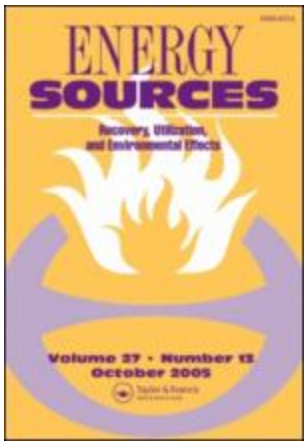

Advances in the Production of a Cement Substitute from Fermentation Residues

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Complete List of Authors:	Maroušek, Josef; The Institute of Technology and Business in České Budějovice Marousková, Anna; The Institute of Technology and Business in České Budějovice, Kůs, Tomáš; The Institute of Technology and Business in České Budějovice
Keywords:	fermentation residues, pyrolysis, particulate matter, bioeconomy, developing countries

Title:

Advances in the Production of a Cement Substitute from Fermentation Residues

Authors:

Josef Maroušek*, Anna Maroušková, Tomáš Kůs

Abbreviation (all):

Institute of Technology and Business in České Budějovice

Contact:

* corresponding author (josef.marousek@gmail.com, +420 777 987 683)

Abstract:

The amount of fermentation residues from biogas stations is rising rapidly worldwide. Nevertheless, farmers are losing their interest in its incorporation into soil since the level of nutrients as well as the agrochemical value of the organic matter present is low. Unlike combustion (carried out in the presence of oxygen), the product of which is ash, pyrolysis (without oxygen) turns biowaste into a highly porous carbonaceous material that can substitute cement, one of the most energy-intensive mass-produced materials in the world. This practice is booming in developing countries. Due to saving and inexperience, farmers or local communities do not apply the technologies to utilize or filter the hazardous gaseous products of pyrolysis. However, these contain high levels of particulate matter (PM) that absorbs hazardous cocktails and facilitates their spread to the surroundings. An inexpensive and easily producible shower cooler was designed and analyzed in full operation. The results obtained suggest that the proposed solution can significantly reduce a wide range of PM sizes, creating preconditions for reducing negative impacts on the environment as well as health of locals.

Keywords:

fermentation residues; pyrolysis; particulate matter; bioeconomy; developing countries

Introduction:

Biogas production, in addition to food industry and biorefining technologies, produces huge amounts of fermentation residues worldwide (Maroušek et al., 2018). The application of

fermentation residues into arable land is raising economic and agronomic concerns among farmers because the level of nutrients is low (some 3 % in total) and these must first undergo mineralization via soil biota (slow and lossy process) to become available for plant nutrition (Vochozka et al., 2017). Furthermore, the agrochemical value of the organic matter in fermentation residues is disputed (Kolář et al., 2008) which altogether results in soil degradation, in particular worsening of the water retention capacity (Smetanová et al., 2013). There are growing reservations towards the landfilling of biowaste since it is considerably bulky and its subsequent biodegradation worsens the stability of landfills, not to mention the unnecessary production of greenhouse gases (Chavez et al., 2019).

The building industry and, particularly, the cement industry are among the most significant emitters of CO_2 in the world (Stehel et al., 2018). Ordinary concrete typically contains about 12% of cement (Salas et al., 2016). The global production of cement has grown rapidly in recent decades; it is the third–largest source of anthropogenic emissions after fossil fuels and land use change (Andrew, 2018). Cement plants release some 7% of global CO_2, while 900 kg of CO_2 is emitted per 1.000 kg of cement. Its cumulative emissions from 1928 to 2016 were 39.3 ± 2.4 Gt CO_2, 66 % of which have occurred since 1990. However, such a picture is far from complete; batching, mixing, transport, placement, consolidation, finishing and, especially, waste management are not considered in these estimations (Salas et al., 2016). At the same time, concrete industry is one of the largest industrial consumers of fresh water (Maroušek et al., 2019a; Guo et al., 2017). In addition, producing a ton of Portland cement, the most common hydraulic cement, requires about 4 GJ of energy, making it one of the most energy-intensive mass-produced material on the globe (Malhotra, 1999). To make matters worse, the cement manufacturing process produces millions of tons of cement kiln dust each year, contributing to respiratory and pollution health risks (Huntzinger and Eatmon, 2009). Emissions from cement production (typically NO_x) react with O_3 and volatile organic carbons (VOCs) to form secondary particulate matter (PM). At ambient temperature and with an excess of O_2, NO is oxidized to NO_2 (toxic by inhalation and causing irritation to the human eye, nose and throat) which is a precursor to the hazardous HNO_3 (Seangkiatiyuth et al., 2011).

There are two main aspects of cement production that result in a significant environmental burden. The first is the chemical reaction involved in the production of the main component of cement, clinker, as carbonates (largely limestone, $CaCO_3$) are decomposed into oxides

(largely lime, CaO) and CO_2 by the addition of heat (Boden et al., 2017). It should be noted that the calcination process (driving off CO_2 from $CaCO_3$ to form CaO) accounts for roughly half of the CO_2 emitted, while the remaining carbon results from energy usage during the production process (Huntzinger and Eatmon, 2009). The second source of emissions is the combustion of fossil fuels to generate the significant energy required to heat the raw ingredients to well over 1000 °C (preferably above 1400 °C), and these "energy" emissions, including those from purchased electricity, could add a further 60 % on top of the process emissions (Andrew, 2018). Furthermore, mining large quantities of raw materials such as limestone and clay and fuel such as coal often results in extensive deforestation and soil degradation (Vochozka et al., 2017; Mehta, 2001).

Benhelal et al. (2013) proposed three strategies of CO_2 reduction, including energy saving, carbon separation and storage, as well the use of alternative materials. In the case of energy saving approaches, moving from a wet to a more efficient dry process with a calciner has since then reduced up to 50% of the required energy and mitigated almost 20% of CO_2 emissions, but other options are not widely known (Salas et al., 2016). Modern cement plants tend to have high energy efficiency and the scope for reducing CO_2 emissions by further efficiency improvements was found to have little environmental saving in comparison to the dry process (Barker et al., 2009). Economic challenges (40 € t^{-1} of CO_2 avoided for a European cement plant and 23 € t^{-1} for a plant located in Asia, Barker et al., 2009) present considerable obstacles to implementing carbon capture and storage processes in cement plants (Maroušek et al., 2019b). As far as alternative materials are concerned, fly ash, blast furnace slag, palm oil waste, recycled concrete, zeolite and other materials have been used for manufacturing blended cement (Salas et al., 2016). A plethora of experiments have shown that the incorporation of raw biomass into concrete results in inconsistent mechanical properties (Britt and Kangas, 2016). Many have experimented with biomass ash that is more homogeneous (Matalkah et al., 2016). Biomass ash, however, like fossil ash, produces round conglomerates, which show little chemical reactivity due to their minimalist surface and thus tend to degrade the quality of concrete (Katare and Maduwar, 2017).

Following the above, most attention has recently been paid to charred biowaste (also known as biochar) and its abilities to substitute cement (Gupta et al., 2018). Experimental results suggest that the addition of biochar reduces the initial setting time and significantly improves the early compressive strength of mortar. Biochar addition can impart ductility to mortar under flexure, although flexural strength was not significantly influenced. Water penetration

and sorptivity of mortar was significantly reduced due to the addition of biochar, indicating higher impermeability in biochar-added mortar. It was also found that the addition of treated biochar results in significantly higher mechanical strength and improved permeability. The published findings show that 5% replacement of cement (by weight) by biochar improved the 28–day compressive strength of mortar by about 10%. However, the flowability of biochar–mortar is reduced with a higher replacement rate. According to Choi et al. (2012), biochar tends to absorb and hold a significant part of mixing water, thus resulting in a stiffer mix. The physically absorbed water in biochar is later released during the hardening of mortar and can contribute to internal curing. Roberts et al. (2009) state that depending on the type of feedstock and preparation conditions used, biochar has the potential for reducing net greenhouse gas emissions by about 870 kg CO_2 per ton of dry feedstock, of which 62 – 66% are realized from carbon capture and storage by the biomass feedstock of the biochar. It was repeatedly and independently reported (Stehel et al., 2018; Katare and Maduwar, 2017; Vochozka et al., 2017) that the addition of 1 % of biochar prepared at 800 °C improved the modulus of rupture and fracture energy of cement by up to 61%. The study suggests that biochar can act as micro-aggregates and improve the compressive strength, bending strength and fracture energy of cement. The improvement in fracture energy due to the introduction of biochar has been attributed to the tortuosity of the crack path. Biochar particles introduce inhomogeneity in the matrix and attract crack paths towards them. The biochar particles have the ability to absorb energy before failure, which improves fracture energy and bending strength.

There is a wide consensus that biochar from fermentation residues has the potential for successful deployment as an additive or substitute of cement, which would also promote waste recycling and sequester high volume carbon in civil infrastructure. Nevertheless, during biowaste pyrolysis, a large number of reactions take place in parallel and in series, including dehydration, depolymerisation, isomerization, aromatisation, decarboxylation and charring, altogether resulting in various solid, liquid and gaseous components. There is an increasing demand for pyrolysis oil (free flowing organic liquid mixture, which generally consists of a great amount of water and hundreds of organic compounds, such as acids, alcohols, ketones, aldehydes, phenols, ethers, esters, sugars, furans, alkenes, nitrogen compounds and miscellaneous oxygenates as well as solid particles, Razaei et al., 2014) while special devices (flash pyrolysis) are being designed exactly for its production and subsequent upgrading (Kan et al., 2016). Gases released from biomass pyrolysis consist of CO_2, CO, H_2, low carbon

number hydrocarbons such as CH_4; C_2H_6; C_2H_4 and C_3H_8, and small amounts of other gases, such as NH_3, NO_x, SO_x and alcohols of low carbon numbers. In terms of health effects, especially the Particulate Matter (PM) plays a key role as it captures and drifts the VOCs through the air over long distances (Madureira et al., 2016). PM_{10} refers to PM of aerodynamic diameter above 10 μm, furthermore PM_5, $PM_{2.5}$ and PM_1 for smaller (and thus more reactive) diameters are established. Even in low concentrations (at the edge of detection), the presence of PM and VOCs can lead to significant impacts on respiratory health, such as low lung function, asthma and bronchitis. Many studies have shown that PM is also contaminated with heavy metals and other hazardous pollutants such as polycyclic aromatic hydrocarbons (PAHs) that can directly enter the body through inhalation, dermal contact and oral ingestion exposure pathways (Wei et al., 2015). In particular, the smallest PM is of increasing concern because it can be easily inhaled deep into the lungs (Khan et al., 2010). As reviewed by Hossain and Davies (2013), pyrolysis gas has multiple potential applications, such as production of individual gas components, including CH_4, H_2 or other VOCs, or in production of liquid biofuels through synthesis. However, all of these require sophisticated and costly equipment that is completely beyond the economic reality of small farms and communities running the majority of biogas stations worldwide. Thus, for economic reasons, most of the small pyrolysis units operating worldwide do not contain any filtration device.

Hypothesis was build, whether the current situation can be improved by designing undemanding shower cooler of the pyrolytic gas (se shown in Fig. 1) that would be incorporated to the pyrolysis unit.

Methodology:

Fermentation residues were obtained from the Nedvědice biogas station (Czech Republic). The technological design and the corresponding processing parameters of the biogas station as well as the feedstock properties are traceable in Maroušek (2013). Fermentation residues were kept at 4°C until analyzed on biochemical properties according to Kolář et al. (2008), the results being stated in Tab. 1. An analysis of nutrients and heavy metals was carried out according to Ratajová (2014) and is summarized in Tab. 2. The fermentation residues (average particle size of 0.9 mm) were mechanically dewatered by a single helix press (PHARMIX, s.r.o, Czech Republic), which operates at 16 revolutions per minute (rpm),

representing a backpressure tension of 650 N that resulted in dewatering to 43 % volatile solids (VS). The mechanically dewatered fermentation residues were subjected to the continuous (150 kg hr^{-1}) standardized UHL–07 pyrolysis unit (Aivotec, s.r.o., Czech Republic). In brief, the pyrolysing apparatus consists of the entrance hopper equipped with an inner vertical slow motion helix. The slowly rotating helix continuously compresses the biowaste down into the mechanical turnstile located at the bottom of the hopper. The turnstile provides a minimum air leakage to minimize combustion and related ash formation. The turnstile leads to the pyrolysis chamber made up of a thick-walled refractory horizontal wide cylinder, where the material is exposed to the external source of heat (for more construction details see Hašková, 2017). 400 kg of dewatered residues were fed into the reactor and pyrolysed at 250, 300, 350, 400 and 450 °C each (speed of the horizontal helix that is responsible for the hydraulic retention time was set to 0.4 Hz, which corresponds to the delay of the feedstock in the pyrolysis chamber for approximately 6 minutes), while the pyrolysis gas was analyzed using the SPS30 PM sensor (Sensirion AG, Switzerland), which operates on the laser-based scattering principle and is capable of detecting PM at 10; 5; 2.5; 1 and 0.5 µg m^{-3}) every second. Two groups of experiments (with and without the shower cooler, Fig. 1) were performed. Statistics was carried out using the ZunZunSite3 software (zunzun.com) where the fitting target is the lowest sum of the squared absolute error.

Results and Discussion:

Hawken et al. (2013) pointed out that only a negligible amount of all materials actually ends up in the desired products, with most of the virgin materials returned to the environment as harmful solid, liquid, and gaseous wastes. Mardoyan and Braun (2015) highlighted that it is advisable to close the loops in extraction and manufacturing and turn biowaste into valuable products. Almost two decades ago, Mehta (2001) predicted that an alternative concrete system provides a model for the future, making concrete mixtures that shrink less, crack less, and would be far more durable and resource-efficient than conventional Portland-cement concrete. According to Škapa and Vochozka (2019) this prediction is coming true and charred biowaste is the answer.

With regard to Tab. 1 and 2, it can be stated that the biochemical properties of the fermentation residues used during the experiments are in good agreement with other literature world-wide (Stehel et al., 2018), in particular with reports from developing countries (Kolář et

al., 2008) where the cement substitute is currently produced most intensively (Matalkah et al., 2016). This finding is a good prerequisite for the generalization the following knowledge.

Data plotted into Fig. 2 shows many remarkable and so far unpublished findings. It is firstly observed that at lower temperatures, pyrolytic gas contains mostly medium-sized and bigger particles and with increasing temperature, its amount is decreasing. The available literature does not provide a reliable explanation for this phenomenon (Matalkah et al., 2016; Salas et al., 2016; Rezaei; Roberts et al., 2009). It is hypothesized that lower temperatures are not sufficient to release the finest particles; however, such a hypothesis requires more experiments. In contrast, it can be deduced from the visualized data that the amount of the finest PM increases with higher temperatures. Following the available literature (Seangkiatiyuth et al., 2011; Huntzinger and Eatmon, 2009), one might conclude that higher temperatures of the pyrolysis process could thus be more environmentally harmful (Madureira et al., 2016), since these smallest particles reach the deepest parts of the respiratory system, where they are most biologically harmful (Hossain and Davies, 2013; Khan et al., 2010). Although such an observation requires further examination, it can be assumed that this is not the case in the instance of pyrolysis gases - at the same time, it can be assumed that thermal degradation of VOCs occurs at higher temperatures; therefore, the finest PM could be with decreased levels of absorbed hazardous substances (Madureira et al., 2016; Khan et al., 2010).

With regard to the application of the shower cooler (PM data plotted into Fig. 3), the most important of all is the fact that there has been a significant reduction in the PM released - regardless the PM size, the quantities are no longer in thousands but in hundreds. Data visualization also reveals that bubbling and subsequent showering of pyrolysis gas via water relatively efficiently captures large and medium PM, and its quantities remain relatively unchanged regardless of the process temperature. This is in line with latest high-end filtration units (Andrew, 2018; Hossain and Davies, 2013); which, however, are unaffordable in developing countries. Nevertheless, the finest PM penetrates more easily through the showers. Increasing the volume of dispersed water did not show a significant increase in its removal either. It is worth noting that increased temperature also increases the amount of the finest PM released, which is in agreement with Fig. 2. If, however, the above-stated hypothesis regarding the reduction of harmful substances at higher operating temperatures proved valid, this partial drawback would not be so important (Kan et al., 2016; Madureira et al., 2016).

Hawken et al. (2013) claim that: 1) humans mine or grow or harvest materials whose daily flow per person averages 20 times the person's weight; 2) at least 93 % of this massflow is lost in extraction and manufacturing, with no more than 7 % getting into products; 3) 6/7 of those products, by mass, are consumer goods that are thrown away after a single or no use; 4) thus, only 1 % of the original mass is retained in durable products; 5) of the material in those durable products, only about 1/50 later returns to produce more value, either as compost or from recycling and remanufacturing. Moreover, much of the waste is toxic. Following the above, it is advisable to support the production of a cement substitute from fermentation residues via the application of simple shower coolers as depicted in Fig. 1. Provided it is equipped with a centrifugal pump for water (800W should provide the performance of 1.5 to 2 m^3h^{-1} and the pressure of 0.4 MPa, operating with water at a temperature of up to 90°C) and ventilator that operates some 200 m^3h^{-1}, the pressure loss in the shower cooler showed to be some 250 Pa. In the developing world, this construction can be realized within 1 month for an estimated $ 1,000.

Conclusion:

An undemanding shower cooler design was designed that allows reducing the amounts of PM released during the pyrolysis of fermentation residues into a cement substitute by one decimal place. There is a certain threshold where an increased intensity of pyrolytic gas washing no longer results in increased PM captures. The finest PM is the most difficult to capture via the showering technique. There is an indirect assumption that the finest PM that is formed during pyrolysis at higher temperatures is less dangerous provided that higher temperatures break down some of the hazardous compounds that are trapped on the PM.

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VS	COD	LP	RP	pH
69 ± 7	2 088 ± 105	9 ± 2	24 ± 4	7.7 ± 0.2

Tab. 1: Biochemical analysis of fermentation residues, where: VS = volatile solids (%); COD = chemical oxygen demand (mg L^{-1}); LP1 = labile pool of carbon (%); resistant pool of carbon (%), all $n = 12$; $\alpha = 5$.

As	Cd	Cr	Cu	Hg	Mo
0.21 ± 0.15	ND	15.52 ± 1.64	12.09 ± 1.57	ND	ND
Ni	Pb	Zn	Ca	$Mg_{(MgO)}$	$P_{(P2O5)}$
2.43 ± 0.47	1.02 ± 0.63	31.10 ± 3.16	23.44 ± 6.91	16.75 ± 3.39	48.95 ± 4.77

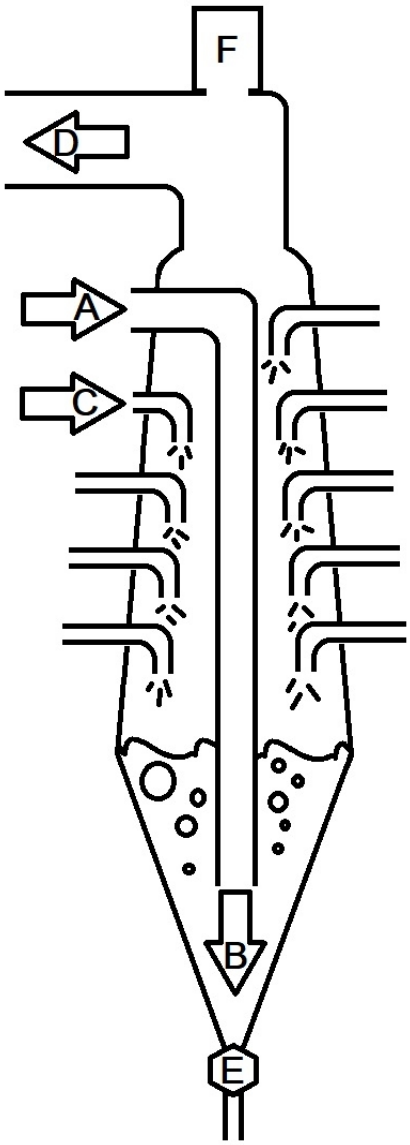
Tab. 2: Analysis of fermentation residues on heavy metals and nutrients, where Ca is expressed as CaO; Mg as MgO and P as P_2O_5, all $n = 12$; $\alpha = 5$.

Fig. 1: Schema of the shower cooler, where: A= pyrolysis gas inlet pipe; B= water washing; C = water showering; D = removal of scrubbed pyrolysis gas; E = water recycling; F = fan to maintain pressure.

Fig. 2: Visualization of PM analyses obtained from standardized UHL–07 pyrolysis unit that was not equipped with the shower cooler, where: the optimal fitting equation was found z = a + bx^0y^1 + cx^0y^2 + dx^1y^0 + fx^1y^1 + gx^1y^2; fitting target of lowest sum of squared absolute error = 2.53E+04 a = -4.93E+01 b = 1.02E+02 c = -7.67E+00 d = 2.18E+00 f = -7.53E-01 g = 5.34E-02; degrees of freedom (error) = 14; degrees of freedom (regression)= 5 Chi-squared: 25366.65; R-squared: 0.95; p-value: 6.44e-09; root mean squared error = 35.61

Fig. 3: Visualization of PM analyses obtained from standardized UHL–07 pyrolysis unit that was equipped with the shower cooler, where: the optimal fitting equation is the same as in Fig. 2; fitting target of lowest sum of squared absolute error = 1.32E+04 a = 1.14E+01 b = -0.51E+04 c = 1.24E+02 d = -0.31E-02 f = -2.09E+03 g = -2.44E-01; degrees of freedom

(error) = 14; degrees of freedom (regression)= 5 Chi-squared: 12.30; R-squared: 0.96; p-value: 3.03e-19; root mean squared error = 12.05.



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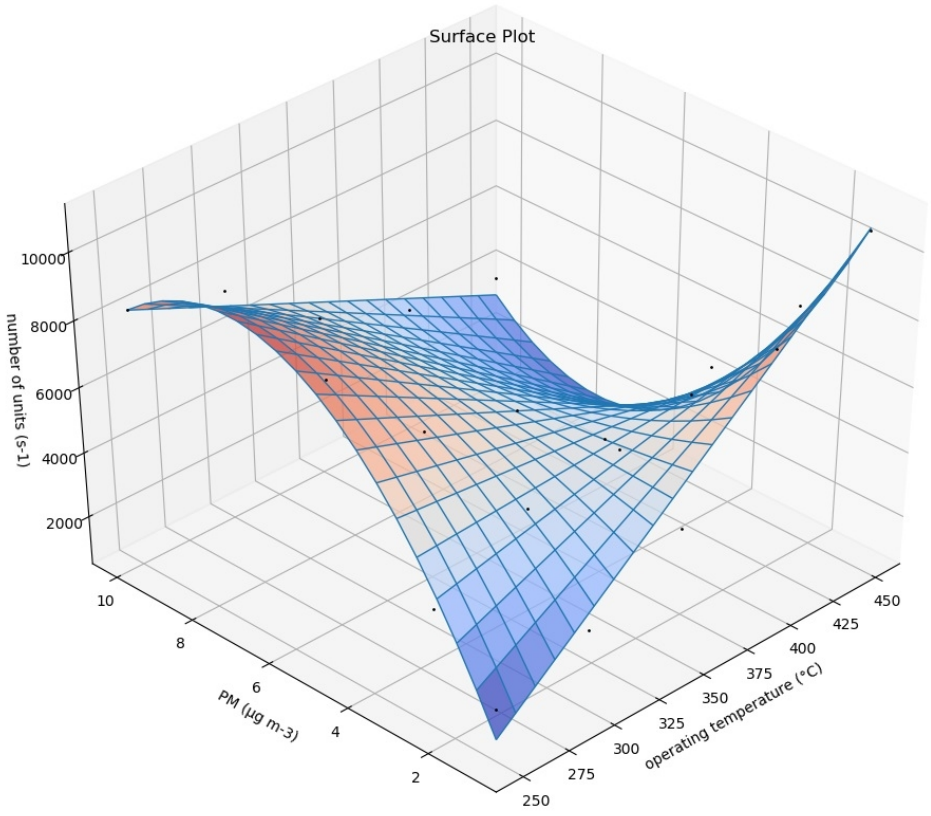
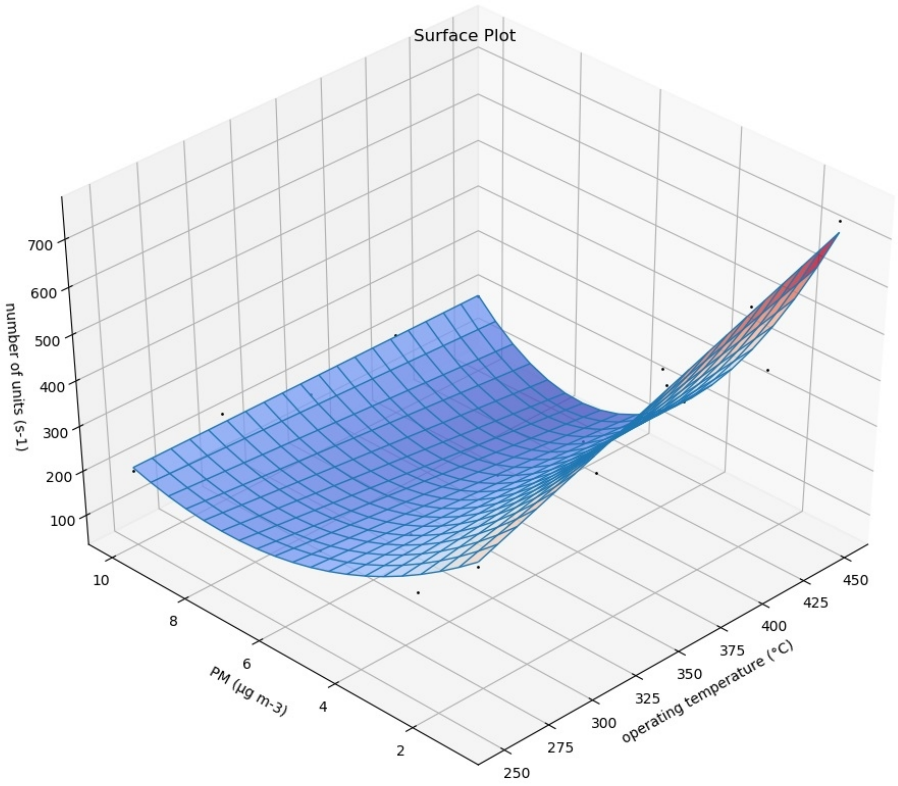
128x296mm (96 x 96 DPI)



Visualization of PM analyses obtained from standardized UHL–07 pyrolysis unit that was not equipped with the shower cooler, where: the optimal fitting equation was found z = a + bx^0y^1 + cx^0y^2 + dx^1y^0 + fx^1y^1 + gx^1y^2; fitting target of lowest sum of squared absolute error = 2.53E+04 a = -4.93E+01 b = 1.02E+02 c = -7.67E+00 d = 2.18E+00 f = -7.53E-01 g = 5.34E-02; degrees of freedom (error) = 14; degrees of freedom (regression)= 5 Chi-squared: 25366.65; R-squared: 0.95; p-value: 6.44e-09; root mean squared error = 35.61

250x211mm (96 x 96 DPI)



Visualization of PM analyses obtained from standardized UHL–07 pyrolysis unit that was equipped with the shower cooler, where: the optimal fitting equation is the same as in Fig. 2; fitting target of lowest sum of squared absolute error = 1.32E+04 a = 1.14E+01 b = -0.51E+04 c = 1.24E+02 d = -0.31E-02 f = -2.09E+03 g = -2.44E-01; degrees of freedom (error) = 14; degrees of freedom (regression)= 5 Chi-squared: 12.30; R-squared: 0.96; p-value: 3.03e-19; root mean squared error = 12.05.

259x218mm (96 x 96 DPI)