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Short Review on the Feasibility Assessment of the Recycling of Oil Refinery Wastes in Cementitious Composites

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The issue of high CO_2 emission is significant in terms of climate neutrality aspiration. Overall, cement and aggregate production play crucial role in global CO_2 emission. Researchers conduct investigations regarding their partial replacement with various waste materials. Currently, enormous amounts of oil refinery wastes (ORW) are generated worldwide, due to the unquestionable significance of petrochemical industry. Since they contain various hydrocarbon compounds and heavy metals, their disposal, in many cases, is limited to landfilling. This might possess a threat to our environment and health, as undesired chemical contaminants release to soil and water, while dust might be moved around by wind. Based on literature knowledge, the recycling of ORW in cementitious composites seems to be reliable research direction, providing potential to bind contaminants in hardened matrix and address aforementioned problem. Waste fluid catalytic cracking catalyst was mainly examined, but still without industrial implementation. Plenty of other ORW were not investigated, providing many research gaps. Considering the above, this paper presents the current state of knowledge, limitations, opportunities, and future development trends, concerning a comprehensive approach to the feasibility assessment of incorporating ORW in cementitious composites. It is vital that appropriate use of ORW might lead to prepare high quality cementitious composites revealing 30 % higher compressive strength. Their adverse impact, e.g. on rheological properties, might be mitigated by tailoring formulations of composites. Their applicability performance might be improved by purification, which has been highlighted in following paper.

1. Introduction

Solving the problem of global warming and co- related issues arising mainly from high CO_2 emission is currently of the highest priority for our planet. At present, the construction sector, despite its undeniably significance for society development, is one of the most environmentally affecting industries. It is mainly caused by the significant CO_2 emission associated with the manufacturing of cement and aggregates, which are crucial constituents of cementitious composites, a major construction materials. Their manufacturing is based on processing natural, non- renewable resources, contributing to their depletion. Accordingly, each 1 t of cement consumes approximately 1,500 kg of raw materials (Ahmad et al., 2021), and generates emission of approximately 600-700 kg of CO_2, which globally accounts for 5-8 % as indicated by the European Cement Association (ECA, 2022) and in the work of Benhelal et al. (2021). For that reason, this sector is currently shifting away from raw materials, in favour of more environmentally friendly, alternative ones, i.e. various wastes. Only few of them have been already consolidated in large scale industrial application (Costa and Marques, 2018). Recently, oil refinery wastes (ORW), resulted from oil refining processes and co- related activities, attracted researchers attention as a promising materials capable of being recycled in cementitious composites, as it has been presented in Figure 1. At present, their recycling is significantly limited as they contain various hydrocarbon compounds (coke deposits) and heavy metals and, instead of being reused, their massive amounts require costly utilization methods, such as landfilling, contributing to general degradation of natural environment.

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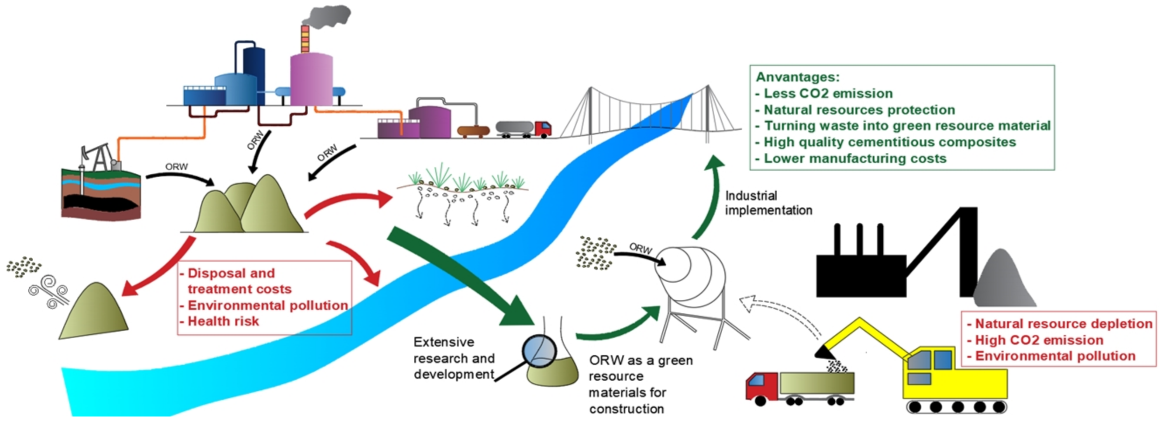

Figure 1: General idea of the recycling of ORW in cementitious composites

According to the literature, the incorporation of spent fluid catalytic cracking catalyst, which is predominately researched ORW, in mortars and concretes, apart from maintaining high quality of composites, might also allow the binding of heavy metals in their hardened state matrix. Additionally, coke deposits might be efficiently eliminated from the structure through the appropriate chemical methods. It is vital that such recycling of ORW provides a key opportunity to implement a circular economy, ensuring numerous benefits both for construction and petrochemical sector. Many research gaps still deny them from industrial implementation. Considering the above, the attention should be put to systematised the current state of knowledge concerning a comprehensive approach to the applicability of various ORW in the construction industry. For this purpose, literature review was carried out for the scope of this paper. As such, some not investigated ORW, e.g. sulphur recovery catalysts, were pointed out, limitations and opportunities were highlighted, and some research gaps were determined. Based on the latter, some new research directions were finally proposed. These are mainly set for more extensive research to be applied on ORW, including their purification methods and their unstudied impact on complete properties of composites. Fulfilling them might be crucial towards industrial implementation of ORW. It should be mentioned that the recycling of ORW might be also limited considering formal issues as waste management requires appropriate national permits.

2. Review methodology and results

To illustrate the research interest on the recycling of ORW in the construction sector, the number of publications comprising the keyword “concrete”, “oil”, “refinery”, “waste”, or their various combinations in abstract, tittle or keywords were checked in the ScienceDirect Elsevier database and showed in Table 1.

Table 1: The Number of articles comprising related keyword or their combinations in abstract, title or keywords from the ScienceDirect Elsevier database. [Date: 23.03.2022]

Keyword	The number of publications	Keyword	The number of publications
1 keyword		3 keywords	
Concrete	75,777	Concrete + oil + refinery	17
Oil	186,502	Concrete + oil + waste	243
Refinery	7,074	4 keywords	
Waste	150,322	Concrete + oil + refinery + waste	8
2 keywords			
Concrete + oil	801		
Concrete + refinery	43		
Concrete + waste	5,409		
Oil + refinery	3,081		

The number of articles containing the only one of following keywords as “concrete”, “oil” or “waste” is very high. Despite this, when their combinations are searched, the number of related articles decreases significantly. Figure 2 presents the number of publications devoted to the applicability of particular ORW in cementitious composites. For this purpose, a formula was used that searches words ‘refinery waste’ in the title, keywords or abstract and ‘*oil* refinery *waste* AND (concrete OR mortar)' anywhere in the article. The phrase ‘*oil refinery* *waste'* searched anywhere in the article was replaced with several examples of popular ORW. As such, the highest number of results were obtained for hydrocarbon waste, oil sand waste and spent catalyst.

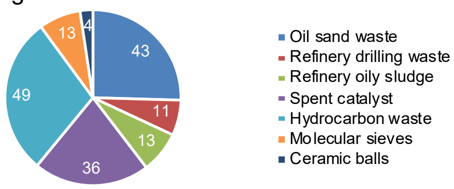

Figure 2: The number of publications devoted to the applicability of particular ORW in cementitious composites

3. Recent state of art

3.1 Effects of using ORW on the properties of fresh concrete mix

Table 2 summarizes the results of the state of art in the subject of the effect of using oil refinery waste in cementitious composites on the properties of fresh concrete mix.

Table 2: Effects of using ORW on the properties of concrete mix

Type of ORW	Application	Properties tested	Results	Reference
Spent Catalyst	Substitute of fly ash	Consistence	The inclusion of ECAT reduces the density of the concrete mix	(Costa and Marques, 2018)
Cracking catalyst	Replacement of 15 % wt. of cement	Workability	Decrease in workability in relation to a control mortar for all water/ binder ratios tested	(Payá et al., 2013)
Spent catalyst	Cement subtype in the amount of 10 %, 20 % and 30 % of cement in SCC mixtures	Flowability	Larger slump flow diameters were obtained which means greater flowability	(Laginha et al., 2014)
		Viscosity	All mixtures satisfy the requirement sets out in European standard EN 206-9 in a research	
		Passing ability	The mixture with the addition of 30 % FCC did not meet the L-box requirements for SCC concretes	
Waste fluid catalytic cracking catalyst	Cement subtype in the amount of 10 %, 20 % and 25 % in SCC mixtures	Flowability	The addition of 10 % FCC resulted in larger slump flow diameters than addition of 20 % and 25 %	(António et al., 2013)
		Passing ability	All FCC mixtures obtained the same grade as the reference mixture - PL2 and PJ2	
Spent catalyst (ECat and EPCat)	Cement subtype in the amount of 5 %, 10 % and 15 %	Flowability	The flowability value decreased with a higher percentage of cement substitution	(Su et al., 2000)
		Bleeding	Bleeding decreased with greater percentage of cement substitution. Also bleeding in EPcat is less significant than that in Ecat	
		The setting time	The setting time shortens as the degree of substitution increases	
Fluid catalytic cracking catalyst residue	Cement subtype in the amount of 6 %, 10 %, 15 % and 20 %	Autogenous shrinkage	The shrinkage of all samples was similar	(Payá et al., 2001)
		Flowability	The inclusion of ECat as a sand substitute resulted in a less flowable mix	
		Setting time	Setting time has decreased	
Catalytic cracking catalyst.	Cement subtype in the amount of 15 % and 30 %.	Flowability	Notice decrease in workability for all tested w/b ratios. In addition, there is no difference between the original FC3R and the grounded FC3R	(Payá et al., 1999)
Spent fluid cracking catalyst	Cement subtype in the amount of 3 %, 6 % and 12 %.	Workability	Adding the catalyst reduces the workability	(Sena da Fonseca et al., 2018)
Organic fraction of oil waste	As a modifying admixture	Workability	The addition of OFOW plasticizes freshly mixed concrete	(Asim et al., 2021)
Spent catalyst	Substitute of sand with 5-15 %	Workability	A decrease in workability and compaction was achieved	(Su et al., 2001)

On the basis of Table 2, it could be seen that the impact of ORW on the properties of fresh concrete mix was mainly assessed through the study of flowability and workability. The addition of the catalyst reduces mentioned above values.

3.2 Effects of using oil refinery waste on physical and mechanical properties of concrete

Table 3: Effects of using ORW on physical and mechanical properties of concrete

Type of ORW	Application	Properties tested	Results	Reference
Waste Oil-Cracking Catalyst	Replacing cement with 5 %, 10 % and 15 % of wFCC catalyst	Water absorption	All mortars containing the catalyst showed lower water absorption than the reference mortar	(Costa, et al., 2014)
		Chloride migration	The use of a waste catalyst increases resistance to chloride penetration	
		Carbonation	The use of a waste catalyst makes it more susceptible to carbonation	
Spent catalyst	Replacement of 25 % by cement	Compressive strength	Compressive strength at both 7 and 28 days is higher with the addition of a catalyst	(Pacewska et al., 1998)
		Flexural strength	Similar bending strengths were obtained	
Catalytic cracking catalyst	Replacement of cement with 6 %, 10 %, 15 % and 20 %	Flexural strength	Early strength decreases with increasing catalyst substitution, after 7 days it is of a similar value, after 28 it decreases with the amount of FCC	(Payá, et al., 2001)
Catalytic cracking catalyst	Replacement of cement with 15 % and 30 %	Flexural strength	Flexural strength was similar to or greater than the control sample, except for the unmachined mineral admixture	(Payá et al., 1999)
	Replacing sand	Compressive strength	A continuous increase in strength was observed with increasing degree of substitution	
Cracking catalyst	Replacement of 15 % by cement	Compressive strength	The compressive strength was higher than that of the control mixture	(Payá et al., 2013)
Spent fluid cracking catalyst	Replacement of 3 %, 6 % and 12 % by cement	Carbonation	The depth of carbonation increased as the level of replacement of cement by the SFCC increased	(Sena da Fonseca et al., 2018)
		Chloride migration	Chloride penetration resistance improved with increasing SFCC replacement of cement	
Spent catalyst	Replacing cement with 15 %	Flexural strength	Similar values were obtained after 3, 7, 14 and 28 days to the reference sample	(Bernabeu et al., 2009)
Spent fluid catalytic cracking catalyst	Replacement of cement with 10 % in combination with 20 % FA	Compressive strength	Compressive strength after 7, 28 and 90 days higher values with the addition of FCC	(Soriano et al., 2016)
		Flexural strength	Strength values fluctuate but have similar, slightly lower values than the reference values	
Spent catalyst Ecat/Epcat	Cement substitute as 5 %, 10 % and 15 %	Compressive strength	Strength increases with the hardening time and is higher than that of samples without additives	(Chen et al., 2004)
	As sand replacement in range 5-25 %	Compressive strength	Replacing sand with 5 % and 10 % gave higher value. For higher substitution the growth disappeared	
Spent catalyst	As fine aggregate replacement with 5 %, 10 % and 15 %	Compressive strength	The addition of 5 % and 10 % ECat increased the strength. The addition of 15 % caused a significant reduction in strength	(Su et al., 2001)
		Water absorb.	Higher water absorption has been noted	

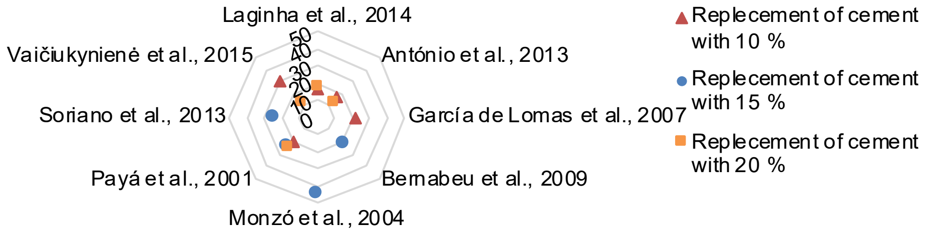

Figure 3: The impact of waste FCC catalyst on compressive strength enhancement of substituted composites

Table 3 shows that the most frequently measured properties of hardened state composites were mechanical properties. Figure 3 reveals the substitution of 15 % of cement with spent FCC catalyst led to the highest compressive strength (CS) enhancement, exceeding 20 % of the CS value of plain concrete at 28th day. When the substitution level was 10 % and 20 %, CS enhancements were found to be 15-20 % and 10-15 %. Researchers emphasize that catalysts derived from miscellaneous oil refinery plants revealed high pozzolanic activity, contributing to CS enhancement of cementitious composites (Abdolpour et al., 2021).

3.3 Investigation and improvement methods of ORW and substituted cementitious composites based on chemistry and material science

Overall, the composition, contaminants, properties and microstructure of different ORW are highly variable, mainly depending of the process form which they are withdrawn and a type of processing crude oil. The role of material science and chemistry is unpriceable considering their recycling feasibility. Understanding complete properties of ORW, such as chemical composition, particle size distribution, direct pozzolanic activity, loss on ignition, specific surface area, capillary water absorption, porosity, specific density, etc. gives grounds to predict their impact on the recycling product. This might be vital as the development of concrete technology occurs mainly by its modification, available with the use of ORW (Czarnecki and Deja, 2021). Additionally, according to the literature, appropriate chemical methods of purification of ORW, e.g. their oxidation with hydrogen peroxide, enable the elimination of coke deposits from their structure and improving their performance (Vaičiukynienė et al., 2015). By means of thermal and surface morphology analysis, the efficiency of purification process might be assessed. Consequently, based on the discussed methods and properties, various ORW, supplied both in original and activated forms, might be compared regarding their potential to be recycled in mortars and concretes. What is more, the hydration products in substituted composites might be quantified with TGA, whilst the health risk associated with heavy metals might be assessed through the toxicity test, e.g. TCLP analysis (Su et al., 2001).

4. Research gaps

Although the interest in the recycling of ORW in the construction sector is growing, they have not yet been thoroughly researched. A number of ORW have not been investigated yet. These are, e.g. waste catalysts derived from Claus and Hydrosulfreen processes or waste ceramic balls from molecular sieves. Waste fluid catalytic cracking catalyst was most commonly researched, but the works were focused mainly on the study of consistency, flowability, volume change and compressive strength of substituted composites. Autogenic shrinkage, bleeding and tensile strength were studied the least frequently. In addition, ORW were investigated mainly in their original forms, more frequently as a substitute of cement compared with sand. The purification of ORW was carried out only through oxidation. The correlation between various properties of ORW supplied in both original and modified forms and their impact on complete properties of substituted composites are not known comprehensively. As such, further research directions aimed for fulfilling the above have been clarified.

5. Conclusions

On the basis of conducted literature review, the main conclusions might be drawn as follows:

- Various ORW appear to possess huge potential to be recycled as a sustainable resource in construction sector, providing vital environmental and economical benefits both in construction and refinery sector.
- Waste FCC catalysts derived from miscellaneous oil refinery plants revealed high pozzolanic activity, contributing to compressive strength enhancement of cementitious composites.
- Apart from partial cement substitution, ORW might also allow to replace a certain level of fine aggregate in mortar and concrete formulations. For this purpose more research should be conducted.
- Considering ORW recycling feasibility, first step should be focused on studying their complete properties.
- ORW recycling in cementitious composites might allow the binding of heavy metals in their hardened matrix. Various methods of purification of ORW are found to be especially interesting. In some cases, these methods seem to be achievable in refinery, as e.g. the calcination of spent FCC catalyst in FCC unit.
- An interdisciplinary approach and collaboration between researchers from various disciplines are highly preferable towards the industrial implementation of ORW in construction sector.

Generally, three main research directions have been clarified, which are vital to be extended in the future.

- There is still a huge room for exploration regarding the recycling of ORW both as a cement and fine aggregate substitute in cementitious composites assuming different formulations and conditions.
- Broadly understood chemical methods seem to be interesting research trend, as they enable the purification of ORW, giving them additional value. Vital properties of ORW might be determined on their basis, which might be crucial for understanding their impact and tailoring formulations of cementitious composites.

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