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Influence of Additives on Thermal Expansion of Silica in Sand Casting Applications

V N Sai Deepak Kolli [a, *], Taishi Matsushita [a], Jessica Elfsberg [b], Attila Dioszegi [a]
[a] Jönköping University, Sweden
[b] Scania CV AB, Sweden
* Corresponding author: E-mail address: deepak.kolli@ju.se

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Abstract

Silica sand is extensively used as a moulding sand for both mould and core making in the cast iron and steel casting industries. The pouring temperatures of cast iron and steel create a nonlinear distribution of temperatures across mould/core. The temperature fluctuations in mould/core establish different heat transport zones and result in a temperature-dependent undesirable expansion of silica. The expansion of silica is one of the primary sources for the formation of surface defects on castings. Additives are incorporated to mitigate the volumetric expansion of mould/core resulting from the granular expansion of silica sand. The paper aims to investigate the thermal dilation of unbonded silica sand integrated with different amounts of additive in the sand (0.8%, 1.0%, and 1.3%) using a horizontal dilatometer. The dilatometric investigations identified a decreasing trend in the thermal expansion behaviour of silica mixture with increasing content of additive inclusions in the mixtures. In theory, the additives in the sand mixtures decompose prior to the α↔β endothermic phase transition of quartz and provide intergranular spacing for the free expansion of silica. DSC and TGA were conducted to identify the phase change in silica and the degradation temperature of the additives, respectively.

Keywords: Silica sand, Phase Transformation, Thermal expansion, Additives, Cast iron, Component casting

1. Introduction

Sand casting is a traditional metal manufacturing process widely practised in cast-iron foundries. The use of sand cores in production is increasing with the demand to fabricate complex geometries. The production of the cores/mould in this process includes moulding sand and a binding agent to hold the granular sand together. Silica sand is the widely used raw material in the fabrication of cores and moulds for sand casting within cast iron foundries. The usage of silica is attributed to its abundant availability, capacity for high sintering temperatures, and economic viability. However, foundries face critical quality issues generating different surface defects on the cast components due to inadequate knowledge of moulding materials.

The thermophysical and chemical properties of moulding sand significantly influence the solidification process and casting skin formation in cast iron. These properties of moulding sand vary relatively with an increase in the temperature of the silica sand during the casting process. On comparing the expansion characteristics of different moulding sands, silica sand expands relatively higher than any other available aggregate sand used in the casting process [1, 2]. The heat transfer due to temperature variation between molten metal and bonded sand (core/mould) induces uneven temperature gradients across the mould/core from the metal mould interface. The varying temperature gradient in the

material results in irregular expansion of the silica granules in the cores/moulds. The cause for this non-uniform expansion of the granules is the polymorphic behaviour of silica sand, which undergoes a phase transition during the temperature rise in sand grains [3]. Elevated temperatures in the core or mould degrade the binding bridges between the sand grains and the granules are susceptible to thermal expansion. The granular expansion of sand is attributed to the temperature driven reversible transformation of silica from α quartz ↔ β quartz [4]. The expansion of silica results in the formation of casting defects such as veining, dimensional inaccuracy of the casting, non-metallic inclusions, metal penetration, rat tails, and scabs [5]. Formation of these unwanted surface irregularities and defects on the cast component reduces the mechanical properties of the product.

The granular expansion of sand is undesirable and needs to be controlled to obtain a better casting product. There are various methods currently in practice across foundries to counter sand expansion to minimize sand expansion. Few foundries attempted to stabilize the tridymite phase by adding aluminium, lithium, and sodium to minimize the expansion effect of silica [6]. High-temperature binders reduce the risk of forming expansion defects and increase the core strength [7], but adding a high amount of binders leads to the generation of problems related to gas evolution in the mould/core. Bašistová et al. [8] have made sand mixtures with different compositions of Cerabeads as additives. Increasing the mountt of Cerabeads in sand reduces the overall grain size and increases the fineness of the material. Irrespective of the composition of the additives, quartz transition was observed in all the samples. Vasková et al. [9] conducted a study on two different additives, one is an organic-based additive and the other is red iron oxide. These two additives increased the strength of the core and the bonding activity between the grains, thus reducing the effect of sand dilatation on the casting surface. Hrubovčáková et al. [10] attempted to counter the nonlinear expansion of silica using a blend of different additives. The casting trials displayed defect-free castings when an iron oxide additive was used along with a carbon-based reducing agent. Iron oxide in the moulding aggregate reduces the softening temperature of silica, which helps to minimize the thermal tension on the surface of sand grains. However, there is an inadequate explanation regarding the mechanism that resulted in defect free cast components with the usage of additional materials. The current study aimed to investigate the thermophysical properties of a sand mixture containing additives produced using a blend of biodegradable material with iron oxide to reduce sand expansion defects formed on castings.

2. Experimental Analysis

2.1. Materials

Silica sand, characterized by SiO_2 purity ranging from 88-92%, serves as the subject of examination for thermal expansion phenomena. The investigated sand variant was extracted from Jönköping, Sweden, and used in the cast iron foundry. The sand displays an average grain size of 0.28 mm with AFS GFN 53 is determined through sieve analysis.

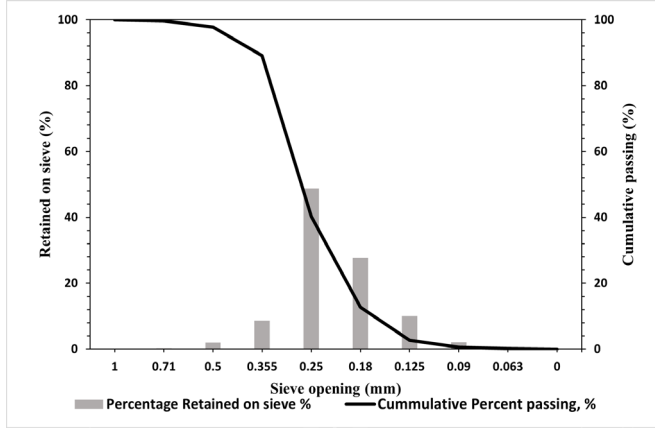

Fig. 1. Grain size distribution of investigated silica sand

A commercially utilized additive with a concentration of iron oxide and substantial proportions of biodegradable particles such as starch and wood flour (containing cellulose) are introduced to the sand mixture to study the expansion characteristics. Due to the physical nature of the additives which is a fine powder structure the incorporation of additives into the mixture is facile. Nominal composition of additives is provided in Table 1 below. However, due to the heterogeneity of the elements in the additives, the precise chemistry of the additives is unknown.

Table 1.
Composition of additives used in the sand mixtures

Compound	Percentage (%)
Starch	50-80
Iron oxide	10-25
Wood dust	1-5
Quartz	<1

2.2. Measurement Methods

The specimens under examination for thermal expansion investigations were unbonded (indicating no use of binding agents). Sand and additives in four different ratios Table 2 were thoroughly homogenized using a laboratory-grade sand mixer in a controlled laboratory environment. Subsequently, the samples were prepared for dilatometric and calorimetric analyses.

Table 2.
Sand with different compositions of additives

Silica sand mixture	Additive (%)
Mixture 1	0
Mixture 2	0.8
Mixture 3	1.0
Mixture 4	1.3

2.2.1. Dilatometric Measurement

The alumina (Al_2O_3) cylindrical crucible sample holder illustrated in Figure 2 was employed for the unbonded sand expansion measurements using a Netzsch horizontal dilatometer DIL 402 C setup. The specific sample preparation method was

originally devised by Svidró et al. [4], exhibits the capacity to accommodate granular samples with an approximate length of 8 mm sealed in the cylindrical crucible with the help of two knobs on either side of the sand sample (as indicated in Figure 2) and possesses a bulk density of $1.47 \pm 0.03 \times 10^{-3}$ g/mm^3. All the samples are prepared similarly to ensure identical packing volumes of the material in the crucible. The sample crucible facing knob 2 is held against a fixed end of the dilatometer through the application of a certain force from another end of the sample holder, with knob 1 connected to the push rod. Subsequently, the furnace is subjected to a steady increase in temperature, with a consistent heating rate of 10 °C/min in an inert atmosphere (Helium gas) to avoid reactive agents in the testing environment. The measurement is performed on the alumina standard to calculate the correction in the measurement system.

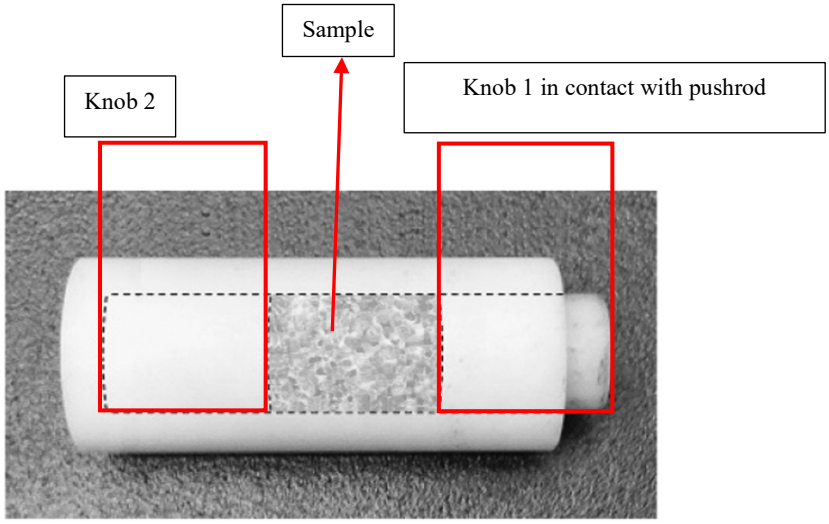

Fig. 2. Sample holder for dilatometer

2.2.2. Calorimetric Measurement

For the determination of the specific heat capacity of the samples, the differential scanning calorimeter Netzsch DSC 404 C is employed. The instrument directly records the DSC signal, and the specific heat capacity of samples is computed using the c_p ratio method using NETZSCH Proteus thermal analysis software. This methodology involves the acquisition of baseline (to calibrate the equipment and avoid distorted signals during the actual measurement), standard (using a sapphire sample), and measurements from different sand samples, using identical experimental procedures for all the measurements. The experimental procedure is conducted under controlled conditions within an inert argon atmosphere, employing a programmed heating rate of 10 °C/min.

2.2.3. Thermogravimetric Measurement

Thermogravimetric analysis is conducted exclusively on additives, which have been introduced as a composite in moulding aggregate for core/mould making. Shimadzu TGA-50 equipment is used for the evaluation of weight loss of the material as a function of temperature or time. The measurement reveals vaporization, decomposition, dehydration, and any other temperature-dependent material characteristics related to the weight loss of the material. The weight loss of the sample is assessed by a precision weight balance system situated within a furnace. The sample weighing approximately about 20 mg or less is placed on a weight balance holder enclosed in a furnace. The heating process is carried out in an ambient atmosphere (air), with the sample subjected to a constant heating rate of 10 °C/min.

3. Results and Discussion

3.1. Mixture with no Additives

The expansion characteristics of non-bonded sand material are characterized by the utilization of a dilatometer. The thermal expansion of the sample $\left(\frac{dL}{L}\%\right)$ is plotted as a function of temperature and the expansion behaviour of pure silica sand is displayed in Figure 3. To ensure a robust representation of the entire mixture of aggregates, a minimum of three sample measurements are acquired for each of the material compositions mentioned in Table 2.

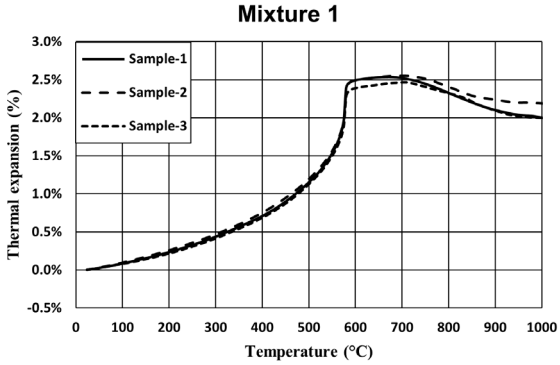

Fig. 3. Thermal expansion of sand mixture with raw sand and 0% additives

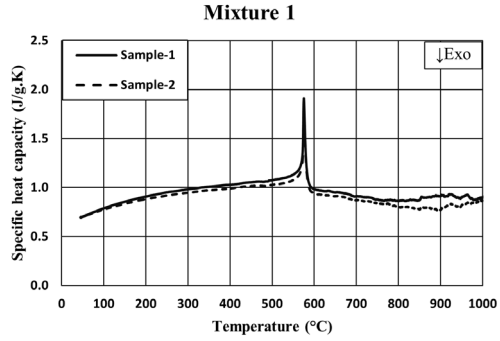

Fig. 4. Specific heat capacity of sand mixture with raw sand and 0% additives

Silica sand mixture 1, which contains no trace of additive, shows a monotonical expansion up to ca. 600 °C (570 °C), and the steep change is observed between 550 °C (570 °C) and 600 °C which is due to the polymorphic transformation of silica from α-quartz to β-quartz (Figure 3). The polymorphic modification of tetrahedral structure (α-quartz) to low-dense hexagonal structure (β-quartz) results in volumetric expansion of the granules. Moreover, the allotropic formation of β-quartz is a heat-absorbing reaction and is coherent from the endothermic peak of DSC measurement in Figure 4 at ca. 573 °C. On further heating of β-quartz, it transforms to β-tridymite at around 870 °C. However, such transformation is not distinguishable in the present DIL and DSC results, as this transformation (β-quartz→β-tridymite) is purely dependent on the purity of silica sand (more precisely presence of alkali elements in the sand). In the absence of granular impurities (alkali elements), the high quartz transforms to cubic structured β-cristobalite. The decrease in the expansion curve following the quartz transition can be from the formation of high-temperature silica allotropes and resulting in the uneven density distribution of the silica granules across the measured sample. Thereby the granular particles used for dilatometric measurements reorganize within the cylindrical crucible (sample holder).

3.2. Mixture with Additives

The rapid change in the specific density of sand Mixture 1 resulting from the crystallographic expansion of silica with temperature change needs to be controlled to mitigate the surface defects formation on the castings. Foundries utilize various types of additives as raw materials for the minimization of silica expansion. In the present study, a commercially used additive material, which primarily consists of starch, Fe_2O_3 (iron oxide), and wood flour (cellulose) is introduced along with other raw materials to minimize the non-directional expansion of granular silica. The expansion curves obtained from the dilatometric analysis of the optimized sand mixture with varying amounts of additives content as mentioned in Table 2 are illustrated in Figures 5, 6 and 7.

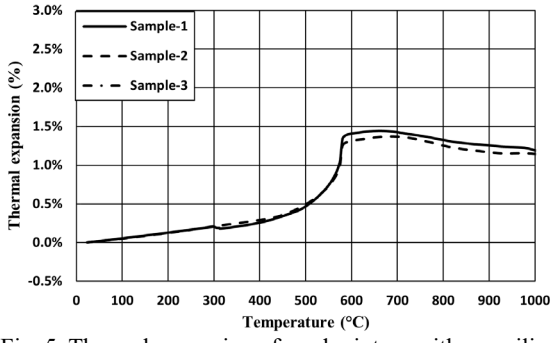

Fig. 5. Thermal expansion of sand mixture with raw silica sand and 0.8% additives

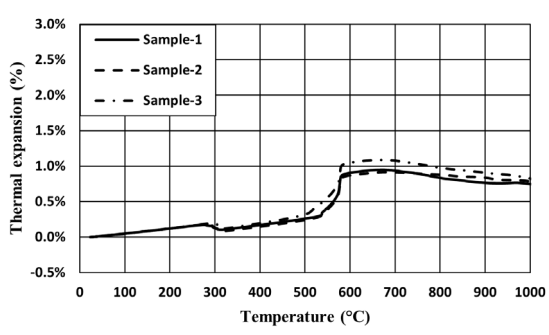

Fig. 6. Thermal expansion of sand mixture with raw silica sand and 1.0% additives

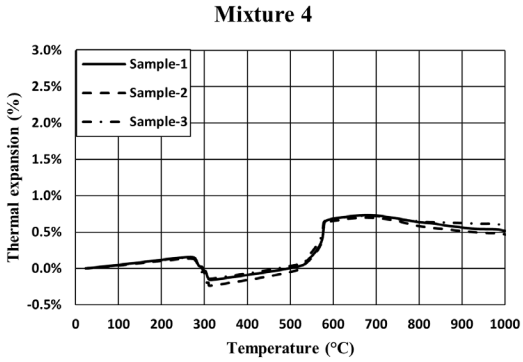

Fig. 7. Thermal expansion of sand mixture with raw silica sand and 1.3% additives

Table 3.
Maximum thermal expansion of silica sand with various percentages of additives in the sand mixtures

Sand Mixture	Additive, (%)	Maximum thermal expansion, (%)
Mixture 1	0	2.5±0.04
Mixture 2	0.8	1.4±0.04
Mixture 3	1	1.0±0.09
Mixture 4	1.3	0.7±0.01

Introducing 0.8% of the additive into the sand resulted in a significant reduction in the maximum thermal expansion of the sand mixture of about 1.1% with a standard deviation of 0.04% relative to the sand containing no additive (Figure 5). Thus, the addition of 0.8% additive decreases the peak expansion by 44% when compared with the expansion of additive-free samples. Besides the decrease in average peak expansion, a noticeable deviation is observed in the expansion curve at ca. 300 °C. Increasing the additive content in the sand mixture further minimized the peak expansion, and the average peak thermal expansion is given in Table 3 (see Figures 5, 6 and 7). In the case of a 1% addition of additives, the peak expansion reduced to 0.96% and subsequently to 0.7% in the case of a 1.3% addition of additives (Figures 6 and 7). Thus, the average peak expansion of the samples is reduced by 62% and 71% with 1% and 1.3% addition of additives, respectively, when compared to the maximum expansion of sand mixture without additive content. The supplement in the sand mixtures created a compaction trend in the thermal expansion curves of the sand samples with additive inclusions at around ca. 300 °C. The negative expansion detected in a linear expansion of sand mixtures with additives is proportional to the amount of additive in the mixture. Also, with the additions the contraction of the curve after the phase change expansion is minimized considerably.

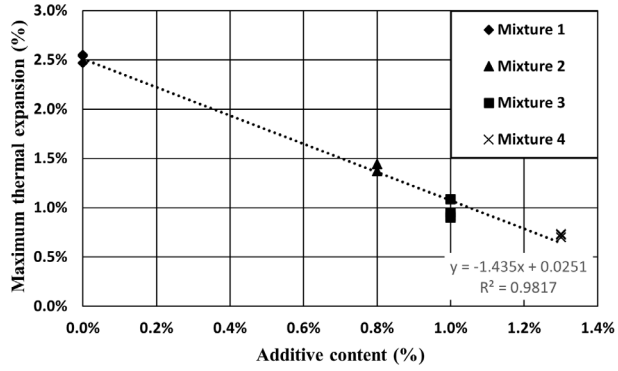

Fig. 8. Maximum thermal expansion of the sand mixture samples with varying amounts of additive

The dilatometric analysis indicates that both the expansion and contraction of thermal expansion curve characteristics depend essentially on the amount of additive in the sand mixture. The correlation between the average thermal expansion of sand mixtures with varying mass percentages of additives in the sand mixture is displayed in Figure 8. The decrease in the peak expansion values is consistent and exhibits a linear character with the quantity of additives added to the sand mixture matrix, and the minimal values are obtained with the additive concentration of 1.3% mass percentage in the sand mixture. The determination coefficient value of the trend line determined from the regression analysis approaches to unity, which suggests thermal expansion declination follows a linear trend concerning additive inclusions in the sand mixtures. Moreover, the negative slope of the curve signifies a decrease in peak expansion values as the quantity of additives within the samples increases.

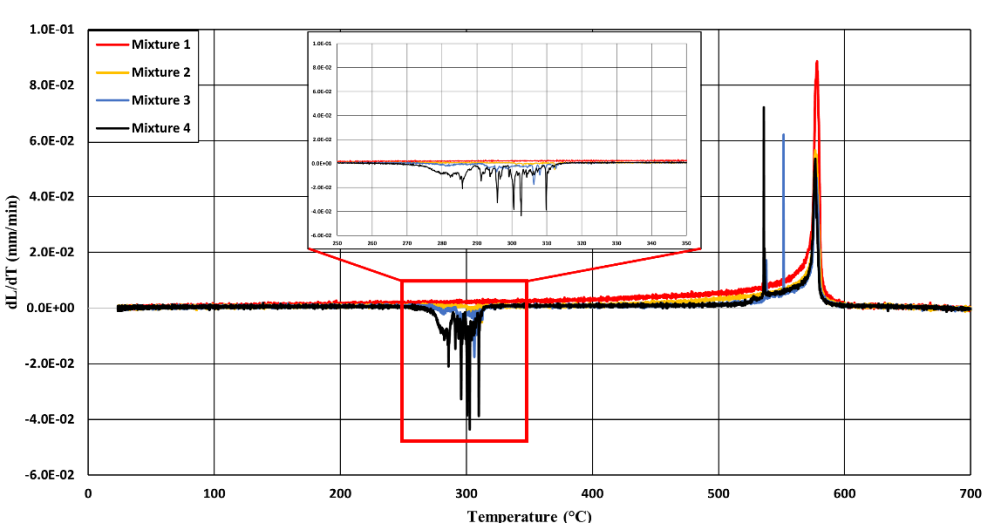

Fig. 9. First order derivative of dimensional change of sand mixtures up to 700 °C

The graph presented in Figure 9 displays the expansion rate of the samples in the cylindrical domain, within the heating cycle of sand mixtures up to 700 °C. The graph reveals the local maxima or minima in volumetric variation of the sand grains in the mixture within the subjected temperature range. Moreover, the graphical representation reveals the localized thermophysical stress generation between the grains due to irregular thermal expansion. From the observation, sand mixture with no inclusion of additives undergoes rapid dimensional variation at ca. 570 °C which is attributed to the crystallographic phase transformation of quartz, this leads to change in the specific density of the grains. The non-linear granular expansion develops thermal and shear stresses locally between the grains leading to probable cause for mould or core failure and causing defects such as rattails and veining. However, integrating additives into the sand mixture matrix has resulted in local minimization of unrestricted dilatometric expansion curve between the temperature of about ca. 270 to 320 °C. The negative deviation trend can be correlated with the addition of additives to the sand mixture, as seen in Figures 5, 6 and 7. The minimization or contraction of the curve is irregular, and the anomaly of the curve increases with the amount of additive. The local maxima in the expansion curve of mixtures containing additives illustrated an additional variation in expansion trajectory at ca. 550 °C. The uncertainty in irregular contraction and expansion can be associated with the heterogeneity of the mixture, the release of thermal stresses developed around the sand grains and diversity in the spatial distribution of additives within the matrix samples.

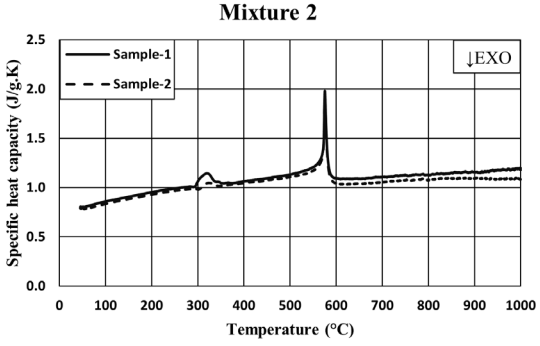

Fig. 10. Specific heat capacity of sand mixture with 0.8% additive

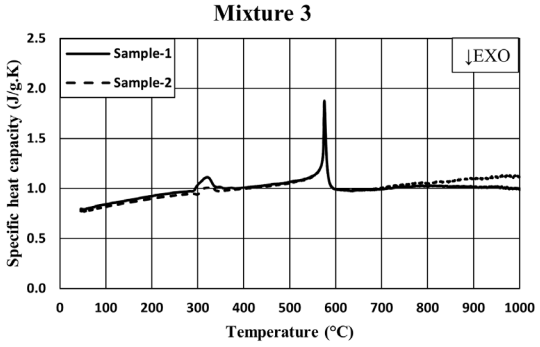

Fig. 11. Specific heat capacity of sand mixture with 1.0% additive

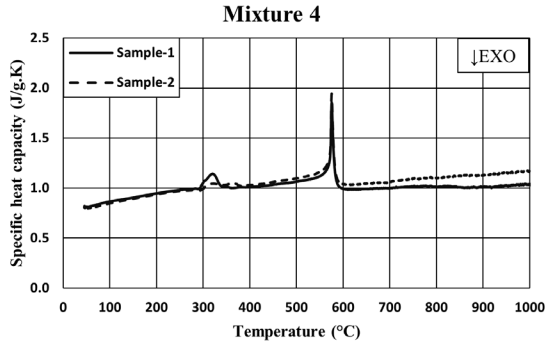

Fig. 12. Specific heat capacity of sand mixture with 1.3% additive

Table 4.
Onset temperature of endothermic peaks for DSC analysis in mixture with additives

Sand Mixture	Additive, (%)	Onset Temperature at Additives decomposition, (°C)	Onset Temperature at Phase transformation, (°C)
Mixture 2	0.8	302±7	572±0
Mixture 3	1	297±5	572±0
Mixture 4	1.3	296±3	572±0

The temperature-dependent specific heat capacity of the sand mixtures with varying content of additives is illustrated in Figures 10, 11 and 12. The samples underwent two different endothermic processes regardless of the amount of additives in the sand mixture. The onset temperature of the reactions is analysed and reported in Table 4. The onset temperature of the initial endothermic curve indicates that the reactions are initiated at the temperature of ca. 296–302 °C. This primary heat-absorbing reaction is observed up to temperature ca. 338±0.5 °C in the sand mixtures containing additives. The heat absorption in this temperature range is attributed to the decomposition of starch and cellulose in additives. The secondary endothermic reaction observed at 572 °C corresponds to the α-quartz to β-quartz transition. The structural reorganization of sand grains following the polymorphic phase variation of quartz results in irregularities in specific heat values.

3.3. TG-DTG Analysis of Additives

To strengthen the DSC results and understand the degrading mechanism of additives in the sand mixtures, thermogravimetric analysis is carried out independently on additives by excluding sand in the measuring samples. The overall weight loss percentage of the sample is plotted as a function of temperature and presented in Figure 13 below. The observations provide the potential degradation mechanism of the additive granules with temperatures reaching up to 1000 °C. The thermogravimetric analysis of the sample identifies a two-stage degradation of the additives across the modulated temperature range.

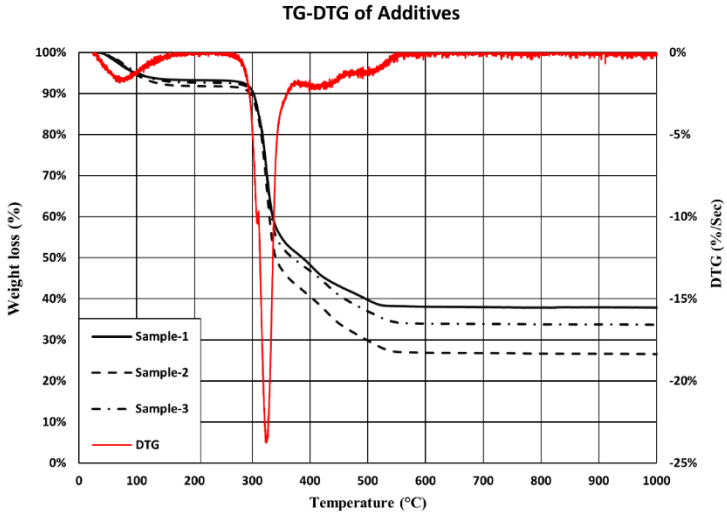

Fig. 13. Decomposition and degradation rate of additives as a function temperature measured using TGA

The initial weight loss occurring at ca. 100 °C accounts for ca. 7% of the total weight of the sample, and this is attributed to the dehydration of the material involving the evaporation of volatile substances in the additives such as moisture and water content. Subsequently, a secondary degradation of the additive starts to eventuate from ca. 280 °C and continues up to ca. 560 °C, and the molecular degradation of the additives results in a substantial weight loss of about 55 to 65 % of the overall weight of the sample. The variation in the weight of the material across this temperature range is attributed to the decomposition of starch and cellulose content, which are primary components in the additive [11, 12]. The decomposition rate represented by the DTG curve in Figure 13 unfolding major contribution to weight loss is from the decomposition of starch and other biodegradable compounds in the additives and is attributed to the elimination of hydroxyl groups, decomposition, and depolymerization of carbon chains within the structures of starch and cellulose. Moreover, the literature indicates the possibility of originating carbonaceous compounds consequent to the secondary degradation step of starch [13-15]. The synthesis of carbonic compounds is purely dependent on the local granular atmospheric condition generated throughout the pouring and solidification process of the melt. It is challenging to characterize the type of compounds eventuated from the degradation of additives as the material itself is highly heterogeneous in composition. The weight loss measurements obtained from thermogravimetric experiments can be correlated with the primary endothermic reactions prompted at ca. 300 °C in DSC measurement curves of sand mixtures containing additives (Figures 10, 11 and 12). After reaching the maximum temperature of the analysis, the residual mass of the samples ranged from 27 to 38 % of the initial mass.

Table 5.
Thermal degradation stages in additives.

Stage	Temperature, (°C)	Reaction
I	30-150	Dehydration
II	280-380	Depolymerization and Decomposition
	380-560	Possible formation of C-compounds

The results from TG-DTG explicitly state that weight loss is observed in the interval where thermal expansion curves registered shrinkage. The dilatation and thermogravimetric results infer that necessary voids are generated from the decomposition of biodegradable material in the additives between the temperature interval ca. 270 to 320 °C. At the mesoscopic level, the voids between the granular media, called solid intergranular voids, accommodate the free expansion of silica grains. Iron oxide content in the additives softens the granular material and provides thermal relaxation by increasing the hot plasticity of the grains from the thermal stresses generated on the grain surface [10, 16].

3.4. Effectiveness of pores generated from additives decomposition

Thermal expansion is thought to be suppressed by the formation of pores due to the decomposition of starch in the additives at around 300°C, which "absorbs" the expansion of silica sand. In this section, the effect of the newly created pores on the thermal expansion is discussed. An analytical calculation is made with some assumptions. The primary uncertainty to address is quantifying the amount of degradable starch present in the additives. The TGA experiments suggest the fractional degradation of additives is between 55 and 65% mass of additives in the samples. The volume fraction of starch is derived from the assumption that each experimental sample mixture contains a 60 % mass proportion of degradable mass with a density of 5.4×10^{-4} g/mm^3 [17]. Figure 14 shows the correlation between thermal expansion and the volume fraction of starch in experimental samples. The volume of starch in the additives increases with the proportion of additives in the sand mixture.

To investigate the effective absorption of non-stationary expansion of quartz granules by the newly created interstitial pores from the decomposition of additives, a calculation was made to determine the amount of suppressed expansion. The quantitative analysis was made with the consideration that the maximum expansion was achieved in the mixture with 0 % mass of additives. The results for each amount of additives are shown as "Theoretical" in Figure 15. As can be seen from this figure, when 0.8% to 1.3% of additive is added, the maximum thermal expansion is suppressed linearly from 55% to 85%, respectively. The experimentally obtained percentage absorption of maximum expansion is illustrated in Figure 16. From these results, it was found that 84% to 94% of the newly formed generated pores from additives decomposition absorb the expansion of the silica sand (Figure 15). The absorption capacity per unit volume of pore with the increasing amount of additives is constant with an experimental error (ca. 80-95%). These results indicate that, as previously believed, the pores generated by decomposition effectively contribute to the suppression of thermal expansion, fulfilling their role significantly. However, this also suggests that even if the decomposition of starch is optimized, further suppression of thermal expansion cannot be expected. Therefore, if further suppression of thermal expansion is desired, it is necessary to consider alternative suppression mechanisms.

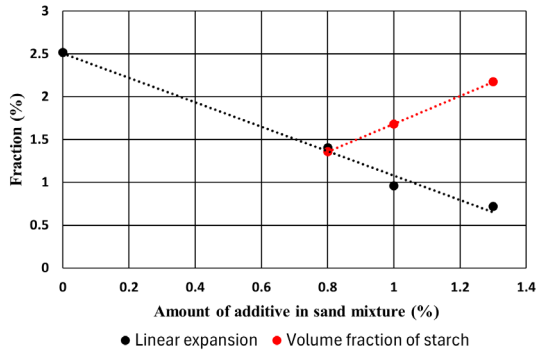

Fig. 14. Correlation between linear thermal expansion and volume fraction of degradable components in the additives

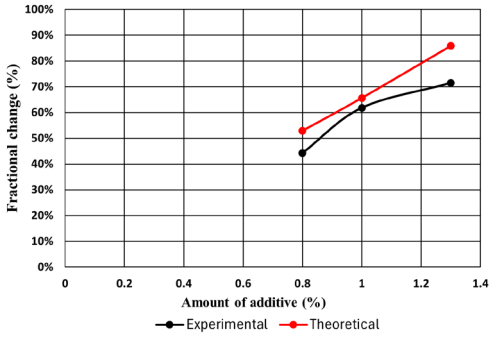

Fig. 15. Percentage reduction of maximum expansion

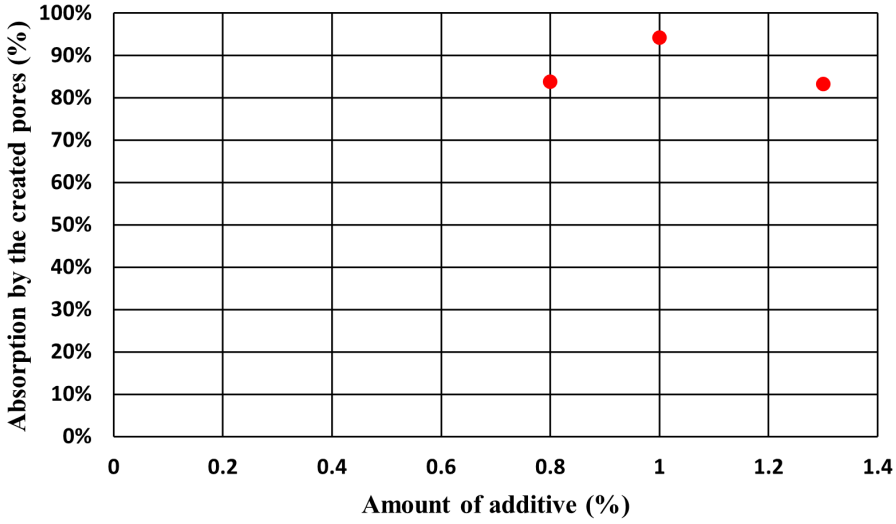

Fig. 16. Effectiveness of volume absorbed by pores created from additives degradation

4. Conclusions

The casting temperatures of cast iron and cast steel result in uneven temperature gradients across the sand core made of silica. The interaction of bonded sand granules with molten melt varies the thermophysical properties of the quartz granules. The dilatometric measurement observed a linear expansion of 2.5% in pure sand, which results in volumetric expansion of the core yielding different surface defects on the finished castings. The findings identified that unrestrained expansion is reduced by incorporating the additives into the aggregate mixture, and the maximum reduction in the aggregate matrix is observed in the mixture with 1.3% additives. Results from the DSC and TGA experiments infer that 60% of degraded additives generate interstitial voids between the sand grains in the granular media, which facilitate the phase expansion of the silica granules at ca. 573 °C. The estimated findings suggest that the voids originating from the additives decomposition are not entirely occupied by subsequent granular expansion. The quantitative findings indicate that the pores absorb the expansion with an efficiency of 84 to 94%. The correlative investigations in this paper provide a comprehensive understanding of the influence of additives on the expansion dynamics of quartz granules and provide essential knowledge for the development of moulding and core-making processes, thereby improving the casting quality. Furthermore, the volume of gases released from the additive's decomposition is undetermined, whichh could contribute to the total gas volume generated at the mould-metal interface and might cause gas entrapment defects in the liquid metal. An extensive investigation is required to quantify and characterize the byproducts obtained from the thermal decomposition of additives.

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