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BIOMATERIALS - BASED COATINGS WITH INDOOR EMISSION CONTROL PROPERTIES

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Abstract

At the international level, current concerns of scientists have been observed, through which concerted actions are being taken, both in the spirit of developing the circular economy, and to reduce carbon dioxide emissions. To harness synergistically the potential of the two global challenges, our work has focused on developing innovative construction products by integrating rice husks and hemp stalks, two natural agro-industrial by-products. Biomaterials - based coatings with responsive potential have been obtained, characteristics that have been confirmed by verifying the innovative coatings through their indoor emission control properties by means of TVOC emission, CO_2 and/or CO absorption capacity. From the comparative analysis of the way in which the three responsive characteristics are correlated for each tested product, it is highlighted the importance that the distribution of the nature and size of the pores in the structure of the respective materials has on the mechanisms underlying the mentioned characteristics of the obtained products. Further research is necessary to specify and capitalize on the specific mechanisms of this type of innovative coatings.

Key words*: circular economy, climate change, indoor air quality, natural agro-industrial by-products.*

INTRODUCTION

Indoor air quality (IAQ) is one of the essential concerns internationally recognized, the growing attention paid to it being based on the increase in the time people spend indoors (Klepeis et al., 2001; Hu et al., 2017; Yuan et al., 2023; Gonzalez-Martín et al., 2024), together with the higher level of pollutants found in indoor air compared to outdoor air (Montaluisa-Mantilla et al., 2023; Gonzalez-Martín et al., 2024).

This concern has grown significantly, especially as a result of the fact that the serious effects of indoor air pollution on human health range from allergies and asthma to high-morbidity diseases such as ischemic heart disease, respiratory diseases or cancer (Gonzalez-Martín et al., 2024; Hilly et al., 2024), with 3.8 million deaths recorded worldwide annually (World Health Organization, 2018).

One of the categories of pollutants with which indoor air pollution is associated is the class of volatile organic compounds (VOCs), a group that includes a wide variety of compounds, continuously growing as new sources of pollution are identified (Hu et al., 2017; Gonzalez-Martín et al., 2021). Poor degradability, severe toxicity and high volatility are the main characteristics of VOCs, having negative effects on the environment and human health. Long-term exposure to VOCs can have a wide range of consequences, from skin irritation, dizziness and fatigue, to impaired lung function (Morin et al., 2020; Caron et al., 2020), leukemia and symptoms related to sick building syndrome (Huang et al., 2021; Gan et al., 2023). Finishing products and furniture elements represent the most common and studied sources of VOCs indoors, the level of total volatile organic compounds (TVOC) constituting an element of the risk regarding the effects on the health of the occupants, used as a general indicator for assessing indoor air quality.

Another significant category of pollutants is the class of volatile inorganic compounds, carbon oxides (CO and CO_2), ozone (O_3), nitrogen oxides (NO and NO_2), sulfur dioxide (SO_2) being among the most relevant types of

pollutants in the indoor environment (Bernstein et al., 2008; Năstase et al., 2018).
In indoor spaces, CO can be present through transfer from outside air or from household activities such as combustion processes (cooking, heating with wood etc.). Among the harmful effects that CO can have on human health are headaches, dizziness, nausea, on pregnant women, young children, the elderly and people with heart and lung diseases being more sensitive to the effects of CO.
Carbon dioxide (CO_2) is an inorganic compound, naturally present in the earth's atmosphere, the predominant source, in non-industrial indoor environments, being human metabolism. Exhalation produces 37 g h^{-1} per person (Yuan et al., 2023), studies showing that high levels of CO_2 can have a significant impact on both productivity and personality psychology (Azuma et al., 2018).
Therefore, large amounts of various categories of pollutants can accumulate in the indoor air of a building, and such dangerous substances pose a threat to human health, necessitating detailed research on how to eliminate them efficiently and economically (Hu et al., 2017).
The alarming increase in global CO_2 emissions, which represent 70% of the total volume of greenhouse gases (Khosrowshahi et al., 2023), constitutes a good premise for the development of a new research direction, through which the construction sector can contribute to the reduction of CO_2 and VOC emissions.
In this context, the paper presents the research carried out for the development of biomaterials-based coatings with responsive potential and verifying their indoor emission control properties. The products were designed through an approach that combines basic ideas, principles, materials and conditioning, as follows: integrating the principle of poly-ionic liquids and nano-fluids, by using a set of raw materials, considering that each of them activates a distinct possibility of increasing CO_2 absorption; the use of synthetic polyvinyl acetate binder, as a polymeric solvent for CO_2 absorption, and to improve the mechanical performance of responsive coatings; integration of solid materials in powder form, with micrometric-sized particles, namely lime, soda-lime (absorbent material, traditionally used in the medical field, in the present research acting as an alkaline solvent for CO_2 absorption), rice husks and chopped hemp stems, all creating in the presence of water a type of nano-fluid for CO_2 retention. The method used, the results obtained and the conclusions are described below.

MATERIALS AND METHODS

Thirteen biomaterials - innovative coatings with compositionally responsive potential - were obtained and in terms of their response to specific requirements. This consisted of determining the indoor emission control property, by monitoring three characteristics, namely TVOC content emission, CO_2 absorption capacity and CO absorption capacity.
The components of the innovative products were: synthetic binder, lime, corn starch, soda-lime, rice husks and hemp stalks.
The products were designed considering the following criteria: different percentage content of synthetic binder; mixing time of the biomaterial, the products being mixed manually, and nature of the plant component, even, rice husks, hemp stalks or 1:1 rice husks: hemp stalks mixture.
The studied biomaterials-based coatings, noted EC1 - EC13, were composed of two successive layers, applied by brush on a plasterboard surface, with dimensions of 20 x 20 x 1 cm. Each bilayer biomaterial-based coating variant was subjected to emission monitoring, inside of S2 experimental stand (Figure 1, left), with DirectSense IQ-610 equipment (Figure 1, right), also used in other international studies (Canha et al., 2017; Johnson et al., 2018; Majd et al., 2019), according to the monitoring protocol described in Vasile et al. (2023), for 24 and 48 hours.



Figure 1. S2 experimental stand (left) and DirectSense IQ-610 equipment (right)

RESULTS AND DISCUSSIONS

The interpretation of the experimental results obtained for the innovative products tested in terms of TVOC emissions was carried out by analysing and comparing the graphical representations of the TVOC emission profiles (Figure 2) and the average values of TVOC concentrations, presented in Table 1, all these results being compared after the same two time points, namely the first 24 hours and the 48 hours of monitoring.

The interpretation of the experimental results regarding CO_2 absorption, respectively CO absorption, was achieved by analysing and comparing the graphical representations regarding the variation of the CO_2/CO concentration (Figures 3 and 4), as well as the average values of the CO_2/CO concentrations presented in Table 1, for all tested products, all these results being compared according to the same time points, namely the first 24 hours and the 48 hours of monitoring.

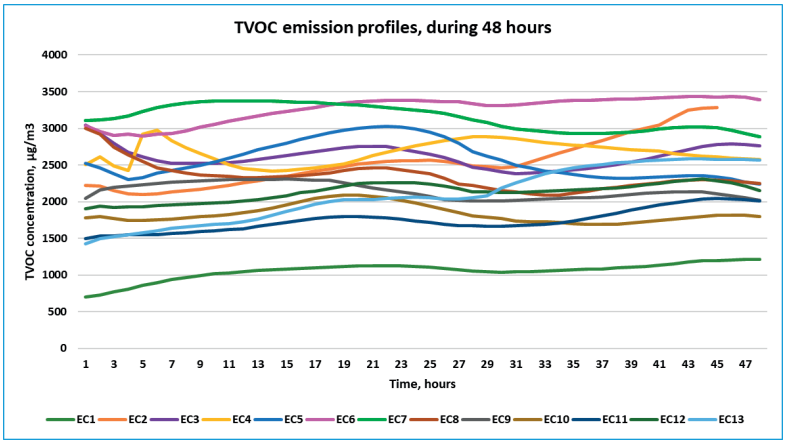


Figure 2. TVOC emission profiles for all monitored products, for 48 hours

Table 1. Average values of TVOC, CO_2 and CO concentrations, at 24 hours and 48 hours of monitoring

Product	Average TVOC concentration values, µg m^{-3}		Decrease in TVOC concentrations µg m^{-3}	Average CO_2 concentration values, ppm		CO_2 absorption, ppm	Average CO concentration values, µg m^{-3}		CO absorption, µg m^{-3}
	24 h	48 h	48 h	24 h	48 h	48 h	24 h	48 h	48 h
EC1	999	1054	-	354	349	1.41	511	438	14.2
EC2	2308	2579	-	350	349	0.28	383	325	15.14
EC3	2667	2616	51	345	345	-	392	345	11.99
EC4	2595	2667	-	361	355	1.66	474	400	15.61
EC5	2689	2570	119	365	368	-	425	367	13.65
EC6	3145	3264	-	374	370	1.07	445	381	14.38
EC7	3294	3150	144	352	353	-	269	263	2.2
EC8	2470	2344	126	363	363	-	358	274	23.46
EC9	2236	2150	86	363	364	-	142	151	-
EC10	1899	1834	65	371	370	0.27	315	239	24.12
EC11	1659	1744	-	376	375	0.28	-	-	0.76
EC12	2064	2131	-	354	360	-	226	191	35
EC13	1784	2090	-	369	367	2	127	115	12

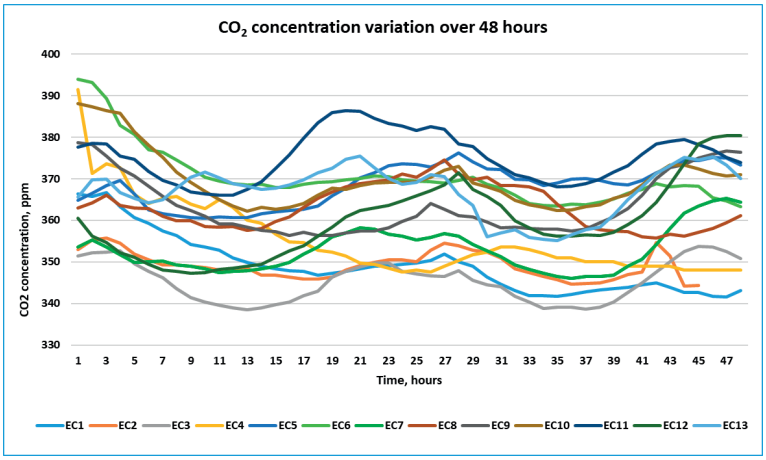


Figure 3. CO_2 concentration variation for all monitored products, for 48 hours

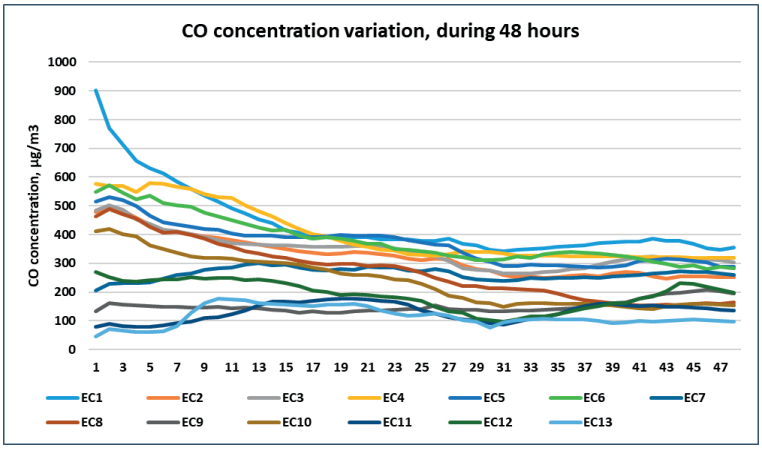


Figure 4. CO concentration variation for all monitored products, for 48 hours

Analysing the experimental results obtained as well as the different forms of their processing, regarding the TVOC emissions of the potentially responsive products tested, the following observations were made.

In general, the TVOC emission profiles are relatively similar to each other, especially in the first 24 hours of monitoring.

During the 48 hours of monitoring, it is observed that half of the products (EC3, EC5, EC7, EC8, EC9, EC10) have downward trends in TVOC emissions, while the others are slightly upward.

From the analysis of the recipes, it results that this evolution could be determined by structural changes in the biomaterials, in correlation with the mixing time, most of the products with a downward trend in emissions having 1 minute of mixing, comparing to those having 3 minutes of mixing.

Analysing the products both as a trend of emission profiles but especially as average values of TVOC concentrations, the product EC1 (with a low content of synthetic binder and rice husks) is particularly noteworthy, which, although with a slightly upward trend, maintains the lowest average values of TVOC concentrations emitted.

Regardless of the quantitative compositional particularities of the biomaterials, it is considered that, in situations where the concentrations of TVOC emitted are lower or decreasing, the favourable influence of three determinant factors are manifested synergistically: the low content of synthetic

binder, the highest possible content of rice husks or/and hemp stalks and the 3-minute mixing time of the biomaterial, before its application on the support surface.

The favourable influence of the presence of the plant component in the biomaterial, in terms of reducing TVOC emissions, is clearer in the case of incorporating the rice husks, or a 1:1 rice husks: hemp stalks mixture, it being desirable to incorporate as large a quantity of these natural materials as possible, within limits that do not affect the workability of the mixture.

Regarding the evolution of CO_2 concentrations, respectively their decrease as a result of the increase in CO_2 absorption by the tested coatings, the following were found:

Based on the individual profiles of CO_2 concentrations, respectively of the decreases in recorded concentrations, it results that the tested products have a certain responsive potential manifested through CO_2 absorption, a characteristic highlighted for seven of the products, especially in the first 14-19 hours of monitoring but also later, at the end of monitoring.

Given the general path of variation of CO_2 concentrations, with multiple irregularities, it is considered that the average values obtained are not quantitatively relevant at this stage of the research. It is noted that no obvious correlations can be established between the components/recipes of the biomaterials and the decrease in CO_2 concentrations during monitoring, and it is assumed that dimensional differences in the porous structure of the coatings could provide clarification, with future research being necessary in this regard.

It is appreciated that there are elements that could lead to an improvement in the responsive potential of the tested materials, regarding the increase in CO_2 absorption from the atmospheric air of the enclosure, mainly, mechanical mixing, increasing the mixing time, and applying protection to larger support surfaces.

The evolution of CO concentrations, respectively their decrease as a result of the increase in CO absorption by the tested coatings, showed the following:

Based on the individual profiles with a downward trend in CO concentrations and the variation in the average values of CO concentration, in the first 24 hours and during the 48 hours of monitoring, it results that the tested products also have a certain responsive potential manifested through CO retention.

The highest decrease in CO concentration (35%) was recorded for the EC12 product, with a content of 58.73% synthetic binder, 3.17% hemp stalks and 12.68% soda-lime, the latter two components supporting the responsive characteristic of the product in terms of CO absorption. Similarly, a significant decrease in CO concentration (24.12%) was recorded for the EC10 product, with a content of 50% synthetic binder, 3.57% hemp stalks and 14.28% soda-lime.

Contrary to expectations, a similar decrease in CO concentration (23.46%) was recorded for the EC8 product, a control product, therefore without soda-lime content, with a slightly higher content of synthetic binder (51.5%). It is considered that the effect observed was generated synergistically by the presence of lime and the 1:1 rice husks: hemp stalks mixture.

During the 48 hours of monitoring, seven of the thirteen tested products had CO concentration developments that indicated decreases between 11.99-15.61 $\mu g\ m^{-3}$ CO, values that indicated, from a compositional point of view, the synergistic effect of the presence of lime, the soda-lime product with a CO_2 absorbent role, and a plant component, mainly rice husks (1.85-3.47%), respectively only 1.75% hemp stalks. However, it is appreciated that the evolution of CO absorption phenomena, similarly to the case of CO_2, is based on the distribution of the nature and size of the pores in the structure of the resulting materials, but further research in this regard is necessary.

From the comparative analysis of the data in Table 1, regarding the way in which the three responsive characteristics (TVOC emission, CO_2 and/or CO absorption capacity) are correlated for each tested product, it is considered that this also highlights the importance of the distribution of the nature and dimensions of the pores in the structure of the respective materials, on the mechanisms underlying the emission and/or retention capacity of the responsive products obtained and studied.

Further research is needed to characterize and control the specific mechanisms of this type of innovative coatings.

CONCLUSIONS

Thirteen innovative biomaterial-based coatings, composed of synthetic binder and two natural agro-industrial by-products, were studied in terms of their response to specific requirements that consisted of monitoring the emission of total volatile organic compounds (TVOC) content, carbon dioxide (CO_2) and carbon monoxide (CO) absorption capacity.

Based on the experimental results obtained, it is observed that most of the products have downward trends in TVOC emissions, while some profiles remain slightly upward, an evolution that could be determined by morphological and structural changes in the biomaterials.

Regardless of the quantitative compositional particularities of the biomaterials, it is considered that, in situations where the emitted TVOC concentrations are lower or decreasing, the favourable influence of the low content of synthetic binder and a significant content of vegetable component (rice husks, hemp stalks or 1:1 rice husks: hemp stalks mixture) is manifested synergistically.

Starting from the experimental results obtained regarding the evolution of CO_2 concentrations, based on individual profiles, it can be considered that some of the tested products have a responsive potential manifested through CO_2 absorption, especially in the first 14-19 hours of monitoring and subsequently, at the end of monitoring.

Based on the individual profiles with a downward trend in CO concentrations and the variation in the average values of CO concentration, in the first 24 hours and during the 48 hours of monitoring, it results that the tested products also have a certain responsive potential manifested through CO retention.

From the comparative analysis of the way in which the three responsive characteristics (TVOC content emission, CO_2 and/or CO absorption capacity) are correlated for each tested product, it is considered that this also highlights the importance that the distribution of the nature and size of the pores in the structure of the respective materials has on the mechanisms underlying the mentioned characteristics of the innovative responsive products obtained and studied. Further research is needed to customize, control and capitalize on the specific mechanisms of this type of innovative products/coatings.

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