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FULL RESEARCH ARTICLE

3D-printed clay-based ceramic water filters for point-of-use water treatment applications

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Abstract
Water is necessary for the survival of all living beings, essential for health and should be a fundamental right for every human being. Thus, to provide enough water with good quality to populations, researchers are currently focusing on the development of innovative processes, such as ceramic water filters, that are easy to operate and inexpensive. The objective is to allow access to sufficient quality water at the point of use. To this end, one of the innovative techniques that has been applied in various fields of science is three-dimensional (3D) printing. In the present work, a 3D printer, Ultimaker 1, initially intended for plastic-based printing, was adapted for use in the manufacture of ceramic filters. These filters can find applications in water treatment for consumption at the point of use. Our study shows that this innovative concept could become a viable alternative in the near future for supplying a sufficient quantity of good-quality drinking water in developing countries.

Keywords Ceramic filter · Additive manufacturing · Water treatment

1 Introduction

Water is an indispensable resource for the survival of all living beings, essential for health and should be a fundamental right for every human. Due to rapid population growth and steady increase in cases of waterborne diseases around the world, the need for sufficient quality water is increasing and urgent [1, 2]. In most developing countries, the water supply is provided by treatment units that fail to serve the entire population [3]. In addition, water is often subject to recontamination before use due to transport from the source to a household or the method of preservation used at home. People then face public health and socioeconomic issues that drive the development of new treatment processes or new technologies that consume few external resources in terms of space needed for implementation and cost. Among these techniques, filtration is one of the most important in the water industry. A filtration process eliminates different compounds present in water according to thresholds and represents a major technological asset because it can eliminate bacteria or even viruses to achieve levels of disinfection.

Due to economic and ecological concerns, research is now focused on the valorization of local clay materials in a filtration process to meet specific needs. Indeed, recent works by many authors show the possibility of using local clay materials in the manufacture of filters for water treatment at the point of use [4, 5]. However, control of the process and the development of this technology still have some design and threshold problems, and the filters are not often adapted to the quality of the water resource to be treated.

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The use of ceramic filters is the most common because of their good performances [6–8]. They are often made locally but sometimes are marketed by specialized industries. During the manufacture and use of these filters, there are often some difficulties, such as [9, 10]:

- A small quantity of water is obtained after filtration due to the poor contact surface of the water with the filter or the relatively small filter thickness (approximately 2 L/h).
- Defects in the manufacturing conformity are not always dependent on the operator given the number of factors influencing the production of these filters (i.e., two filters manufactured under the same conditions often do not have the same performance).

Thus, we were interested in the implementation of another method of manufacturing that produces filters with reduced thickness and a large filtration surface with consistent quality. To this end, it has been a question of moving towards additive manufacturing, which is an innovative process that has found application in various fields (industry, medicine, etc.) [11, 12], but this process has not truly been exploited in the past to make water filters.

The challenge was to realize ceramic filters based on natural clays using additive manufacturing. In the literature, ceramic materials have already been made using 3D printing [13–15]. The scope of this research was to realize filters with small thickness and a large contact surface with the water to be treated. We focused our research on two points: (1) the realization of filters through additive manufacturing and (2) the exploitation of the filters for water filtration.

2 Experimental procedure

2.1 Raw materials

The raw material used for 3D printing was clayey material from the "Sè" region in the Benin Republic and clay powder from Keramische Massen (The Netherlands). The main composition of the natural clayey material collected in "Sè" included quartz and kaolinite. The details of the raw material characteristics are presented by [16]. To form the ceramic paste, water was added, and rice husk was also used as a pore-forming material. Twenty percent (20%) of rice husk (w/w ratio) was used in the ceramic paste. Rice husks were of variable size (<45 µm, 45–200 µm and 200–500 µm). The optimal water amount to be used was determined by trial and error after several experiments. Table 1 shows the details of the different formulations.

Table 1 Formulation of ceramic pastes

Filter	Rice husk size (µm)	Amount of clay (g)	Amount of rice husk (g)	Amount of water (g)
F1	<45	240	60	135
F2	$45 < D < 200$	240	60	168
F3	$200 < D < 500$	240	60	195
FC1	$D < 45$	240	60	125
FC2	$45 < D < 200$	240	60	155

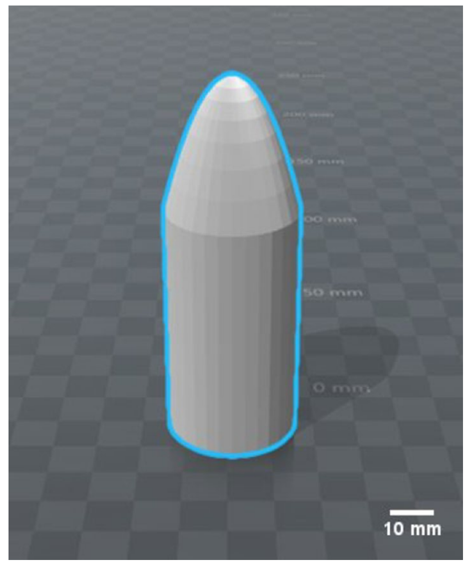

Fig. 1 Drawing of the filter design

2.1.1 3D printing device and procedure

Prior to printing, the software 123D design was used to draw the filters. A drawing of the filter design is presented in Fig. 1. The filter has a diameter of 30 mm and a thickness of 3 mm. The height of the cylindrical part is 50 mm, and the conical part has a height of 35 mm.

The 3D printer used was an "Ultimaker 1", designed by the Dutch company "Ultimaker" and intended primarily for plastic material additive manufacturing. Modifications related to the development of a clay material printing system were made on the printer. A syringe was adapted to the printer head. This syringe contained the clay paste for the manufacture of the ceramic piece (Fig. 2). To attach the syringe, a plastic material support was made using the 3D printer. A closure was also made for the syringe to allow the ceramic paste to be pushed out progressively under 4 bars of pressurized air. The tip of the syringe was used for extrusion (1 mm) and the thickness of the filters (3 mm). The manufacturing device in its final version is shown in Fig. 3.

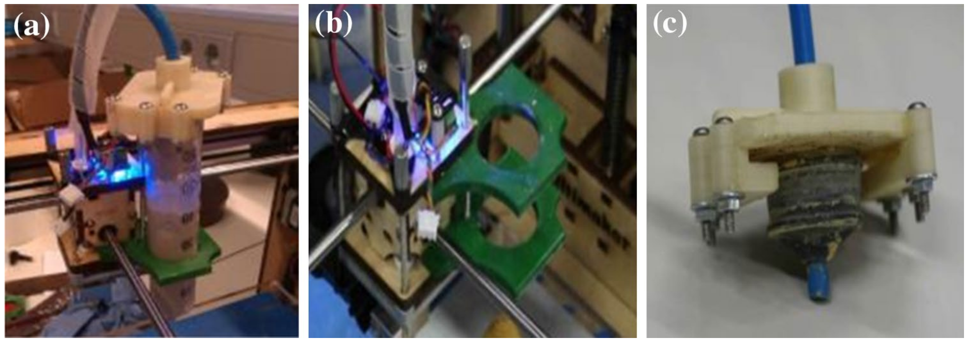

Fig. 2 Changes made to the Ultimaker 1. **a** A syringe containing the ceramic paste; **b** syringe attachment bracket; **c** hood for closing the syringe and for applying air under pressure

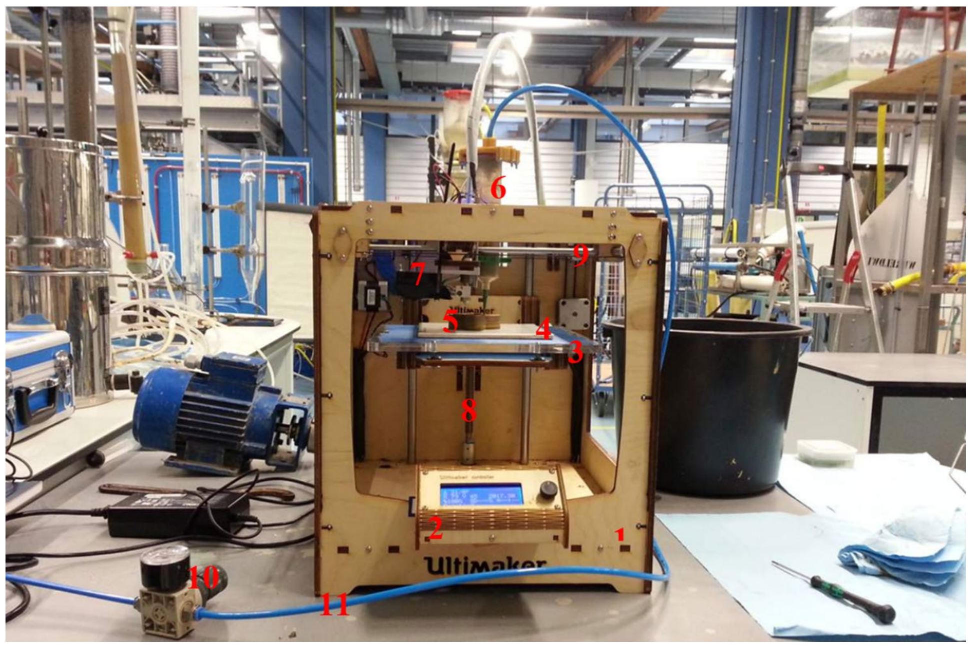

Fig. 3 Overview of the additive manufacturing device dedicated to the realization of the filters. 1. 3D printer "Ultimaker 1". 2. Device for controlling the printing speed. 3. Printing platform. 4. Ceramic support. 5. In-process materials. 6. Syringe containing ceramic paste. 7. Fan for drying. 8. Actuator. 9. Travel bar allowing movement of the print head. 10. Manometer for pressure control. 11. Pressurized air duct connection

The printing was performed layer by layer, and the printed pattern of each layer corresponded to the cross section of the designed figure. The printing parameters of the layer thickness and saturation level were set at 1.2 mm and 100%, respectively. The printing speed was 7 mm/s.

2.2 Thermal treatment program

After processing the 3D printing, the printed specimens were subjected to 1 week of drying at room temperature and then sintered in a high-temperature furnace (Keramikos, the Netherlands) at 900 °C for 2 h with a heating rate of 1.6 °C/min, followed by slow cooling to avoid cracks. The obtained filters were immersed in water to ensure the absence of bubbles. The porosity was measured by noting the weight difference between the dry and saturated filters. SEM images were taken using a scanning electronic microscope (SEM FEG Supra 40 VP Zeiss). Filters were kept dry in an oven at 120 °C until use.

2.3 Application to water treatment

The filter performance was assessed using surface water treatment. The experimental setup used for this purpose was composed of PVC columns with 1-m height and 50-mm diameter (Fig. 4). The PVC columns were interconnected and fed by a pump (Masterflex, Model 77200-60, USA) to maintain the same water level (and, therefore, constant pressure) on the different filters.

The ceramic filters were attached to the bottom of the PVC columns using acrylic glue. Figure 5 shows an example of an attached ceramic filter.

Water samples were taken after filtration and analyzed for their physicochemical and microbiological characteristics. For the physicochemical analysis, pH and conductivity were measured with a multiparameter analyzer model HI 98311 from Hanna Instruments according to the manufacturer's instructions. Turbidity was measured using a Turbiquant 110 Ir Merck. Absorbance at 254 nm was measured with a Shimadzu UV Mini 1240 spectrophotometer. The permanent

Fig. 4 Experimental setup for water filtration tests

Fig. 5 Ceramic filter attached to the base of a filtration column

index was measured according to the standard method NFT 90-050.

Microbiological parameters, i.e., *Escherichia coli*, total coliform (TC) and fecal coliform (FC), were measured using Rapid E-Coli media according to the standard method NFV-08-05.

3 Results and discussion

3.1 3D-printed specimens

The 3D-printed clay ceramic filters obtained are shown in Fig. 6.

A comparison of the various filters for visual inspection indicates that it was easier to make filters with smaller rice husk sizes, and these filters had a more suitable appearance. We were not able to make filters from commercial clay using rice husk with a size between 200 and 500 µm. Densification of the structure can also be seen in Fig. 6.

After drying and sintering, shrinkage that varied according to the formulation was observed in the ceramic pastes (Fig. 7).

Table 2 lists the details of the filter size before and after sintering.

After sintering, filters F1, F2 and F3 had the same dimensions, and the FC1 and FC2 filters had the same dimensions. The observed shrinkage was probably related to the characteristics of the raw materials and was more noticeable with natural clay materials. For the rice husk size used, it seemed that porous materials did not influence the shrinkage of the filters.

3.2 Flow rate

Table 3 shows the variation in the flow rate obtained using the filters for distilled water filtration. This result reveals that a better flow rate and porosity were obtained for a filter with a larger rice husk size. We also noticed that the commercial clay materials had better porosities and flow rates than the natural clay materials. This difference can be explained by the fact that the characteristics of the raw material influence the pore volume, and therefore have an influence on the efficacy of the filters.

3.3 Water treatment efficacity

To assess the filters' ability to treat water, a filtration test was performed using surface water. Table 4 summarizes the physicochemical and microbiological results obtained. A filtration test was used to evaluate the performances of the FC1 and FC2 filters. With regard to filters F1, F2 and F3, a fouling phenomenon was quickly observed. The fouling may be due to both inorganic and organic compounds. Indeed, in the literature, the main fouling phenomena are adsorption, polarization concentration and filter cake formation [17, 18]. Each phenomenon contributes to an increase in the flow resistance of the permeate flow. In this case, with regard to turbidity (9.07 NTU), the pore size and filter cake formation are the most likely causes of the observed fouling. When the pore volume decreases, the pores become more narrow, resulting in an increase in the hydraulic resistance [19, 20]. The mechanism leading to particle capture is often described by an analogy with depth filtration in particle beds, i.e., by a collision (or capture) efficiency that corresponds to the ratio of the rate of particles intercepted by the support by the rate of particles brought by the flow towards the support. On a physical level, it is related to the phenomena of particle

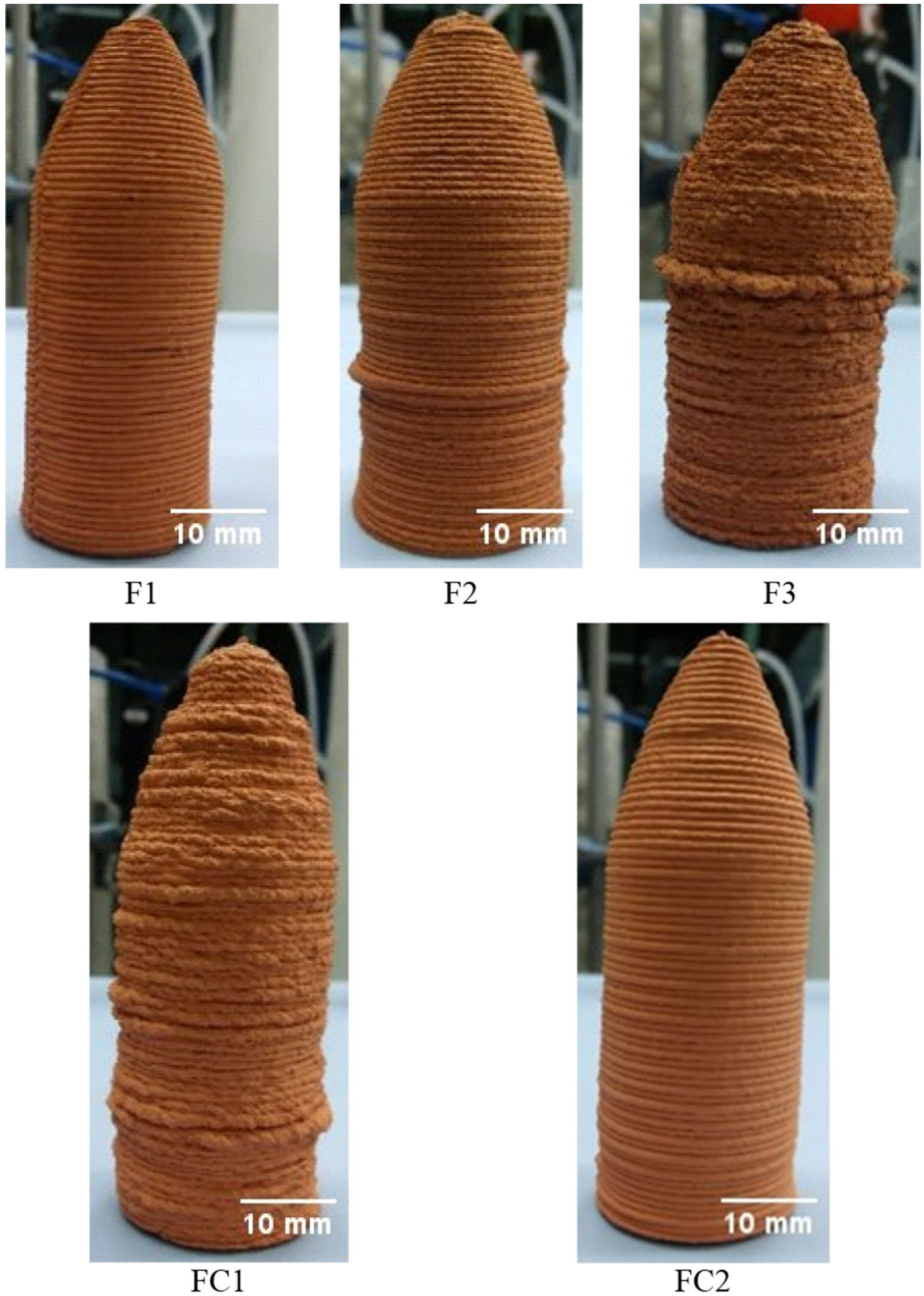

Fig. 6 Photo of 3D-printed ceramic filters. F1: rice husk size $<$45 µm. F2: rice husk size between 45 and 200 µm. F3: rice husk size between 200 and 500 µm. FC1: commercial clay with rice husk size $<$45 µm. FC2: commercial clay with rice husk size between 45 and 200 µm

transport on the surface of the pore volume (by diffusion or hydrodynamic interception mechanisms) and to the affinities of the particle for the surface (electrostatic force). The fouling by deposition on the surface is due to the flow of materials brought to the wall by convection, which causes an accumulation on the surface of the porous material. Moreover, various phenomena that were not studied, such as the hydrodynamic conditions (temperature, flow rate, pressure, etc.), may affect the fouling observed.

The turbidity, absorbance at 254 nm and permanganate index were measured for samples from F1 and F2, and the results are presented in Table 5.

The results show a very good elimination of turbidity ($>$99%), which confirms that the filters are very efficient in terms of their primary function, i.e., steric exclusion (removal of particles larger than the pore size). Good removal of organic matter was also observed (F1: 67%, F2: 68%, FC1: 65%, FC2: 67%). This removal of

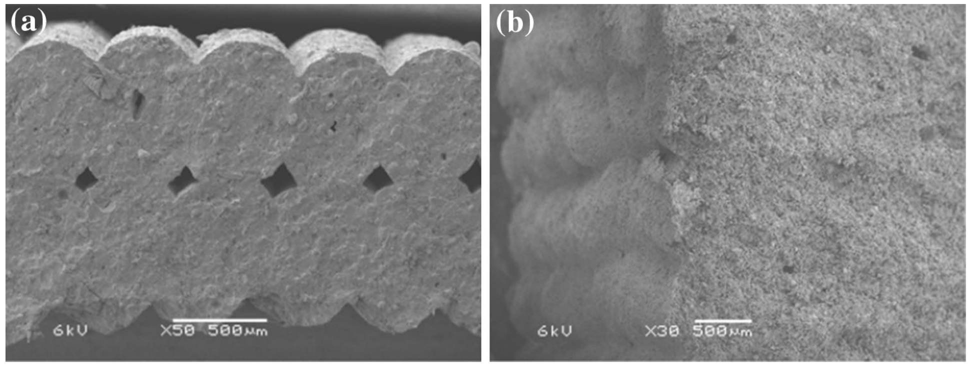

Fig. 7 SEM images of 3D-printed ceramic filters (FC1). **a** wall; **b** cross section of the filter

Table 2 Filter size before and after sintering

	Initial size (mm)	Final size (mm)	
		F1, F2 and F3	FC1 and FC2
Thickness	3	2.5	2.8
Diameter	30	20	20
Cylindrical part height	50	40	47
Conical part height	35	20	30

Table 3 Porosity and flow rate of the filters

Filter	Water porosity (%)	Flow rate (mL/h)
F1	23.5	35
F2	26.7	55
F3	29	75
FC1	43.5	208
FC2	45.8	214

Table 4 Analytical results for raw and treated water using 3D-printed ceramic filters (1)

	Raw water	Treated water	
		FC1	FC2
pH	6.63	7.93	8.13
Temperature (°C)	24.4	24.4	24.4
Conductivity (µS/cm)	82	151	168
Turbidity (NTU)	9.07	0.065	0.062
Abs 254 nm	0.083	0.065	0.062
Permanganate index (mg/l)	8.64	3.04	2.88
Escherichia coli	02	0	0
Fecal coliform	156	0	0
Total coliform	203	0	0

organic matter is likely associated with the elimination of the particulate fraction of the organic matter because we observed a good reduction in the turbidity and the

Table 5 Analytical results for raw and treated water using 3D-printed ceramic filters (2)

	Raw water	Treated water	
		F1	F2
Abs at 254 nm	0.083	0.020	0.016
Permanganate index (mg/l)	8.64	2.88	2.80
Turbidity (NTU)	9.07	0.064	0.069

adsorption phenomena of the dissolved organic matter. The increase in the conductivity values was probably due to the mineralization of the sample by ions present in the clayey material. This phenomenon was already observed in the literature during the first hours of the operation of the ceramic filters [9].

Microbiological analyses indicated the presence of bacterial pollution in the raw water but a good microbiological quality for the treated water. It has been well established in the literature that common ceramic filters are capable of reducing key microbes in water [21–23]. These filters were, therefore, effective for the retention of microbial contaminants.

4 Conclusion

Clay ceramics with designed shapes and sizes for water filtration were obtained using 3D-printing technology. This study reveals that it is possible to make ceramic filters using natural clayey materials. The filters mainly eliminate microbiological pollution with good performance. It appears that to improve the treatment, pretreatment of the raw water needs to be performed. We conclude that 3D printing is an effective technology for ceramic filter manufacturing for water treatment at the household level. These 3D-printed ceramic filters are an innovative water quality treatment at the point of use that needs to be further studied.

Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there are no conflicts of interest.

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