Conductive geopolymers as low-cost electrode material for microbial fuel cells

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Supporting Information 1: Metakaolin and waterglass raw materials

Used **metakaolin** is a commercial product of industrial-scale calcination (and subsequent grinding) of a quartz-rich clay which originated from a primary geological deposit. The clay was calcined in an industrial rotary kiln for about 4 h at 700–750°C with a production capacity of about 11 tons/hour. Quantitative powder X-ray diffraction by means of DIFFRAC.TOPAS (Version 5, Bruker) software for Rietveld refinement and using 10 wt.% spiked corundum resulted in 46 wt.% amorphous, 40 wt.% quartz and 10 % Muscovite in the metakaolin (full mineralogical composition given in Figure S1). Metakaolin had a Blaine specific surface area of 10 000 cm²/g and a median grain size of 41 µm.

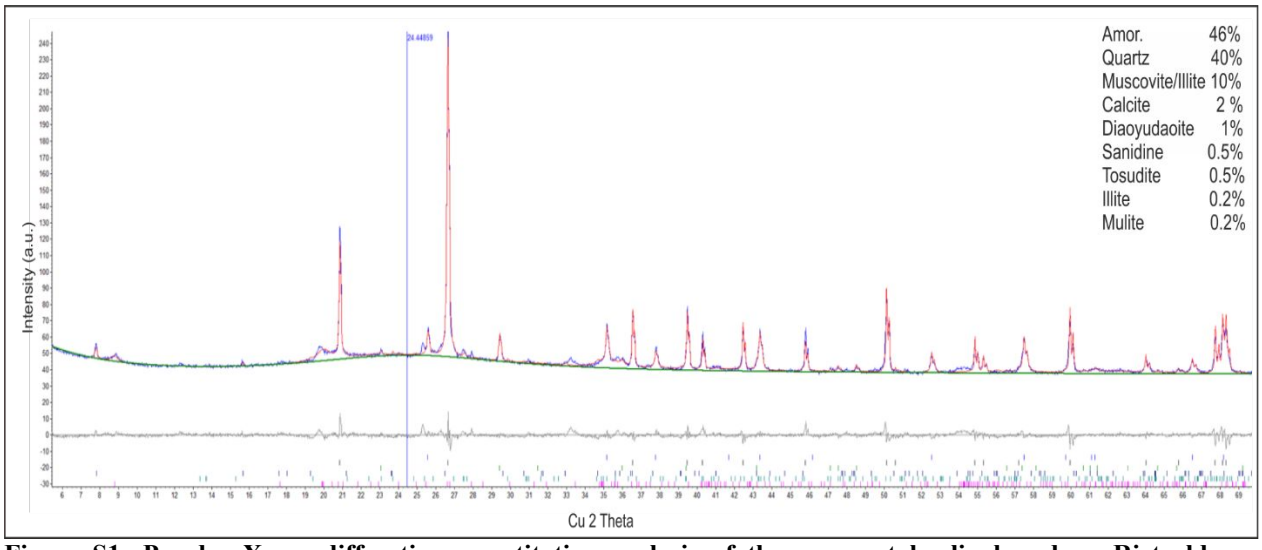

Figure S1: Powder X-ray diffraction quantitative analysis of the raw metakaolin based on Rietveld refinement (using DIFFRAC.TOPAS Version 5, Bruker).

Potassium silicate solution (**waterglass**) was used as alkaline activator, with a molar SiO_2/K_2O ratio of 1.5, 45% total SiO_2 and K_2O "solid" content, 20 mPas viscosity and 1.51 g/cm³ density.

Chemical composition of metakaolins and K-based silicate solution (waterglass) is given in **Table 1.**

Table S1. Chemical composition of metakaolins and K-based silicate solution (waterglass).

Material	SiO_2	Al_2O_3	CaO	TiO_2	Fe_2O_3	MgO	Na_2O	K_2O	H_2O
Metakaolin MK1	67.0	27.0	1.0	1	4	0.1	0.1	0.2	–
K-waterglass	22	–	–	–	–	–	–	23	55

<u>Supporting Information 2</u>: Graphite powder raw material

The graphite used is a high purity, specially ground natural graphite. The technical data of graphite is given in **Table S2**.

Table S2. The technical data of natural graphite.

Material	Carbon wt. %	D_{10} µm	D_{50} µm	D_{90} µm	Spec. surface m^2/g	Bulk density g/l	Electrical conductivity S/m
Graphite	>99	1-3	4-6	8-11	10	250	183

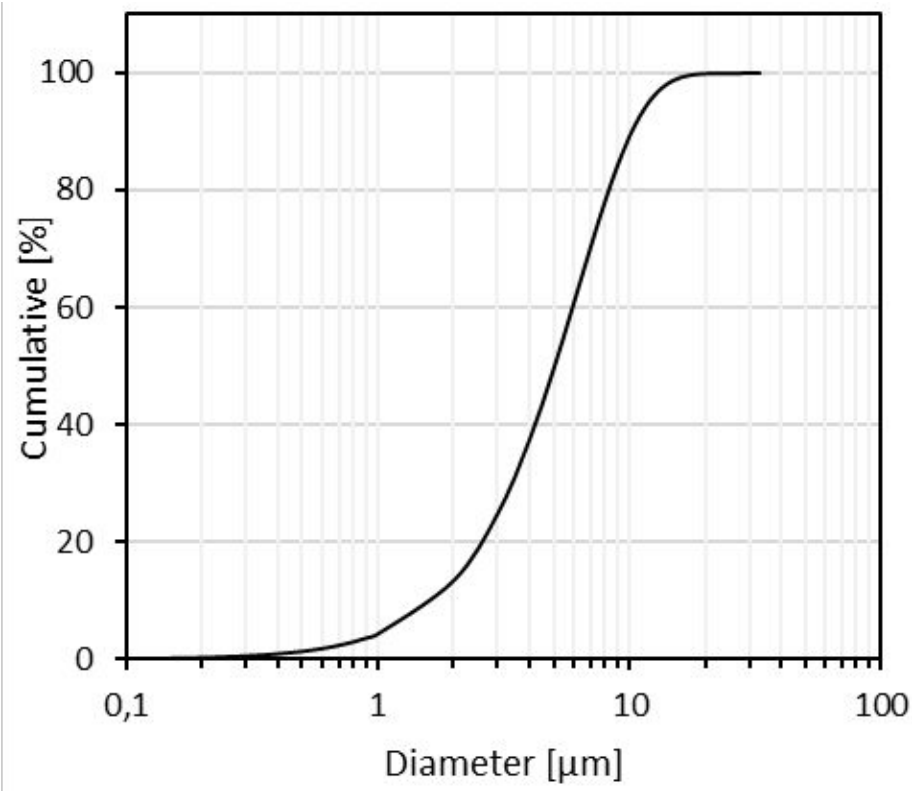

Figure S2. Particle size distribution of graphite powder.

Graphite powder was analyzed by scanning electron microscopy (Philips XL-30 FEG , Netherlands) using an electron beam at 30 kV as the accelerating voltage. Figure S3 shows SEM images of the natural graphite. The flake graphite particles have thin plate shapes.

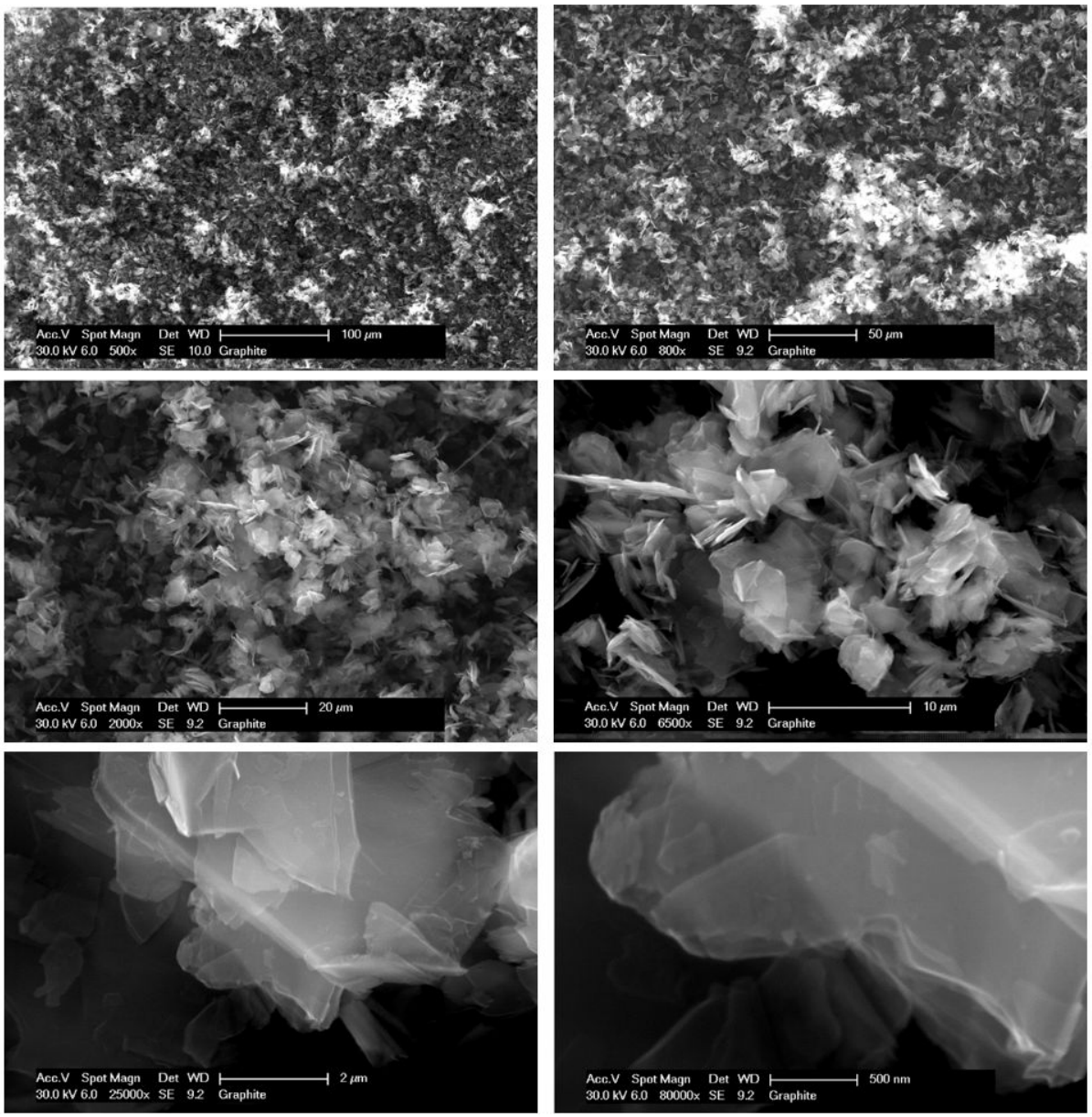

Figure S3. Scanning electron microscope images of the graphite powders.

Supporting Information 3: Mixture design of PCG and GPG is given in Table S3.

Table S3. Mixture design of PCG and GPG.

PCG	Graphite [g]	Cement [g]	Water [g]	PCE [g]	Fly ash [g]
PC Ref.	0	948	570	0	0
PC06 1C	28	948	570	0	0
PC06 3C	57	948	570	0	0
PC06 4C	85	948	570	0	0
PC06 9C	199	948	570	19.9	0
PC06 3F 8C	176	733	572	17.6	220
PC06 3F 9C	198	723	564	19.8	217
PC06 3F 10C	220	714	557	22	214
PC75 8C	176	843	632	17.6	0
PC75 9C	198	832	624	19.8	0
PC75 10C	220	820	615	22	0
GPG	**Graphite [g]**	**Metakaolin[g]**	**Waterglass [g]**	**PCE [g]**	**Water [g]**
GP Ref.	0	1173.2	938.5	0	0
GP08 1W 1C	22	1133.3	906.6	2.2	22
GP08 1W 2C	44	1093.4	874.7	4.4	44
GP08 1W 3C	66	1053.5	842.8	6.6	66
GP08 1W 5C	110	973.7	779	11	110
GP08 1W 7C	154	894	715.2	15.4	154
GP08 1W 8C	176	854.1	683.3	17.6	176
GP08 1W 9C	198	814.2	651.3	19.8	198
GP08 1W 10C	220	774.	619.4	22	220
GP08 1.2W 10C	220	722	578	22	264
GP08 1.7W 10C	220	594	475	22	374
GP08 2W 10C	220	516.2	413	22	440

Supporting Information 4: Laboratory Electrochemical H-Cell

The H-shaped **reactor** consists of two modified 100 mL laboratory bottles, which are connected to each other by flanges. Between the flanges, which are fixed by means of a clamping ring, a circular proton exchange membrane on a sealing ring is located.
A second flange is attached to the working electrode chamber (WE chamber), over which the working electrode is attached from the outside by means of a clamping system. A 1 mm thick circular silicone seal between the working electrode and the flange is used. The inner diameter of the flange is 2.5 cm and thus the geometrically accessible area 4.9 cm2.
The counter electrode chamber (CE chamber) contains a graphite electrode (submerged geometric area approx. 20 cm²), which can be immersed via the bottle neck, through a screw cap with septum.
To ensure constant potential control, a reference electrode can be introduced into the WE chamber through a screw cap with fixing rings. Figure S4 shows a completely mounted H-cell prepared for sterilization.

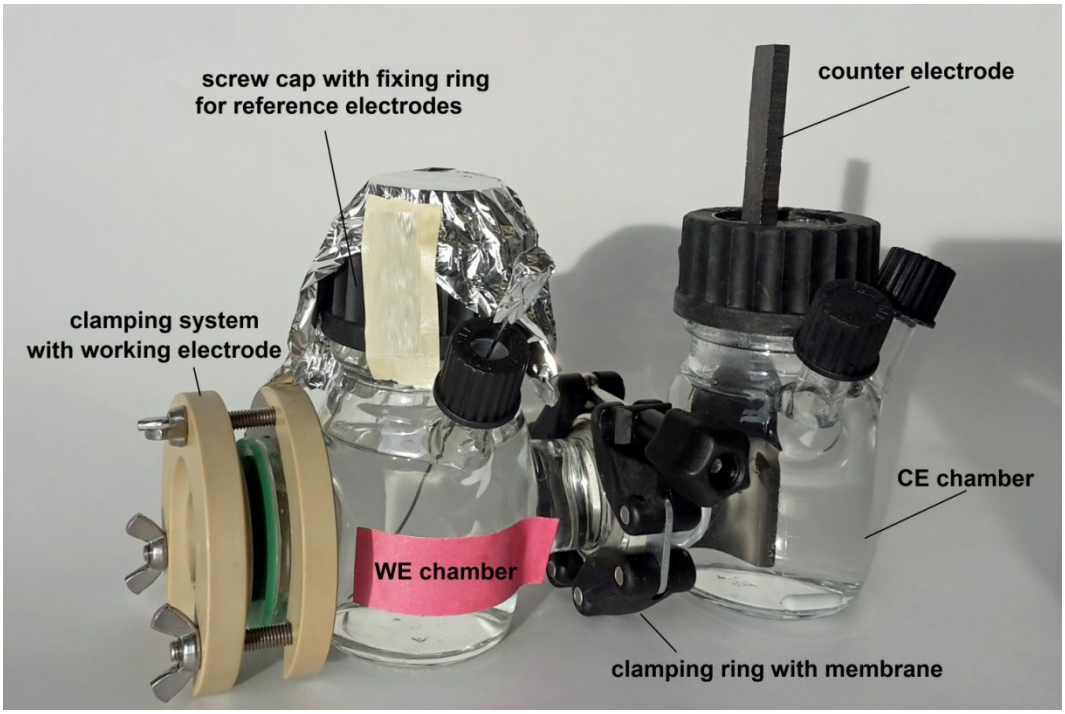

Figure S4. Mounted H-cell without reference electrode

Supporting Information 5: Description of the working steps of the MFC experiments

At day 1 the cultivation of a 7-day culture was prepared: In a sterile workbench, 1 mL of a stationary grown culture was taken from the septum bottle of the 7-day culture prepared one week ago using a syringe and cannula and transferred to a new septum bottle containing 50 mL of the growth media. The newly prepared 7-day culture was stored at 30 °C.
At day 4 the cultivation of the precultures for further deployment in the MFCs was done: In a sterile workbench, 1 mL of the culture media was taken from the septa bottle of the 7-day culture produced on day 1 and inoculated to 4 septum bottles each containing 50 mL of the growth medium. The septum bottles were incubated at 30 °C.
At day 5 the assembly and sterilization of the fuel cells was carried out: 4 MFCs were assembled with the electrode materials (WE) to be tested. The fuel cells were filled with ultrapure water and sealed with aluminium foil. Subsequently the MFCs were sterilized in an autoclave for 20 min at 120 °C. After sterilization they were temporarily stored under a clean-bench.
At Day 6 the preparation of the MFCs was completed and the tests were started: The four sterilized MFCs were emptied in a sterile workbench and filled with growth media lacking disodium fumarate. The WE chambers were filled with 120 mL each and the CE chamber with 105 mL, respectively.
The Haber-Luggin capillaries were inserted into the WE chambers after being sterilized in 70 % ethanol via the screw caps with fixing rings and sealed gas-tight with foil. Then, the Ag/AgCl/KCl_{sat}-reference electrodes (Sensortechnik Meinsberg, Waldheim, Germany) were inserted into the Haber-Luggin capillaries filled with KCl_{sat}. Finally, cannulas attached to sterile filters (pore size = 0.22 µm) were placed for gassing the WE chambers at 40 mL min^{-1} with a gas mixture of 80 % N_2 + 20 % CO_2.

<u>Supporting Information 6</u>: Current density curves of tests with PC063F10C.

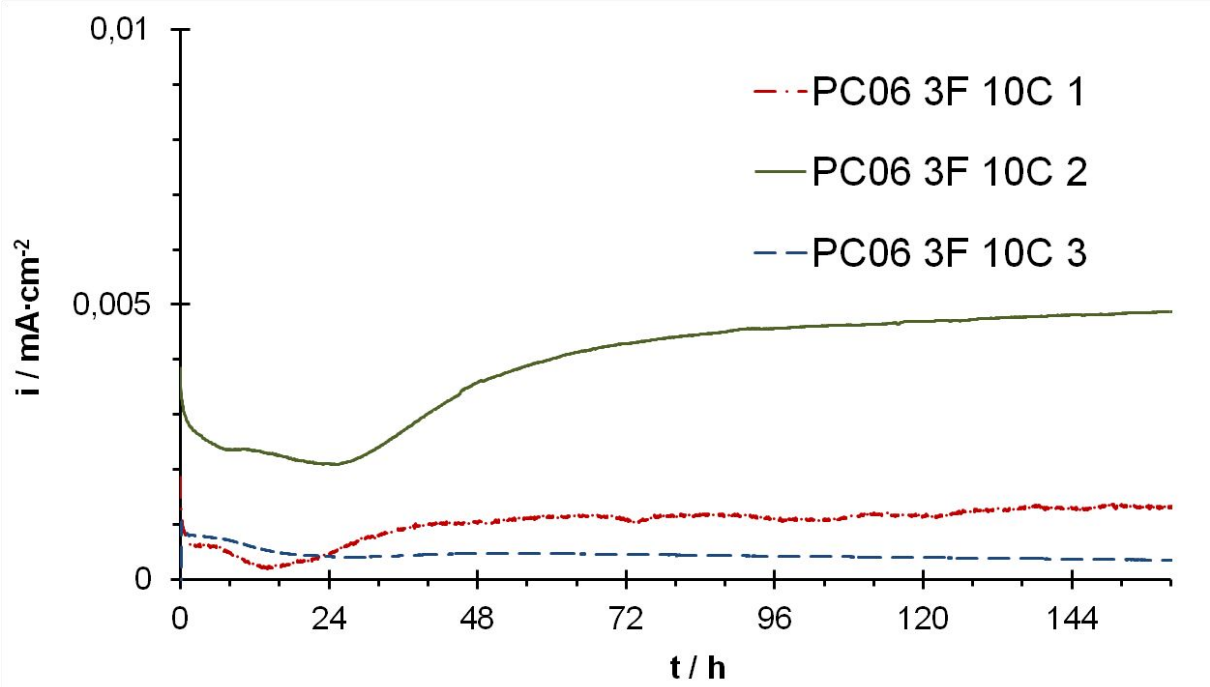

Figure S5. Current density curves for cultivations of G. sulfurreducens in H-cell MFCs at PC063F10C (anoxic Geobacter medium without fumarate + acetate; electron acceptor: anode; 30 °C; anaerobic). Current density is referred to the geometrical WE surface (4.9 cm^2).

Photo of the multi-MFCs setup.

Figure S6: MFCs connected to a multipotentiostat placed on multi-position magnetic stirrer inside an incubator.

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