particle size distribution of HEP waste. The pore size is observed to be varying from 1 to $50\ \mu\text{m}$ in the modified fired bricks. The presence of low porosity in the bricks, due to small amount ($< 2\%$) of HEM waste additives as seen in Figs. 5 (a) - 5(c) contributed to increased compressive strength. On the other hand, the presence of high porosity due to higher amount of waste additives ($> 3\%$) as seen in Figs. 5(g) - 5(i) resulted in increase in amount of water absorption. However, with the presence of higher porosity, the modified fired bricks are light weight, sturdy and ease to handle. The SEM images may also be used as a quantitative tool to observe the porosity distribution.

The brick is proportionate mixture of clay, sand and high energy propellant waste. With sufficient quantity of water, the brick is moulded and dried with low shrinkage, cracks and warping. During firing, it enables silica to fuse and to bond the solid particles by neck formation. The firing temperature plays vital role in determining compressive strength and water absorption efficiency, because it controls porosity in the bricks. The heat evolution from the propellant will enhance the firing process of bricks by uniform heating.

The test results of water absorption of bricks with HEP waste additives are presented in Fig. 6. Initially with addition of 0.5% HEP waste, the rate of water absorption of bricks decreases drastically. This could be attributed to increase in densification due to in-situ thermal energy generated and addition of HEP waste. However the porosity generated is low and no formation of interconnecting pore channels for capillary action of water. Increase in the addition of HEP waste additives leaves increased pores during firing, causing increased porosity and allows the formation of pore interconnecting channels for capillary action

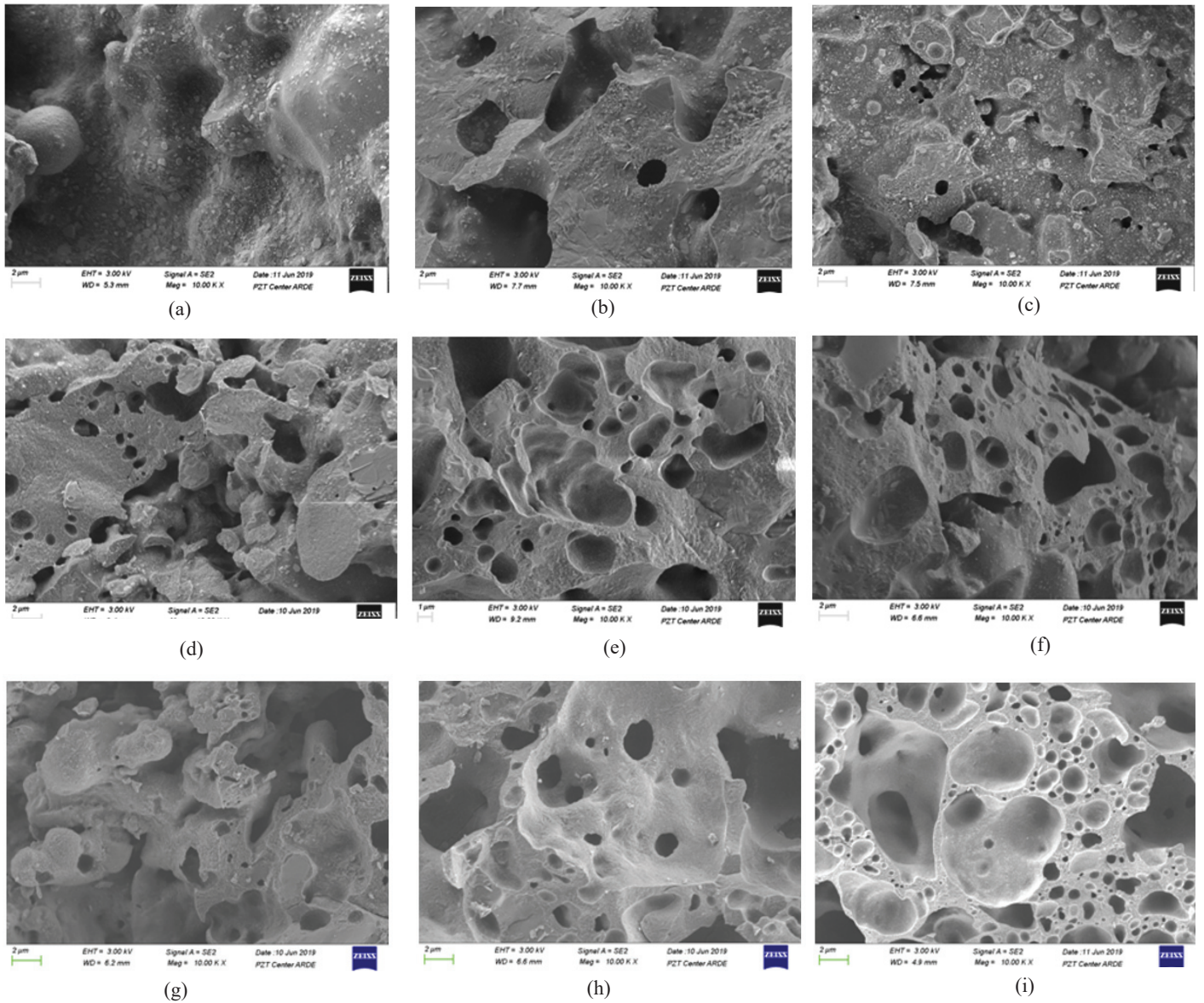


Figure 5. SEM images of fired brick samples: (a) 0 B, (b) 0.5 B, (c) 1 B, (d) 1.5 B, (e) 2 B, (f) 2.5 B, (g) 3 B, (h) 3.5 B and (i) 4 B.

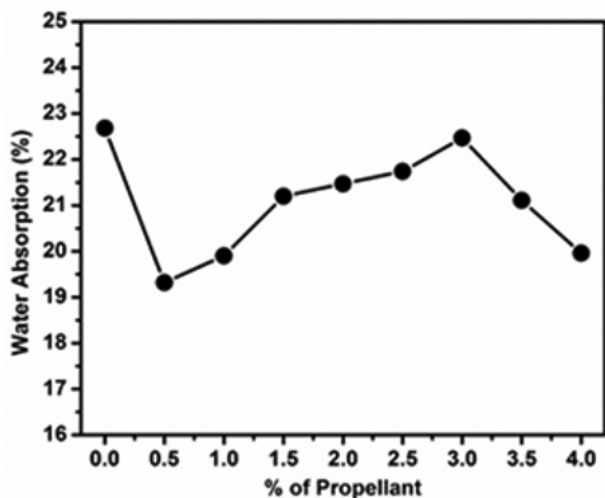


Figure 6. Effect of HEP waste additives on water absorption of bricks.

which in turn increases the rate of water absorption of bricks. It is well known that the porosity dominates over the densification behaviour of the bricks. Further increase in HEP waste additive dominates the densification behaviour which in turn reduces the rate of water absorption. With above 3 % usage of HEP, the densification dominates the porosity behaviour because the pores generated interior is not connected by capillarity with surface pores. Hence the water connecting channels are disengaged and that leads to decrease in water absorption. The rate of water absorption significantly depends on (i) nature of additive, (ii) interaction of additives with sand particles, (iii) thermal properties of the additives and (iv) affinity to moisture absorption. However the present study reveal that lower HEP waste additive up to 1 %, is more beneficial due to reduced water absorption capacity and classified under moderate weather resistant bricks¹.

The test results of compressive strength of the bricks with varying HEP waste additives are shown in Fig. 7. Compressive

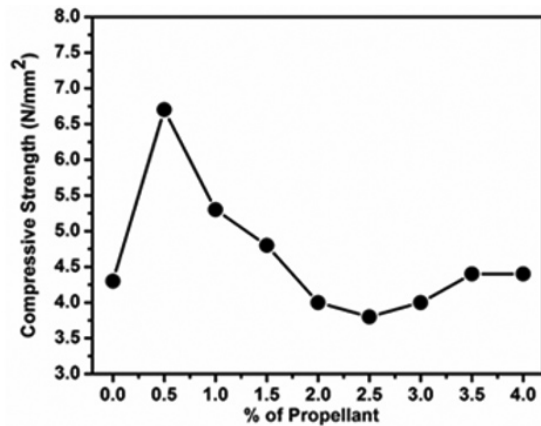


Figure 7. Effect of HEP waste additives on compressive strength of bricks.

strength usually depends on porosity, pore size, pore distribution and secondary phases in the material. Initially compressive strength increases with addition of 0.5% HEP waste, and then it decreases with further addition of HEP waste. The increase may be due to alumina dispersion strengthening of bricks, which is present in the HEP waste modified bricks after firing. The dispersed alumina in the bricks seizes micro crack formation and prevents the crack to propagate till failure. The water absorbing efficiency of bricks is not directly related to compressive strength; however its pore structure and porosity may play a vital role. Wagh³⁰, *et al.* proposed a model to study the dependency of modulus with porosity. It is assumed in the model, ceramics are continuous network of solid cylinders with porous channels between them.

$$E = E_0(1 - P)^m \quad (3)$$

where E is Young's modulus of bricks, and E_0 is Young's modulus of free pore matrix, P is porosity and m is the function of characteristics of cylinder.

It is clearly depicted from the above Eqn. (3), lower the porosity in the bricks, higher the compressive strength and reduction in efficiency of water absorption.

Similarly, lower the addition of propellant in the bricks (0.5 wt %) results in lower porosity, and increase in compressive strength due to packing efficiency of the particles. Higher the propellant content, the packing efficiency will get disturbed because during the process of heating the bricks, the gases present in the interior of the initially open pores widen and generate large voluminous gaseous products. The packing nature of cylindrical particles is basically different from spheres because the former exhibit orientation freedom, and contains flat, curved surfaces and corners.

However, with further increase in HEP waste, the porosity dominates the behaviour of the bricks. The presence of porosity also affects the cohesion and dispersion strengthening effect of alumina which in turn lowers the resistance of brick material to failure. It can be attributed to the stress concentration arising at open pores, which may lead to loss of structural integrity and affects compressive strength. It may be noted that compressive strength values are exactly varying opposite to rate of water absorption in HEP waste modified bricks.

4. CONCLUSIONS

HEP waste as additive in manufacturing of bricks has been experimentally investigated for the first time and results in terms of functional properties were discussed in detail. The results presented in this article have been verified for compliance of all test standards and are industry scale by brick industry. From the present study, the following conclusions may be drawn;

- Utilization of HEP waste as additives in manufacturing of bricks can be considered as one of the waste disposal methods and subsequently used as ecological structure in the buildings for creation of wealth out of waste.
- XRD studies revealed the presence of various minerals and alumina peaks in the bricks modified by HEP waste. This will validate that no secondary phases were present, which can cause damage to the bricks.
- The microscopic study revealed the formation and uniform dispersion of pores in the modified bricks with HEP waste additive.
- Water absorption test indicated that, the rate of absorption decreased drastically with small amount of HEP waste additive, and further increase in additives increased porosity that in-turn increased the rate of water absorption. The optimum quantity of HEP waste will result in lower density bricks.
- From the compressive test results, it is observed that, small amount of HEP waste additive improves the compressive strength due to alumina dispersion strengthening in the bricks.
- The bricks sintered with HEP in small amount of weight percentage make lighter bricks with improved compressive strength. The reduction in weight of bricks without sacrificing compressive strength will have cascading effect in bringing down the cost of civil engineering structures using this bricks as basic building blocks.

BIS specification of average compressive strength of bricks of 'class 5' to be minimum 5 N/mm² and water absorption not to exceed 20 %. The final properties of HEP (0.5 %) added bricks are meeting the BIS specifications for 'Class 5' bricks. Total energy generated by HEP is of nearly 100 kJ per brick whereas energy consumed in firing of a blank brick is of 2MJ per brick. The process is energy intensive and addition of HEP will trim down the quantity of fuel nearly 5 %. Though the HEP % is small but the volume of total number of bricks baked in very large quantity hence the entire process will be more energy efficient and correspondingly cost effective. HEP being process waste will be available with bare minimum cost and it will contribute towards the total energy required for firing the bricks. Based on the above observations, it may be concluded that less than 1% of HEP waste additives is beneficial to obtain favourable physical and mechanical properties of bricks. And also process is considered as one of the favourable waste disposal method of HEP waste.

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In the present work he has helped in formatting the manuscript and guided the author.

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In the present work he has carried out characterization of the bricks and formatted the manuscript.