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Literature review on the state of the art of the circular economy of Ceramic Matrix Composites

Lars Wietschel [a], Florian Halter [a], Andrea Thorenz [a], Denny Schüppel [b], Dietmar Koch [b,*]

[a] Resource Lab, Institute of Materials Resource Management, University of Augsburg, Germany
[b] Institute of Materials Resource Management, University of Augsburg, Germany

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1. Introduction

Ceramic Matrix Composites (CMC) are light weight materials with high specific strength, a high temperature resistance, and high resistance to corrosive environments [1]. Ceramic fibers embedded in a ceramic matrix thereby overcome the disadvantages of conventional technical ceramics and allow applications that go beyond the possible use of metals [2,3]. The superior properties are demonstrated by brakes based on CMC for example, which have very low wear rates and enables brakes that endure the entire service life of different applications [4], and with an appropriate remanufacturing (refurbishment) even a second or third life gets feasible (rebrake [5]. However, the production of CMC applications has a huge energy demand and depending on the CMC type, considerable volumes of high-grade silicon and ceramic or carbon fibers, which are very expensive and which are produced with a high amount of energy, are required. Preliminary life cycle assessment (LCA) results on environmental impacts of the CMC production indicate high environmental impacts in its production phase [2]. Obviously, there is a strong need to evaluate processing of CMC in the future in order to define a suitable tradeoff between properties and reduced environmental impact. Furthermore, this gives rise to the need of using CMCs as efficiently as possible and to close loops in the different life-cycle stages. The circular use of products and materials provides the opportunity to partially offset the high environmental impacts of the production phase [6,7]. The production volumes of ceramic matrix composites are still low compared to other material classes, however the annually produced volumes are steadily increasing in recent years [2]. As production volumes grow, with a certain delay increasing material volumes will reach their end-of-life, making the issue of circularity increasingly important.

This paper gives a brief overview on the current scientific discussion on the circular use options of CMCs. Chapter 2 gives a state of the literature by introducing methods for the assessment of material circularity in general and a literature review on the circular economy efforts of CMCs. In chapter 3 concludes and discusses the findings of the literature review and gives an outlook on relevant research topics.

2. State of the literature

Different strategies can be implemented to increase the circularity of materials. In the European Union, the *waste framework directive* [8] prioritizes circularity strategies: the prevention of waste by use-intensification has the highest priority, followed by the preservation of the product or components of a product through a preparation for reuse. The preservation of the materials by a recycling or downcycling is the following option. The penultimate strategy is the recovery of the embodied energy by combustion and the least option is the disposal of residues on landfills [8,9]. The prioritization of circularity strategies is based on the principle of minimizing environmental impacts and protecting the human health [8].

2.1. Methods to measure material circularity

In order to investigate all relevant flows, stocks and losses of a material throughout its service life for the assessment of its circularity, a life-cycle perspective has to be taken [10]. Two of the best-known methodologies for measuring the physical flow of materials in economies are Input-Output analysis and Material Flow Analysis (MFA) and both methods allow the monitoring of the circular economy [11]. The

* Corresponding author.
E-mail address: dietmar.koch@mrm.uni-augsburg.de (D. Koch).

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Input-Output method facilitates linking the circular economy to the economic dimension and allows to quantify material flows across different value chains [12]. Material Flow Analysis is a tool for the detailed quantification of material flows associated with the provision of a product or service to society [13]. Fig. 1 shows the usual differentiation of MFAs in the production (the processing of raw materials into intermediate products, fabrication (the production of finished products), use phase, and the end-of-life and quantitatively covers the material flows between these stages and in-use stocks. Dynamic MFAs allow the assessment of material flows over time and thereby enable the determination of material stocks in an industry or society [13]. Throughout the whole life cycle, certain shares of a material can leave the intended material path. Those materials can either be collected in a structured waste management in order to reuse it, or it can go lost by dissipation to the environment, third-party materials, or landfills [15]. On the one hand, knowledge of material losses in the individual life cycle phases allows material efficiency to be measured. On the other hand, this knowledge allows the investigation of different waste management options such as the reuse, remanufacturing, or recycling of residual material flows. Circular Economy Indicators which focus either on products or on processes can be used to simplify and standardize the measurements. Common indicators for products are for example the *collection rate* (CR), which measures how much of the in-use products are forwarded to the waste treatment, or the *end-of-life recycling rate* (EoL-RR) that quantifies the share of EoL-products that is recycled. For processes, the *recycled content* (RC) quantifies the share of secondary materials of the total input materials, or the *recycling efficiency rate* helps to evaluate the performance of a recycling process [16].

The circular use of materials is only useful, if thereby certain economic or environmental, goals are reached. While the economic evaluation based on monetary values is relatively simple, the evaluation of the environmental benefits is more intricate. Life cycle assessment (LCA) is probably the most appropriate method for the investigation of the environmental performance throughout the entire service life of a material. A main challenge in life cycle assessment applied on circular use considerations of materials is the consistent accounting of environmental benefits from re-using materials in continuous loops [17]. However, the inclusion of all possible material loops is especially important in comparative LCA of materials with different lifetimes.

Only a few material flow analysis of composite materials have been done and published so far, with a focus on carbon fiber reinforced polymer (CFRP) [13]. [18] published an MFA on global in-use stocks of CFRP in the wind power sector in order to estimate the annual volumes of CFRP waste per region. The MFA study reveals the need for launching a CFRP waste management program and thereby points out the importance of establishing circular economy strategies. The same authors published a similar MFA study on CFRP waste volumes from the commercial aeronautical sector. They conclude that the sector increasingly uses CFRP, wherefore the waste volume will increase significantly by 2050, which also indicates the importance of circularity strategies [19]. The studies show that an MFA can be an important method for the estimation of in-use stocks and end-of-life material volumes, which is important for the development of reuse, remanufacture, and recycling strategies. In combination with Life Cycle Assessment, the environmental benefits of different circularity strategies can be investigated. Hybrid studies that combine MFA and LCA are frequently found in literature on decision-making for environmentally optimal circular economy strategies [20,21]. Apart from these general aspects, one has to keep in mind that for CMC even the steps of production and fabrication are still not evaluated in terms of material footprint, necessary energy inputs, and environmental impacts. There is no literature found which describes the manufacturing of CMC, accordingly. This makes it very difficult to define material flows according Fig. 1 precisely.

2.2. Literature review CMC

To investigate the state of research on circular economy strategies of CMCs, we conducted a structure literature review on Web of Science in September 2022 under the use of the PRISMA-Method, seeFig. 2. Thereby, the work focuses on CMC containing exclusively carbon and silicon carbide as well as combinations of these materials like C/SiC or SiC/SiC due to their particular importance in the high mechanical and high-temperature sector. By combining the term 'circular economy' with 'CMC', 'ceramic composite' or 'silicon carbide' in a search string, only a couple of results can be found from which none contributes to circular economy for ceramic or other composite materials. Therefore, the search string has been adapted by exchanging 'circular economy' with the related and commonly used terms of 'reuse', 'recycling' and 'remanufacturing'. This results in 682 publications from which 602 can be sorted out by analyzing titles. The resulting 81 publications do not concern CMCs as a secondary resource, however, some of them are closely related to waste treatment. For example [22] considers SiC composites as a photochemical material for wastewater treatment while others like Zhu et al. (2021) evaluate the recycling of membranes made of ceramic composites which cannot be transferred to structural composite materials like C/SiC or SiC/SiC.

Table 1 shows the most relevant literature in context of the circular use of Ceramic Matrix Composites with its investigated re-options and the applied assessment methodologies. A promising recycling strategy for CFRP composites is to recover the carbon fiber for reuse by matrix

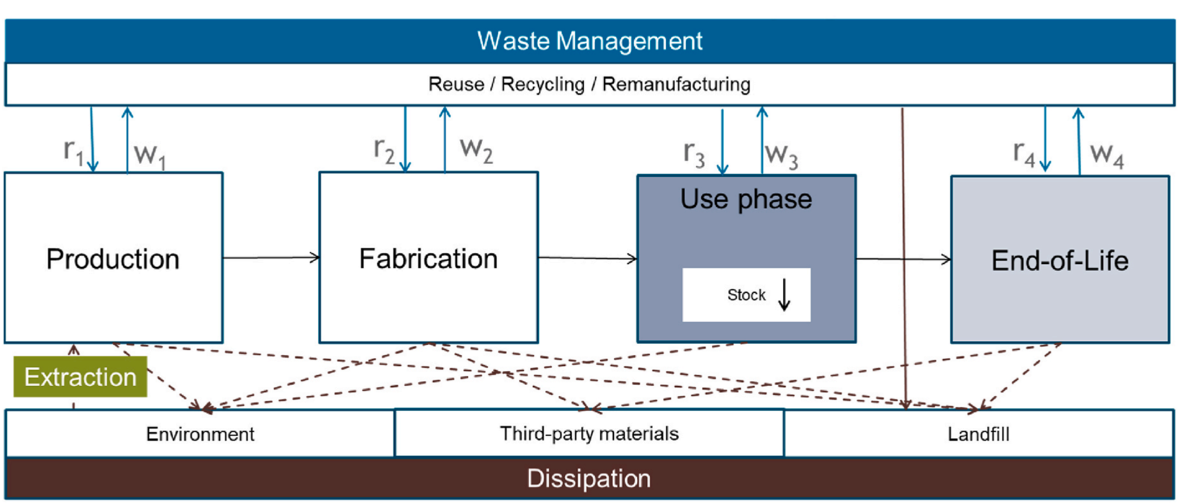

Fig. 1. General material flows and processes in material flow analysis (own depiction based on [14].

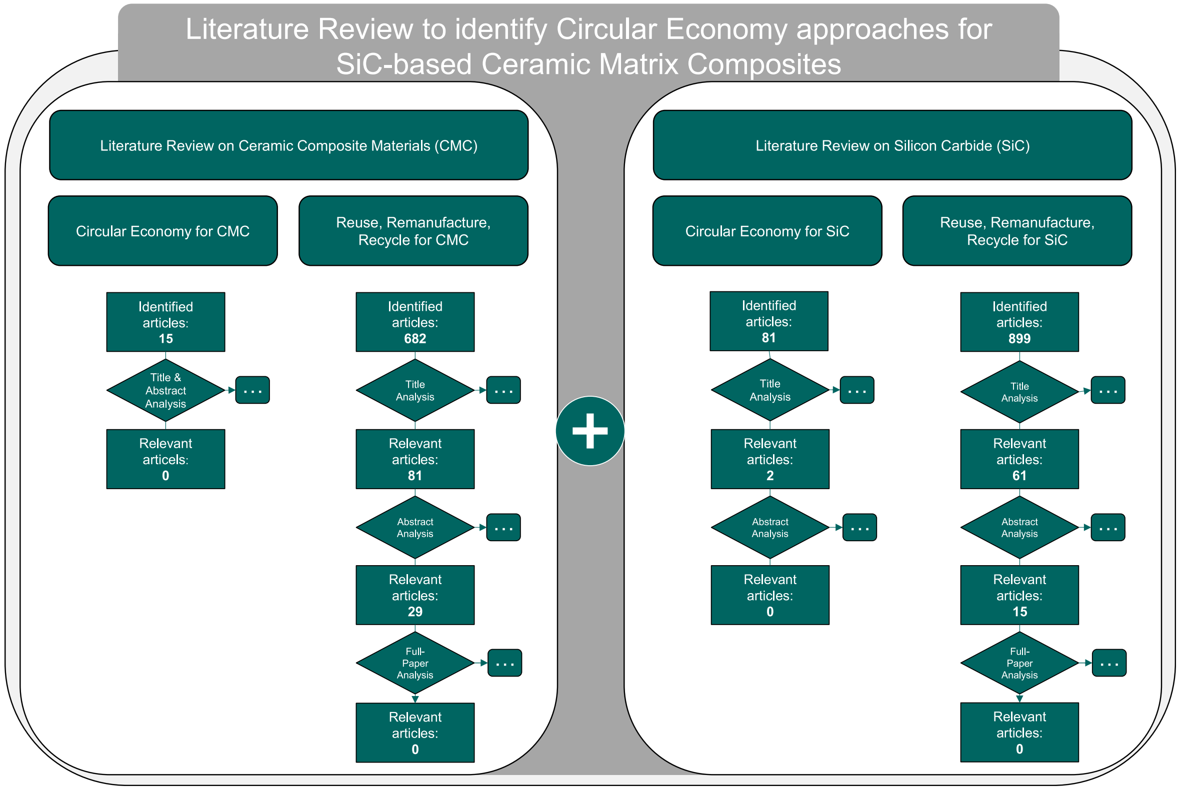

Fig. 2. Search strings with the number of identified and excluded articles.

Table 1
Literature overview with key characteristics.

#	Article	Material	Re-Option			Assessment method		
			Reuse	Remanu-facturing	Recycling	CE Indicator	(dynamic)MFA	Technical feasibility
[01]	[23]	CFRP			X			X
[02]	[24]	CFRP			X			X
[03]	[18]	CFRP			X	X	X	
[04]	[25]	CFRP	X					X
[05]	[26]	SiC			X			X
[06]	[27]	SiC			X			X
[07]	[28]	SiC			X			X
[08]	[29]	C-comp.			X			X

fiber separation. This can either be done by thermal pyrolysis [23] or chemical solvation of the polymer matrix [24]. Due to the high thermal and chemical resistance of ceramic composites, the recovery of the fibers by these methods is less promising. However, there are several studies focusing on the separation of SiC and Silicon out of powder saw dust or waste sludge especially originated from solar panel production. These studies can become relevant for CMC recycling as CMC products could be grinded at the end-of-life state to receive dust containing SiC and separated Silicon and Carbide contents [26]. for instance used a mixture of chemical and high-gravity centrifugation to separate Si from SiC to recycle kerf loss while [27] used hydrocyclone assembly to separate Si from SiC out of wire saw slurry.

To validate this option and to identify further potentials the literature review has been extended by reviewing circular economy activities for silicon carbide in general. At first the Terms 'SiC' and 'silicon carbide' were combined with the term 'circular economy'. This resulted in 81 publications from which none was relevant for SiC as a secondary material. Most of these publications focus on circular economy for other use cases like for example for WEEE [30]. Secondly, the terms 'SiC' and 'silicon carbide' were combined with the terms 'reuse' 'recycling' and 'remanufacture', which yielded significantly more matches. This search resulted in 899 publications from which 15 additional relevant papers can be extracted after title and abstract review as well as the removal of duplicates. As for the previous search string, most studies focus on the separation of SiC and Silicon from other dust particles or sludges. Two out of 15 publications address the recycling of carbon fibers. While [25] investigated the reuse of carbon fibers in brake pads [29], sketched different methods of recycling carbon composite wastes without considering silicon carbide specifically. The publication of [28] was the only identified work that directly addressed SiC recycling. The authors synthesized SiC ceramics with good mechanical properties from processing residues of the primary SiC production by the *starch consolidation method*. Summing up, the literature review on circular economy strategies for CMC materials and products did not yield relevant results. Therefore, the review is complemented by further literature sources.

The EU classifies ceramic waste as non-hazardous *mineral waste from construction and demolition* (code 12.1), without further differentiation into different kind of ceramics [31]. In Germany for example, ceramics in general are not collected separately, but are disposed of with the residual waste or as construction waste without differentiation between oxide and non-oxide ceramics [31,32]. Due to their properties, ceramics cannot be incinerated and are not biodegradable wherefore they either have to be landfilled or reused. Structural ceramics and whitewares at their end-of-life can be used as filling materials or as coarse aggregate in the concrete production [33]. Transferring these waste management strategies would lead to significant functional and economical downcycling of CMCs [15].

In contrast to the search in the scientific literature, some approaches for recycling or repairing CMCs can be identified via patents and company information. The patent of [34] deals with the recycling of silicon and carbon containing materials. SiC crystals are synthesized from secondary materials with high silicon and carbon shares into high purity SiC crystals that meet industrial requirements. This process was developed to recycle by-products and residues of the SiC production by the Acheson-Synthesis. Thereby, the process has the potential to significantly reduce the waste volume compared to the original Acheson-Synthesis. If the silicon content of end-of-life products containing SiC is high enough, the recycling process could be transferred to CMC waste [34,35]. Other patents deal with the repair of local damages of CMC parts (especially turbine applications), which extends the lifetime of CMCs [36–39]. The patent of [37] comprises the repair of local damages in different kind of melt infiltrated CMC parts (such as turbine shroud, turbine nozzle, turbine blade, combustor tile, or combustion liner) caused by pitting, delamination, cracking or other damages. In a first step, the adjacent area around the damage is removed, followed by an insertion of reinforcing fibers and a subsequent melt infiltration. The Patent of [38] comprises the reparation of local damages in CMC parts such as turbine application by converting a gas precursor with laser assisted chemical vapor deposition at the repair area to ceramic material. Another example is the reuse of ceramic brake discs by remanufacturing. The process is comparable to the PIP-process and is capable of rebuilding matrix flaking and reconditioning surface structures (rebrake 2022). This remanufacturing method could be used for further CMC components and products. Since only the matrix and not the fiber content can be recreated, this method is limited to reuse applications with equal product requirements and geometries. The example also shows that in the case of product reuse in several cycles, it is important that the following life cycles are already taken into account in the product design phase. The brief review of published patents and information of industrial companies unveils different approaches for the remanufacturing, reparation, recycling, and reuse of CMC parts. To the best of our knowledge, however no scientific research was published so far on approaches that extend the longevity of CMC-based products.

3. Discussion and conclusion

CMCs are lightweight materials with interesting technical properties that allow applications that would not be possible with conventional technical ceramics or metals. Due to its superior properties and despite its high energy demand in the production phase, CMC products face increasing interest. The circular use of CMC products could be an important strategy to increase the service life of CMCs and thereby offset the environmental impacts of the primary production. Furthermore, the increase of longevity can provide economic benefits. While patents and industrial companies unveil different approaches for the circular use of CMCs, the review of scientific literature shows that existing literature did not yet address the environmental and economic benefits associated to the circular use of CMC products. Based on the need for an elaborate circularity strategy for the energy intensive material class ceramic matrix composites and a lack of literature that addresses the reuse, repair, remanufacturing, or recycling of CMCs, the following conclusions may be drawn:

- Only a few reuse, repair, and recycling options are known for CMCs, wherefore there is a high probability that production residues and end-of-life products end up in being used as filling-material in road construction or concrete supplementary, which would mean a sever downcycling of this high-tech material class.
- MFA can support the identification of valuable waste streams that might be directly circulated or transferred into a structured waste management system that enables recycling and reuse.
- Defining a set of circular economy indicators that are suitable for CMC can help producers and users to identify potential improvements to lower the environmental impact of the composites.
- Especially for this material class with few known options for material recycling (unlike e.g. metals), reuse options should already be considered in the design phase of products.
- The separation of SiC and Silicon from end-of-life products without reuse options could be relevant for CMCs. Both the separation process itself and possible applications for the recyclates should further be investigated and tested for their suitability for additional CMC products.
- Circular design of CMC products with the aim of product standardization and component modularization, the use of homogenous materials and a design that allows an easy product disassembly is one key enabler of increasing the circularity.
- As a comparatively new material, it might be possible to implement circularity strategies for more sustainable CMC – production before standard processes have been implemented.
- Finally, Circular economy strategies could significantly reduce the high environmental impacts of CMC components and also reduce costs when the use of CMC products is intensified. The recycling of EoL products could reduce energy consumption and processing time for secondary products.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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