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Influence of Molding Mixture Composition on TOC and BTEX Emissions

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Abstract

The work deals with the issue of BTEX (benzene, toluene, ethylbenzene, xylenes) and TOC (Total Organic Carbon) emissions arising from molding mixtures due to the decomposition of organic compounds at high temperatures and lack of oxygen. The measurements are focused on the combination of bentonite molding mixture with cold-box cores, which is very often used for casting cast iron castings, especially in serial and mass production. The number of emissions is controlled by changing the air access to the mold by choosing the grain size of the silica sand and optimizing the mold-metal ratio by changing the height of the molding mixture above the casting. The highest measured TOC value was in the mold made of mixture ŠH32/E/4CB/20, which exceeded the still valid limit of 150 mgC/m^3. The TOC values of the other molds were around the limit of 50 mgC/m^3. The amount of BTEX emissions correlates with TOC measurements, where up to 3 times more emissions were released from the molds prepared of ŠH32/E/4CB/20. The idea behind the measurements is find a simple and easily applicable methods for foundry industry leading to a reduction in TOC and BTEX emissions. The results so far show that by optimizing the composition of the molding mixture and adjusting the ratio between the molding mixture and the metal, the amount of these emissions can be effectively reduced. The influence of the type of used silica sand, especially its squareness, origin and the ability of different types of bentonites to absorb the resulting harmful compounds and thus reduce emissions released from the molds remains an open question.

Keywords: BTEX, TOC, Green sand, Cold-box technology, Emission

1. Introduction

Today, one of the most widely debated issues in foundry engineering are emissions. Despite being a significant source of pollutants during the casting process, green sand molds continue to dominate foundry production. The oldest, simplest, most cost-effective, and still widely applied method for producing molds is the use of bentonite mixtures. These mixtures are suitable for casting most produced alloys, particularly various types of aluminum alloys and cast iron. When casting iron, green sand mixtures typically contain so-called “lustrous carbon carriers,” which improve the surface quality of cast iron castings. However, they also represent one of the largest sources of BTEX emissions (benzene, toluene, ethylbenzene, and xylenes). Another major source of emissions is the use of cores, most often manufactured from mixtures with organic binders. These binder systems generate BTEX emissions under high temperatures and oxygen-deficient conditions. Nevertheless, they remain the most widely used technology for core production, as they ensure dimensional accuracy, shape stability, good shake out properties, storage durability, and the ability to manufacture complex geometries. The key unresolved question in this context is how to avoid or at least minimize the generation of BTEX and total organic carbon (TOC) emissions to acceptable levels.

Among the most monitored emissions are BTEX compounds (benzene, toluene, ethylbenzene, and xylenes). These emissions are also considered within the overall assessment of pollutants measured in exhaust gases from foundry operations, typically expressed as total organic carbon (TOC). The importance of

controlling both the concentration and the total released amount of BTEX compounds, particularly benzene, arises from their harmful effects. Benzene is a recognized carcinogenic and mutagenic substance. Organizations such as the World Health Organization (WHO) and the International Agency for Research on Cancer (IARC) classify benzene as a Group 1 carcinogen. Long-term exposure to elevated concentrations of benzene adversely affects human health and may lead to serious diseases. Therefore, monitoring BTEX emissions is of critical importance from both an environmental protection and an occupational health perspective [1-4]. Legislative limits for both BTEX and TOC emissions are established in accordance with European Union regulations. Their definition is guided primarily by the BREF (Reference Document on Best Available Techniques) and BAT (Best Available Techniques) frameworks. The concept of Best Available Techniques (BAT) is embedded in numerous international policy and regulatory documents addressing environmental protection. The application of BAT in industrial practice is intended to ensure a consistently high level of environmental performance and compliance with evolving sustainability standards. [5-7]

Emission-reduction technologies currently applied in foundries have remained largely unchanged for several decades. As a result, it is increasingly challenging for the industry to keep pace with contemporary requirements in emission control. In other industrial sectors, however, a wide range of methods for eliminating various pollutants are already well established. For example, particulate matter (PM, referred to as TZL in Czech terminology) is commonly removed from air streams using filters (fabric or ceramic) and separators (electrostatic or mechanical) [8, 9]. More advanced air protection technologies include photocatalytic coatings, which actively degrade nitrogen oxides (NOx) and volatile organic compounds (VOCs) under light exposure. In the case of greenhouse gases, direct air capture (DAC) technologies are being developed to remove CO_2 directly from the atmosphere, followed by underground storage [10]. With respect to BTEX and TOC emissions generated in foundries, specialized abatement systems can also be employed. For instance, Advanced Catalytic Purifier (ACP) technology utilizes catalytic oxidation to remove TOC, VOCs, amines, phenols, and formaldehyde [11]. Another option is the use of ultraviolet purification (UVP), in which exhaust pollutants are decomposed into less harmful compounds such as H_2O and CO_2 through UV irradiation and photo-oxidation processes [12]. Some of these technologies might be also used in the foundry industry, however their high costs make an obstacle to foundries. Therefore, it is important to find out how to decrease the emission levels from the foundries without these additional high costs.

In industrial practice, a common combination is green sand mixtures together with cores produced using organic binders, most often through cold-box technology. A major source of emissions from green sand, as previously noted, are so-called lustrous carbon carriers, such as coal dust or hydrocarbon resins [13]. Numerous studies have explored alternatives to conventional lustrous carbon carriers with the aim of reducing emissions. For example, the authors of [13] investigated the development of an inorganic–organic binder system based on bentonite clay modified with organic compounds (organophilic bentonite). In another study, the authors of [14] examined various lustrous carbon carriers (coal dust, amorphous carbon, hydrocarbon resins) with respect to their ability to generate lustrous carbon, their loss on ignition, and their influence on mechanical and technological properties of green sand mixtures. Additional reports describe attempts to replace coal dust with hydrocarbon resins [15, 16] or cellulose-based additives [17, 18]. However, these substitutes still contain high amounts of carbon and therefore cannot be considered an effective long-term solution for emission reduction, as their thermal degradation continues to produce significant levels of hazardous BTEX compounds. [14, 19, 20]

From the perspective of foundries that will soon be required to reduce TOC and BTEX emissions, it is essential to identify solutions that are both simple, and effective, and that can be implemented in practice on at least a temporary basis. This necessity arises from the fact that most of the advanced abatement technologies discussed above are prohibitively expensive and thus inaccessible for many foundry operations. One relatively straightforward approach involves improving air access and enhancing gas evacuation from molds, thereby minimizing the pyrolytic decomposition of lustrous carbon carriers. This can be achieved, for example, through optimization of the molding mixture composition and the mold-to-metal ratio as performed also in [3]. The following experimental section therefore focuses on assessing the influence of bentonite mold composition and mold-to-metal ratio on the generation of TOC and BTEX emissions. In this context, only a limited number of studies reflect real foundry conditions, particularly with respect to the mold-to-metal ratio and the combination of different binders within the mold system. Existing studies typically employ an experimentally controlled pouring methodology using a testing chamber optimized for efficient exhaust gas capture. This approach enables the monitoring of pyrolysis and combustion by-products generated during the thermal decomposition of binders. [3,4]

2. Materials and methods

For the purposes of the experiment, one of the most employed combinations in cast iron foundries was selected. Accordingly, green sand mixtures for mold and core preparation were produced using the cold-box process. The experimental device was devised according to the best possibility of capturing the emissions and controlling the gas evaluation and also to reflect the conditions and the process in the foundry industry.

The prepared molds were poured at a portable casting station. After the melt (grey cast iron) reached the required casting temperature 1380°C, it was poured into the mold. Immediately after pouring, the portable casting station was positioned beneath a lid mounted on the mold frame. The lid was connected to an exhaust duct. An exhaust gas sampling probe was installed in the exhaust duct and connected to a SmartFID analyzer for the determination of total organic carbon (TOC). In addition, an activated carbon adsorption tube was integrated into the exhaust system and connected to a suction pump. The pump ensured controlled extraction of the exhaust gases, which passed through the activated carbon tube, where the emitted compounds were captured by adsorption. The pump also recorded the total volume of the extracted gases. This parameter is essential for the calculation of emission concentrations in the exhaust stream. Emission measurements of TOC were conducted over a period of

60 minutes and the collection of BTEX emissions were conducted simultaneously.

2.1. Preparation of bentonite molding compound

The starting materials for the preparation of the molding mixture were selected as quartz sand from Šajdíkove Humence (ŠH) in two grain sizes. Specifically, quartz sand ŠH was used in the fractions ŠH34 (d_{50} = 0.24 mm) and ŠH32 (d_{50} = 0.38 mm). These sand fractions represent the most utilized materials for green sand mold production and correspond to the lower and upper limits of grain sizes suitable for foundry mold. Bentonite was chosen from Clariant and contained carbon-forming additives (ECOSIL® D02 S78).

The green sand mixture was prepared using a +GF+ wheel mixer. Initially, the dry components were mixed for 1 minute, after which water was added, and the mixture was further homogenized for an additional 3 minutes, resulting in a total preparation time of 4 minutes. The prepared mixture was then transferred to sealable plastic containers and allowed to rest for 24 hours. Key technological properties of the green sand mixture, including permeability and strength, were evaluated using laboratory cylinders (Φ50 × 50 mm). The composition and average technological parameters of the bentonite mixtures are summarized in Table 1.

Table 1.
Composition of green sand mixtures

Parameter		ŠH32/E	ŠH34/E
Water [%]		2,5	2,5
Bentonite [%]		5	5
Strength [kPa]	Immediate	57,3	57,4
	24 h	57,1	57,5
Permeability [j.p.SI]	Immediate	476,0	225,5
	24 h	442,6	200,4

2.2. Preparation of cores using cold-box technology

The core mixtures were prepared using a laboratory S-mixer with the ECOCURE SILVER 1010/2011 binder system from ASK Chemicals. The dosage of both binder components was set at 0.7% by weight of sand, corresponding to 2 kg. Initially, the sand was mixed with component 2 of the binder system for 1 minute. Component 1 was then added, and the mixture was further homogenized for an additional 1 minute. Cores were subsequently formed at a pressure of 2 bar and cured with a tertiary amine of the DMPA type, applied at a dosage of 2 ml for 20 seconds. The specifications of the core mixtures are summarized in Table 2.

Table 2.
Composition and average strength values of core mixtures

Parameter		ŠH32	ŠH34
Binder [%]		0,7 / 0,7	0,7 / 0,7
Bending strength [MPa]	Immediate	2,2	2,5
	1 h	3,4	3,7
	24 h	3,9	4,2

2.3. BTEX Measurement - GC

Gas chromatography is widely regarded as one of the most suitable techniques for the detection and analysis of organic compounds [13]. It is an analytical separation method based on the repeated establishment of an equilibrium distribution of the analyte between the stationary and mobile phases [21-23]. Its primary advantages include rapid and straightforward analysis, efficient separation of components, and the requirement of only a minimal sample quantity for analysis [13].

BTEX emissions were collected using an adsorption method, employing Dräeger-type G activated carbon detection tubes (220 mg). One end of the tube was connected to an air extraction apparatus, while the other end was attached to a pump to ensure the withdrawal of exhaust gases from the sampling pipe. Measurements were conducted over a period of 60 minutes, with detection tubes being replaced at 15-minute intervals to prevent saturation of their adsorption capacity. Following the measurement, the used detection tubes were immediately sealed to prevent the release of captured emissions. The collected samples were then prepared as liquids for quantitative analysis using gas chromatography.

An Agilent 7890 Plus gas chromatograph with FID was used to evaluate the analyte. The chromatograph is equipped with an Agilent 7693 A autosampler, a capillary silicone column Φ0.5 mm – 50 m. Hydrogen H with a purity of ≥ 99.999% is used as a carrier gas and a flame source in the FID. The samples are dosed into a heater with a temperature of 280°C by the autosampler.

2.4. TOC measurement with SmartFID

For continuous monitoring of TOC emissions, a portable Smart FID device (ErsaTec) was employed. The system consists of a heated sampling tube (ϕ50 mm, 2 m in length) maintained at 180°C, and a detector housing containing a flame ionization detector (FID). During measurement, gas samples are continuously drawn from the suction apparatus and conveyed through a heated hose to the FID detector, where a hydrogen flame combusts the sample. Organic compounds are decomposed in the flame to form CH fragments, which subsequently react with atomic oxygen to generate CHO ions. These ions are then subjected to an electric field, producing a small current proportional to the TOC concentration in the sampled gas.

Prepared molds for casting and subsequent measurement were positioned on a mobile casting platform. A dedicated apparatus for emission measurement, integrated with a gas extraction circuit, was employed. Immediately after casting, the molds were placed

beneath this measurement structure (Fig. 1). Emission measurements were conducted over a period of 60 minutes.

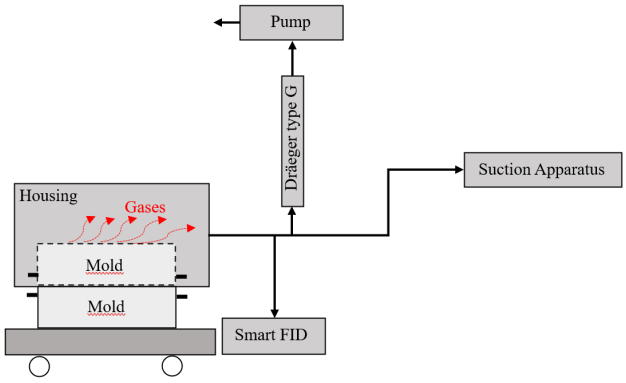

Fig. 1. Apparatus used for measuring BTEX and TOC

3. Description of achieved results

A total of three mold types were prepared and subsequently compared. The preparation and properties of the green sand mixtures and cores are detailed in Sections 2.1 and 2.2. The compositions of the molds are summarized in Table 3.

Table 3.
Composition of the forms used

Sand	Bentonite	Cores [ks]	Height [cm]	Marking
ŠH32	ECOSIL	4	20	ŠH32/E/4CB/20
ŠH32	ECOSIL	4	5	ŠH32/E/4CB/05
ŠH34	ECOSIL	4	5	ŠH34/E/4CB/05

The idea behind the aforementioned mold compositions was to establish conditions that would promote both the maximal and minimal formation of BTEX and TOC emissions. This was achieved by varying the grain size of the molds and the height of the molding mixture over the casting, thus the mold-to-metal ratio differs.

3.1. Measurement results TOC

The results of TOC measurements using the Smart FID device are presented in Figure 2. Prior to measurement, the device was calibrated with propane (93.5 ppm) to ensure accuracy and linearity. Immediately following casting and heating of the mold with cores, combustion and thermal degradation of organic compounds occur, which is reflected in an increasing TOC value. Within the first seconds after metal pouring a sharp increase in TOC is associated with the initial thermal decomposition of the binder and a pyrolysis of carbonaceous substances. 1 After reaching the peak the emission rate gradually decreased and was held under the 60 mgC/m³. The highest TOC concentration of 173.6 mgC/m³ was recorded at a mixture height of 20 cm above the casting. This value exceeds the current maximum limit of TOC emission of 150 mgC/m³. Future regulatory limits are expected to be reduced to 50 mgC/m³. The TOC concentration reached this level only after 19 minutes of measurement, following an initial sharp increase, after which the TOC amount steadily decreased.

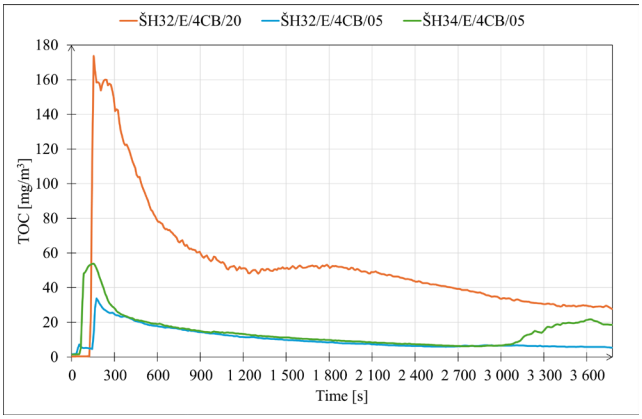

Fig. 2. Record of TOC measurement values

For a mixture height of only 5 cm above the casting, air access to the mold is enhanced, creating conditions more favorable for the combustion of organic compounds, as reflected in the measured TOC values. In the case of the finer-grained ŠH34/E/4CB/05 mixture, the maximum TOC concentration reached 53.82 mgC/m³, whereas for the coarser-grained ŠH32/E/4CB/05 mixture, the maximum value was 33.73 mgC/m³.

These results correspond to the researches given in [4] and [24], where also was observed the sharp increase in emissions in first 250 s and later the emission levels were minimal.

3.2. Measurement results BTEX

During the TOC measurement, samples were simultaneously collected for laboratory analysis of BTEX concentrations in the exhaust air. The total concentrations of the individual BTEX compounds are presented in Figure 3.

When comparing the measured BTEX concentrations from the different mixtures, it was observed that the benzene emissions were dominant. In case of mold ŠH32/E/4CB/20 was released up to 2.5 times more benzene and up to 3 times more toluene than the ŠH32/E/4CB/05 and ŠH34/E/4CB/05 mixtures. This finding indicates that increasing the mold-to-metal ratio leads to a higher release of BTEX compounds. In molds with a mixture height of 5 cm above the casting (ŠH32/E/4CB/05 and ŠH34/E/4CB/05), the differences in individual BTEX components and their total concentrations were negligible. The total BTEX concentration from the finer-grained ŠH34/E/4CB/05 mold was 49.864 mg/L, only slightly higher than the 48.163 mg/L measured for the ŠH32/E/4CB/05 mold.

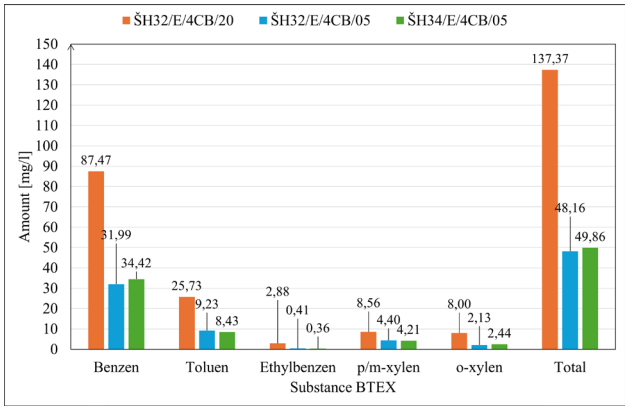

Fig. 3. Amount of BTEX compounds released from molds

This minimal difference can be attributed to the insufficient height of the molding mixture above the casting, which limits its capacity to influence emission filtration across different grain sizes. Greater differences between these two molds were observed in the TOC measurements presented in Figure 2.

4. Conclusions

The purpose of the present measurements was to investigate the conditions governing the formation of TOC and BTEX emissions as a function of input parameters, including grain size and the use of cold-box cores, therefore investigation the combination of the different mixtures in accordance with the emission evolution. Parameters such as the grain size of the sand and the height of the molding mixture above the casting influence mold permeability, which in turn fundamentally affects the rate of combustion of organic compounds present. The experiments confirmed the feasibility and reliability of the applied measurement methodology and corresponds to the research which were conducted under the similar conditions [3,4]. A key insight from these results is that, by appropriately selecting mold-to-metal ratio and the grain size, it is possible to achieve low TOC concentrations, which are expected to comply with the forthcoming regulatory limit of 50 mgC/m³ or approach this value. The results obtained to date indicate that optimizing the composition of the molding mixture, specifically changing the grain size, and adjusting the mold-to-metal ratio, particularly in the upper portion of the mold, can effectively reduce emission levels. The influence of the type of silica sand, especially its angularity, source, and the adsorption capacity of different bentonite types in capturing harmful compounds and mitigating emissions from the molds, remains an open question. Simple technological measures aimed at reducing emission loads represent a first practical step that foundries can implement in the upcoming years without requiring significant investment. Consequently, these measures constitute an important component of the gradual transition of foundry operations toward more environmentally sustainable production.

The limitations of this study are strictly defined by the materials used, the casting conditions, and the mold parameters. The presented results cannot be directly extrapolated to other materials or processes, as each casting process is inherently specific. In particular, the results are strongly influenced by casting conditions such as the type of material, melt temperature, ambient temperature, and related factors. This work reflects real industrial environments and seeks to identify and describe relationships between material characteristics and emission behavior. The study is also aligned with the most recent emission limits established by the European Union.

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