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Performance of Carbon Aerogel/Fiber Paper as Capacitive Deionization Electrodes under Variable Operating Conditions

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ABSTRACT

The capacitive deionization has garnered renewed interest with recent modifications resulting in efficient (energy and cost) alternatives to desalinate brackish water (salt concentration less than 10 g/L). Recent research efforts, to improve the technology, have been focused mainly on electrode material development and modifications of operational parameters for optimized performance. This research is focused on performance evaluation of carbon aerogel/fiber paper as capacitive deionization electrodes under variable operating conditions. The main operational parameters that were varied are applied voltage, influent temperature and initial concentration. The highest adsorption capacity achieved in this on-going study is 4.61 mg/g at 33.5°C with NaCl solution of 300 mg/L initial concentration, at a rate of 12 mL/min flow rate and 1 V of applied DC voltage. The results obtained by varying operating conditions in this study will provide greater understanding on suitability of these electrodes in capacitive deionization based full-scale capacitive desalination plants.

INTRODUCTION

In today's world with increasing population and consequently increasing water demand, we see shortage of usable water. Limited fresh water resources, increased water pollution, and depleting groundwater levels have reduced the availability of usable water. The need for alternative water sources like desalination with newer and efficient techniques is higher than before. Sustainable water management and increased use of greywater would help in reducing the stress on limited water sources (Ahmed et al. 2018). The focus of this research is on desalination with capacitive deionization (CDI), a novel desalination technology, showing potential to be an energy efficient and environmentally friendly alternative (Porada et al. 2013) to traditional technologies like reverse osmosis (Zhao et al. 2013). CDI utilizes electrostatic forces to remove ions by application of voltage on two electrodes arranged in supercapacitor like setting (Suss et al. 2015). Carbon based electrodes have been popular choice among researchers to be used as CDI electrodes due to their properties like good electrical conductivity, relative inertness to chemicals in the salt environment at the low voltage level applied in CDI, porous structure, high surface area (Huang

et al. 2017) etc. Much of the CDI literature have been focused on developing electrode materials through some chemical treatments, metal oxide depositions, composite electrode development and other suitable methods (Thamilselvan et al. 2016). Very few works have focused on exploring effects of operating parameters and mathematical & statistical modelling of data (Zhao et al. 2014).

The need for assessing effects of parameters on adsorption capacity of various electrodes is indispensable for probable future full-scale operations of CDI. Keeping this need in mind, this work focused on exploring possible variables that might affect the adsorption capacity of the electrodes in CDI applications. The experimental data were collected and analyzed to study the effects of applied voltage, influent temperature and initial concentration are reported based on experimental data.

MATERIALS AND METHODS

Materials and cell preparation procedures used in this study is similar to one previously published work by this research group (Ahmed and Tewari 2017). The electrode material used in this study is Carbon/Aerogel-Fiber paper. Sheets of 254 mm x 88.9 mm with thickness of 0.1778 mm were purchased from Marketech International (Port Townsend, WA). These sheets were cut into pieces of 50.8 mm x 88.9 mm to be used as electrodes. A flexible graphite sheet with no wire mesh insert was purchased from Equalseal through Amazon to be used as current collector. These sheets were of 0.7 mm thickness. These sheets were cut into pieces of 120 mm x 10 mm approximately. Acrylic sheets of same thickness (0.7 mm) were cut into pieces of approximately same dimensions as graphite sheets. These graphite and acrylic sheets were pasted on each other using nonconductive Silicon II clear sealant/glue purchased from Lowe's, Ruston, LA. The material was used to prepare CDI cells shown in Figure-1a and 1b and as described below.

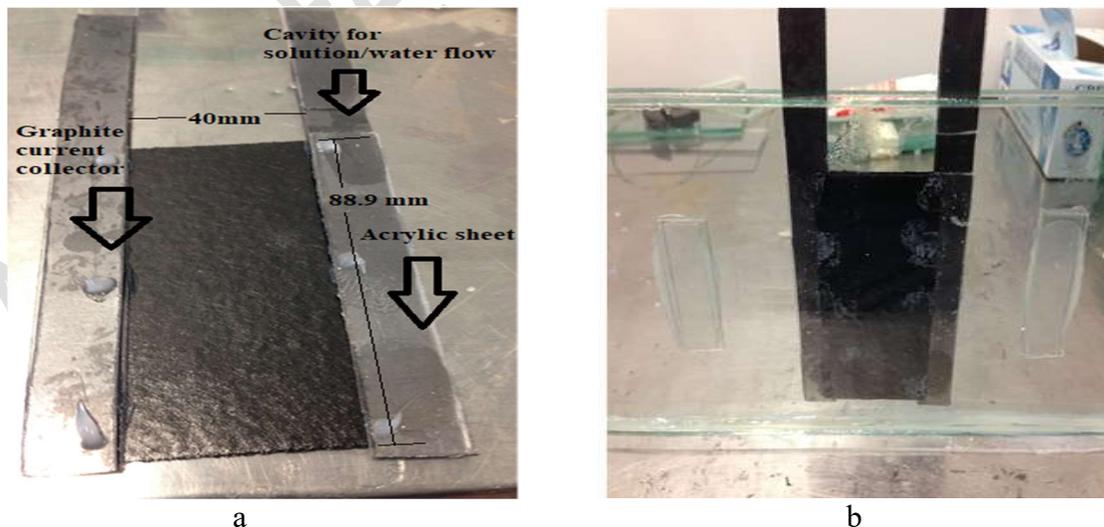


Figure-1. a) Electrode pasted on graphite sheet and acrylic sheet b) Electrode with current collectors inserted into glass cell.

One of the electrode pieces was pasted on the graphite-acrylic sheet assembly in a way that on each two longitudinal edges of the electrode touches graphite on one side and acrylic sheet on the

other side. The other piece of the electrode was pasted on top of the surfaces shown in Figure-1a so that it also touches graphite on one side and acrylic sheet on the other side. The face to face area of the electrode was 40 mm x 88.9 mm. The separation between the electrodes (acrylic sheet+ graphite+ glue thickness) was approximately (1.5-1.6 mm). This assembly was then inserted into a glass cell prepared to be used as CDI cell unit as shown in Figure-1b. Then this whole cell was sealed with Silicone II clear sealant purchased from Lowe's, Ruston, LA. Figure-2 provides a view of arrangement of the experiment set up with a CDI cell in operation.

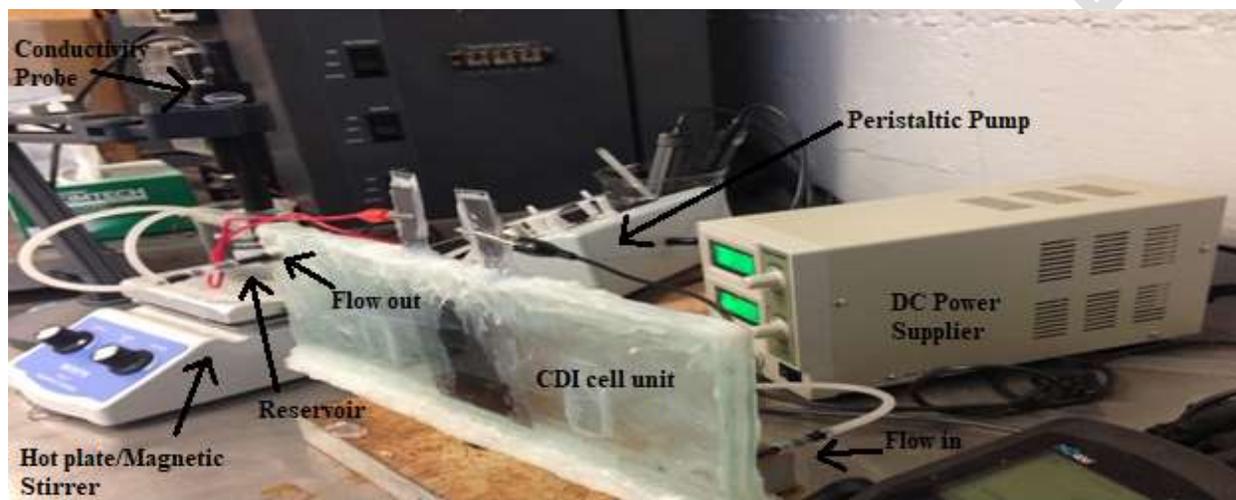


Figure-2. Experimental set up.

In the experiment set up, a reservoir is kept on top of a stirring hot plate. A solution of NaCl is pumped through the CDI cell and back to same reservoir using a peristaltic pump (Model:3387, Fisher Scientific). This pump was calibrated for each dial position to maintain required flow into the CDI cell. A power supplier (TP1803D, TEK-POWER) was connected to the current collectors to supply desired voltage. A CDC401 conductivity probe and HQ40d multimeter by HACH were used to monitor conductivity in the solution and record data.

Applied voltage was varied between 0.6 to 1.2 V in experiments at an increment of 0.2 V. It was not increased more than 1.2 V to ensure the stability of the electrolyte (NaCl solution). As chloride has activation potential of 1.36 V and if a voltage greater than 1.36 V is applied, it may induce non-faradaic reaction (Porada, Zhao et al. 2013). This reaction between chlorine and carbon electrodes can introduce a brown colored substance to the solution as found in a previous study (Ahmed and Tewari 2016).

The all other applied parameters were kept as constant for variable voltage experiments. In this study, temperature of flowing electrolyte was maintained at room temperature (22-24 C), flow rate as 12 mL/min and initial concentration as 300 mg/L. Same electrolyte and cell was used for all the experiments to avoid variations like ion sizes, separation (spacer length) between electrodes, CDI cell size etc. The cell after each experiment underwent rigorous wash cycles to remove any remaining salts before next experiment was run.

The experiments that were conducted to study the effect of temperature involved ice bath and hot plate for temperatures below and above room temperature. The experiments were conducted in about 10⁰C difference among the temperatures of the water in the reservoir. The average temperature that could be maintained in low temperature experiments under ice bath was 9.5±1.02⁰C. The room temperature experiments showed an average of 23.5±0.23⁰C. Then an elevated temperature was used in the hot plate and adjusted from time to time to maintain a near constant temperature on the reservoir. Applying around 55-60⁰C of temperature in the hot plate maintained an average of 33.5±1.25⁰C on the reservoir. The applied voltage, initial concentration and flow rate were kept constant at 1 V, 300 mg/L and 12 mL/min respectively.

The initial concentrations, used to study the effect of, were 100, 200, 300 and 400 mg/L. The applied voltage, temperature and flow rate were kept constant at 1 V, room temperature (22-24⁰C maintained on all trials) and 12 mL/min. Mass balance calculations were performed to calculate salt removal capacity from the conductivity-drop experiments and the following equation was used:

$$\text{Salt removal capacity (mg/g)} = \frac{\text{Slope of calibration curve of salinity vs conductivity of NaCl} * \text{conductivity drop} * \text{Volume of solution pumped}}{\text{electrode mass}} \quad \text{-----(1)}$$

Here, slope of the calibration curve of salinity vs conductivity of NaCl is 0.5057 (Figure-3), total volume of solution = 0.16 L and mass of the electrode was 1.44 g.

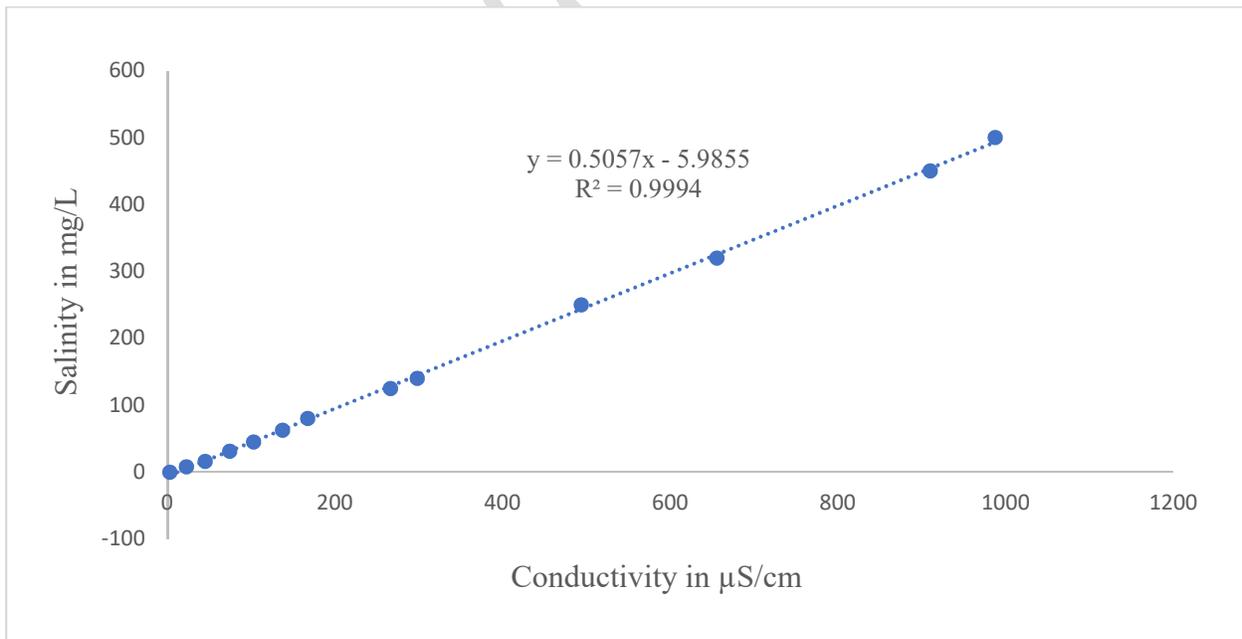


Figure-3. Salinity vs. conductivity for NaCl solution.

RESULTS

Variation in applied voltage has shown to affect the adsorption capacity of the electrodes under study. With increasing voltage, the adsorption capacity also went up (Figure-4). For applied voltage of 0.6 V, conductivity was observed to decrease to 560 $\mu\text{S}/\text{cm}$ from 600 $\mu\text{S}/\text{cm}$ (a change of 40 $\mu\text{S}/\text{cm}$). The change in conductivity level went further up with increasing voltage. In this study we observed conductivity drops of 50, 60 and 74 $\mu\text{S}/\text{cm}$ for applied voltage of 0.8 V, 1 V and 1.2 V. Corresponding salt removal capacity are shown in Table-1.

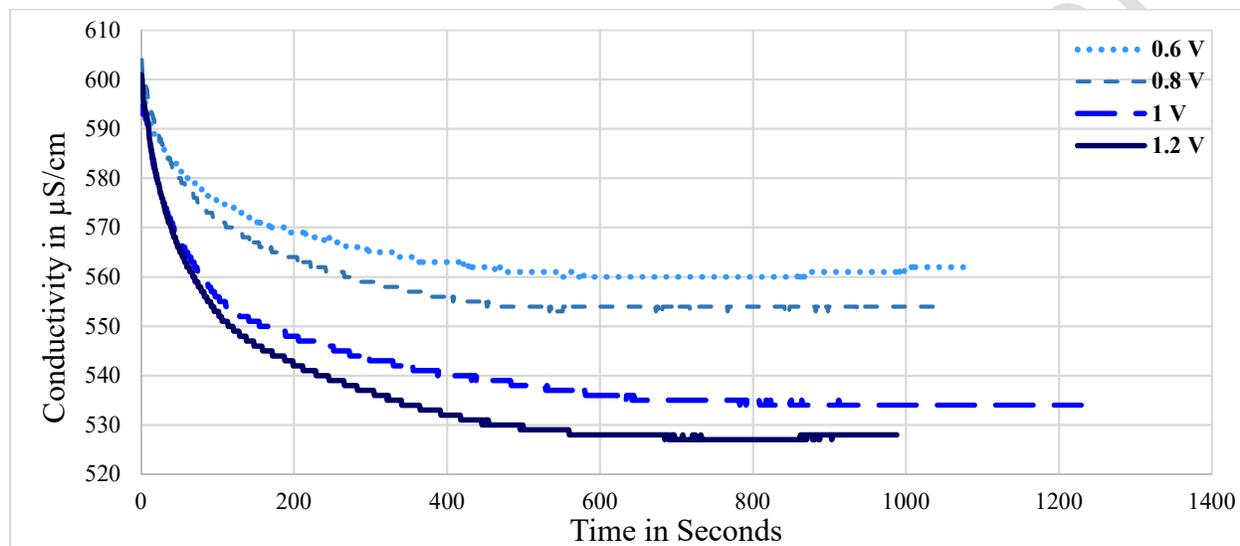


Figure-4. Effect of applied voltages.

The reason behind this increase in conductivity change and corresponding salt removal capacity can be attributed to the fact of stronger electrostatic force with increasing voltage.

Table-1. Salt removal capacity at different applied conditions.

Applied Condition	Applied Value	Salt Removed/mass of electrode (mg/g)
Applied Voltage (V)	0.6	2.25
	0.8	2.81
	1	3.37
	1.2	4.16
Influent Temperature ($^{\circ}\text{C}$)	9.5 ± 1.02	3.15
	23.5 ± 0.23	3.43
	33.5 ± 1.25	4.61
Initial Concentration (mg/L)	100	2.21
	200	2.75
	300	3.26
	400	3.93

With increasing influent temperature, it is expected that conductivity change and corresponding salt removal capacity will increase as ionic activity increases. However, in our observations

experiments done at $9.5 \pm 1.02^{\circ}\text{C}$ and $23.5 \pm 0.23^{\circ}\text{C}$ did not show much difference in their results. But experiments done at $33.5 \pm 1.25^{\circ}\text{C}$ showed significantly salt higher removal capacity (Figure-5). Change of conductivity level were 56, 61 and 82 $\mu\text{S}/\text{cm}$ at influent temperatures of 9.5 ± 1.02 , 23.5 ± 0.23 and $33.5 \pm 1.25^{\circ}\text{C}$. Corresponding salt removal capacities are also reported in Table-1. The change in conductivity and corresponding salt removal capacity showed an increasing trend with increasing applied initial concentration. Conductivity changes of 39.4, 49, 58 and 70 $\mu\text{S}/\text{cm}$ were observed corresponding to 100, 200, 300 and 400 mg/L of initial concentration respectively (Figure-6). Salt removal capacities are shown in Table-1.

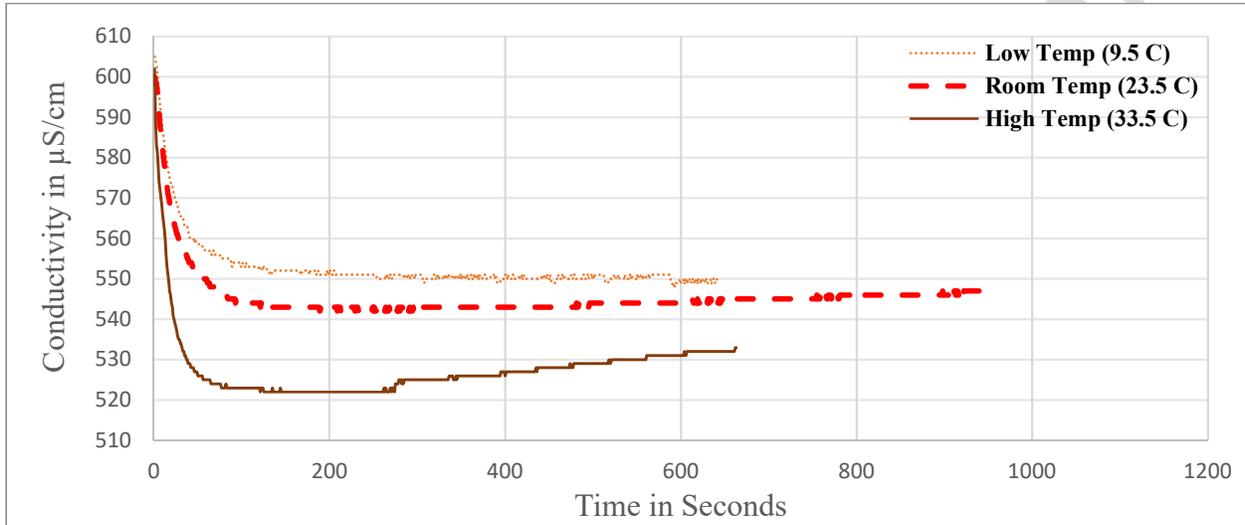


Figure-5. Effect of influent temperature.

This increase of conductivity change or ion removal capacity can be attributed to the fact of higher concentration gradient near electrode surface.

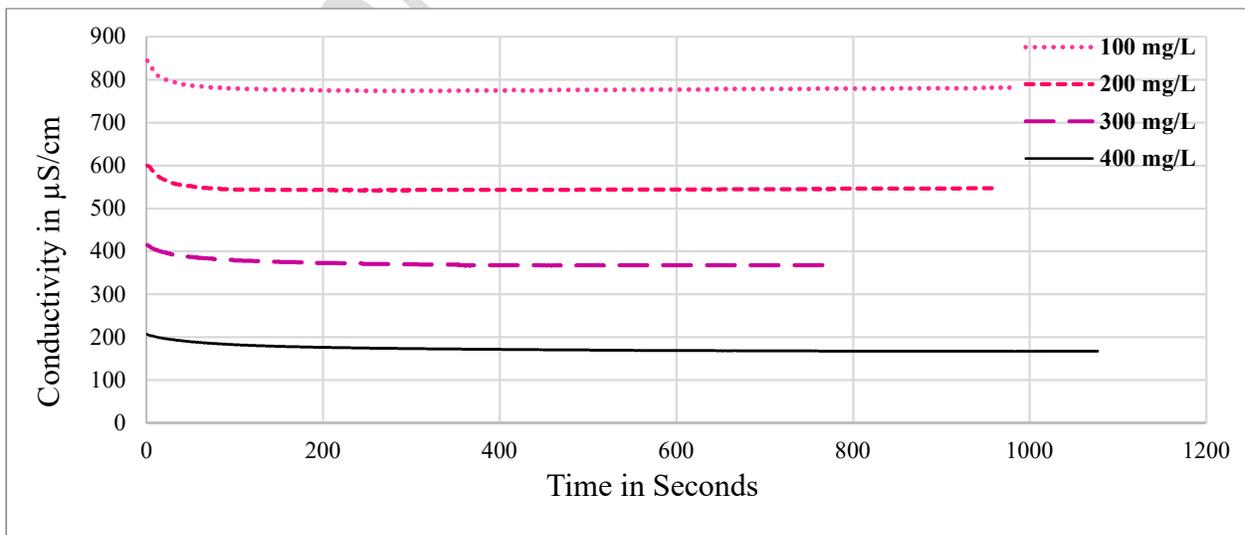


Figure-6. Effect of initial concentration.

The highest salt removal capacity per mass of electrode was observed to be 4.61 mg/g in this study. This value is compared with other published values for different electrode materials (unmodified forms) as presented in Table-2. It seems that the highest salt removal per unit mass of electrode obtained in this study is reasonable compared to the values reported in Table-2. The recent developments of composite electrode materials have yielded many high capacity electrodes. Examples include 3D graphene and hierarchically porous carbon composite (6.18 mg/g) (Wen et al. 2013), phenolic resin based carbon nanofiber electrodes (20.1 mg/g) (Chen et al. 2014), N, P doped meso/microporous carbon (20.78 mg/g) (Xu et al. 2017) etc.

Table-2. Salt removal capacity by different electrodes.

Ref.	Material	Removal Capacity (mg/g)
This work	Carbon aerogel/fiber paper	4.61
Li et al. 2010	Activated carbon	0.8
Myint et al. 2014	Activated carbon cloth	5.8
Gao et al. 2013	Carbon xerogel	1.9
Tsouris et al. 2011	Carbon aerogel	5.8
Yang et al. 2014	Carbon nanotube (CNT)	1.36
Wen et al. 2012	Ordered mesoporous carbon	1.94
Zhang et al. 2012	Graphene	1.1
Wen et al. 2013	3D graphene	4.41

DISCUSSION

Adsorption capacities of the carbon aerogel/fiber electrode papers increased with increasing levels of applied voltage and initial concentration. At lower or room temperature level adsorption capacities were almost similar, but increased significantly at higher temperature. The maximum adsorption capacity was found to be 4.61 mg/g at temperature of 33.5⁰C, applied voltage of 1 V, flow rate of 12 mL/min and initial concentration of 300 mg/L. This capacity may further be maximized by increasing the voltage over 1 V and if influent concentration is over 300 mg/L for real life application. The maximum capacity of the carbon aerogel/fiber paper electrodes observed in this study was high compared to other unmodified electrodes, but significantly lower than composite or chemically modified electrodes. Further studies are required to explore the possibility of composite electrodes made from carbon aerogel/fiber paper exhibiting better adsorption capacity than other composite electrodes.

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