

## Solid Oxide Fuel Cells' Prospects for Landfill Gas Utilization in Russia

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**Abstract**—The problem of municipal solid wastes' (MSW) utilization is one of the most actual all over the world nowadays. In Russia, landfills are still the main way for MSW utilization. However, waste decomposition, leading to landfill gas generation, causes, on the one hand, a lot of complaints from local people mainly due to hydrogen sulphide emissions. On the other hand, landfill gas, containing 40–60 vol % of methane, can be considered as calorific fuel. Solid oxide fuel cells (SOFCs), due to high operation temperatures, can be considered as an efficient device for landfill gas conversion to electricity and heat. Withal some companies and laboratories are developing SOFCs with direct methane oxidation to increase conversion efficiency on account of steam reformer expulsion. In this research, issues of landfill gas' useful utilization are considered. Experiments on different materials' application for model gas mixtures' direct conversion into energy by means of an SOFC and foreign experience in the field of landfill gas purification and estimation of its volumes in Russia are taken as initial data. The estimation results obtained allow the authors to consider SOFC application for landfill gas conversion as a significant market niche for Russian hydrogen technologies. Energy efficiency and environmental issues can also be improved by SOFC application in this niche.

**Keywords:** fuel-cell based generators, solid oxide fuel cells, laboratory scale technology, municipal solid wastes, landfill gas, technical potential

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Fuel cells, especially those with solid oxide electrolyte, have been considered for a long time as promising generating thermal and electrical energy systems, primarily due to the high efficiency of fuel chemical energy direct conversion [1]. Because of high operating temperature (800–1000°C), SOFCs are not very sensitive to carbon monoxide (CO) impurities in the fuel gas, which makes it possible to use it as a fuel [2]. At the same time, an SOFC can be characterized by high sensitivity to some other impurities (compounds of chlorine, silicon, and sulfur) [3]. In order to avoid the solid carbon deposition on the SOFC anode electrode from a fuel gas, natural gas or biogas usually undergoes a steam reforming reaction, after which the resulting synthesis gas is sent to an SOFC [4]. However, the fuel processor, which implements the steam-reforming reaction, consumes the greater part of the thermal energy generated by the fuel cells and makes the system more complex and expensive. Therefore, schemes for the direct conversion of methane inside the SOFC are being developed in different countries. In [5], microtubular SOFC operation using simulated biogas mixtures at an efficiency of 18.5% and a current

density of 524 mA/cm<sup>2</sup> was considered. The tolerance of the system to sulfur impurities and carburization of the anode increases due to Co addition to the anode catalyst. Landfill gas containing 40–60% CH<sub>4</sub> can also be a source of methane alongside with the natural gas.

In recent years, the problem of MSW accumulation at Russian landfills with self-decomposition and arbitrary release of landfill gas has grown to a huge scale. Modern building technologies for landfill reclamation are widely used. Reclamation measures usually include sealed wells and a pipeline system provided in the whole landfill body for biogas diverting to a flare (burning) or for useful utilization. The systems for biogas cleaning and supply from the landfills to consumer gas-supply systems are also widely applied abroad. Landfill gases' possible compositions and techniques for its energy-generating applications, as well as the economic aspects of the problem, are considered in [6].

Gas-piston units with a relatively high efficiency (30–35% when operating on landfill gas) are often applied for this purpose. However, due to high temperatures in their combustion chambers, the nitrogen

oxides' emission occurs during units' operation [6]. Therefore, SOFC can be considered as a relatively efficient and environmentally friendly way to obtain energy from landfill gas. At the same time, the main scientific and technical problems to be solved are related to the landfill gas purification from impurities and the direct methane oxidation scheme implementation.

The issues of SOFCs operating on biomass gasification products (including landfill gas) are considered in [7]. The main focus of the authors is on the analysis of the admissible composition of impurities, noting a small number of experimental works on real or model gas mixtures. In connection with the influence of the processing of organic raw materials on the content of impurities in the fuel gas, various approaches to biomass gasification are analyzed.

Experiments described in this study were performed on biogas obtained from municipal solid waste in SOFCs with a supporting anode (Ni-8YSZ/8YSZ/GDC+LSCF) [2]. Biogas composition was determined, and it was purified from impurities using carbon sorbents and alkali metal oxides. A mixture of similar composition was then prepared using bottled gases. It was shown that the limiting concentration of hydrogen sulfide for SOFCs is 1 ppm (by volume), and the presence of organosilicon compounds in the fuel gas is unacceptable. In order to remove carbon dioxide from SOFC emissions, exhaust gases were used as a raw material for the growth of microalgae, the nutrient medium for which contained the fermentation products of municipal solid waste. This theoretically makes it possible to mitigate the impact on the environment by reducing carbon dioxide emissions from SOFCs.

Methods for the direct use of biogas in SOFCs (without preliminary reforming) were considered in [2]. It was shown that the carbon deposition on the anode and sulfur compounds presence in fuel gas have the most detrimental effect on the SOFC lifetime. As recommendations for reducing these poisoning effects, the structured fibrous anode materials based on  $Mg_6Al_2(OH)_{16}CO_3 \cdot 4H_2O$  parallel with water vapor and carbon dioxide share in the fuel gas control are recommended to prevent anode electrode poisoning. The gas mixture parameters depend on temperature, operating voltage, dilution of the fuel gas with steam, addition of molybdenum, copper, and cerium dioxide to the composition of the anode material. It is also noted that, in most cases, enrichment of biogas with methane contributes to the reduction of carbon deposition. The recommended fuel utilization factor according to the authors of [2] is 0.25.

A review of technologies for landfill gas cleaning from impurities in order to obtain electrical and thermal energy is presented in [8]. Sulfur compounds (primarily hydrogen sulfide), organosilicon compounds, halides, etc. are subject to mandatory removal. The gas

composition average for US landfills participating in the landfill methane utilization program is a mixture of methane (50%), carbon dioxide (45%), oxygen, nitrogen, and other impurities (5%). The organosilicon compounds' content in landfill gas can reach  $67 \text{ mg/m}^3$  and  $417 \text{ mg/m}^3$  for wastewater sewage gas. The influence of hydrogen sulfide on SOFC characteristics directly depends on the cell-operating temperature. Thus, in the case of a hydrogen sulfide content of approximately 1 ppm at  $1000^\circ\text{C}$ , a reversible drop in the operating voltage by 9% occurs; at  $800^\circ\text{C}$ , the voltage drop is much larger and has the character of irreversible degradation [9].

The permissible content of impurities in the fuel gas is,  $\text{mg/m}^3$ :

H <sub>2</sub> S	1
Silicone compounds	1
Halides	3

The main methods of landfill gas purification are absorption, adsorption, membrane separation, and component precipitation by the gas stream cooling. When landfill gas is cooled to  $5\text{--}10^\circ\text{C}$ , moisture is removed from it and the content of organosilicon compounds decreases by 10–50%. The basis of purification at present is adsorption. Relatively new is the method of organosilicon compounds adsorption at elevated temperatures ( $400^\circ\text{C}$ ) on oxides of aluminum, silicon, calcium, and magnesium, although the latter two are also characterized by increased carbon dioxide absorption. It was shown in [8] that activated carbon has a high sorption capacity for organosilicon compounds, but it is the most difficult sorbent to regenerate. The highest rates were achieved on superhydrophobic polymeric sorbents. An additional feature of these expensive materials is the loss of sorption capacity by less than 10% per cycle during ten regeneration cycles (heating in air up to  $100^\circ\text{C}$ ).

The main sorbent for hydrogen sulfide are porous media based on iron oxide. In [10], the sorption capacity of porous sorbents based on iron oxide is given as equal to 0.075 g of sulfur/g of substance. A combination of condensation with several sorbents (activated carbon and iron or aluminum compounds) seems promising for practical use, since each of the sorbents usually captures its own target impurity.

Multistage purification was implemented in the DEMOSOFC project [11]. In this case, the first purification stage was used based on biogas cooling with water to  $5\text{--}10^\circ\text{C}$ , and the second stage was based on sorption on activated carbons specially selected to remove organosilicon compounds, as well as traces of hydrogen sulfide. After purification, the biogas pressure was increased to 0.4 MPa by a compressor to maintain the required pressure in the SOFC stack. The mixture reforming was not carried out, which made it possible to implement a cogeneration power

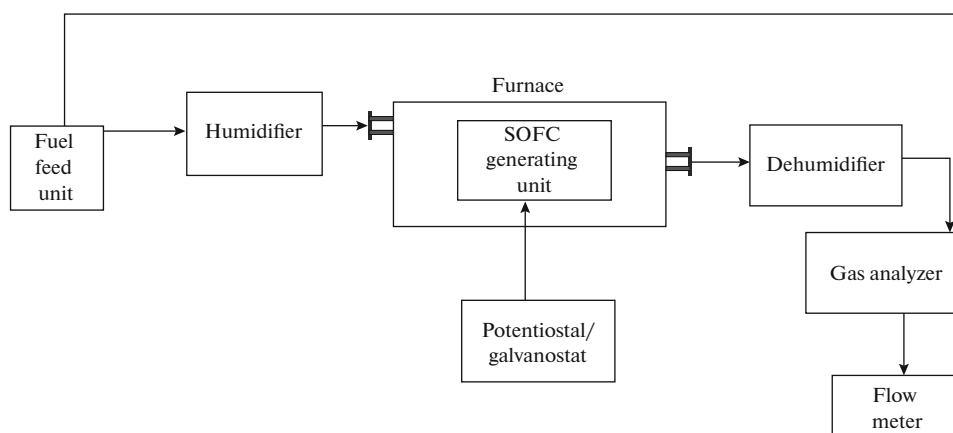


Fig. 1. Scheme of the setup for the experiment.

plant with an efficiency factor (el) of 50–55% and an efficiency factor (therm) of  $\approx 30\%$ . In this case, the electrical efficiency was estimated taking into account the proportion of methane in the initial biogas.

This paper is devoted to estimating the landfill gas technical potential for the heat and electricity production in Russia, taking into account the developed SOFC materials and technologies of the Institute of Solid State Physics, Russian Academy of Sciences. Using model landfill gas composition generated based on literature review data, we studied the SOFC operation to derive the actual cells' performance curve. The energy costs for purification and preparation of fuel gas for SOFC were estimated based on literature data. The scale of the laboratory stack was aimed at demonstrating the possibilities of utilizing landfill gas and generating electrical energy from it. Based on statistical data, we made an assessment of the annual formation of landfill gas at Russian MSW landfills (gross potential of landfill gas). The energy that can be obtained from this gas using ECG SOFC (technical potential of landfill gas) was estimated using experimental results at the Institute of Solid State Physics of the Russian Academy of Sciences.

## MATERIALS AND METHODS

To study the SOFC electrochemical performance under conditions of utilization of model waste gases, bipolar current collectors [12, 13] and end cathode and anode plates were fabricated. Electrical contact between the ceramic cathode and metal connectors was provided using the LSM( $\text{La}_{0.8}\text{Sr}_{0.2}$ ) $_{0.95}\text{MnO}_{3-d}$  contact composition [14]. A high-temperature glass-ceramic sealant was used to separate the gas spaces and seal the assembly. All structural elements were assembled into a generating block ("assembly"). A generating unit of two SOFCs was installed in a test stand (Fig. 1).

The model composition of the gas mixture was set using mass flow controllers from Bronkhorst (Nether-

lands) in the fuel supply unit, after which it was humidified in a Fideris humidifier (Canada) for further steam reforming of methane. After humidification, the gas was supplied to the SOFC generating unit, which was thermally stabilized in a furnace at a given temperature under a mechanical load of  $0.2 \text{ kg/cm}^2$ . Methane conversion took place in the generating unit. After that, the conversion products passed through a dryer and were fed into a gas analyzer manufactured by OOO Boner (Russia) based on Siemens sensors (Germany) to control the gas composition and then into a flow meter.

The parameters under study were measured using a Reference 3000<sup>TM</sup> potentiostat/galvanostat (Gamry) with an external Reference30K Booster<sup>TM</sup> current amplifier connected to it. The device allows for measuring the maximum voltage of  $\pm 32 \text{ V}$  at a maximum current of  $\pm 30 \text{ A}$  and perform electrochemical impedance spectroscopy (EIS) measurements with a frequency of up to  $300 \text{ kHz}$ .

In accordance with the selected sintering program for sealing and activating the generating unit, the following actions were carried out under a load of  $5 \text{ A}$  at constant gas flows before electrochemical measurements:

1. heating the block to  $500^\circ\text{C}$  at a rate of  $1^\circ\text{C}/\text{min}$  in an air atmosphere;
2. exposure at  $500^\circ\text{C}$  for 3 h;
3. heating to  $940^\circ\text{C}$  at a rate of  $2^\circ\text{C}/\text{min}$  in a nitrogen atmosphere from the anode side and in air from the cathode side;
4. exposure at  $940^\circ\text{C}$  for 3 h; after exposure for 1 h, the atmosphere in the anode chamber was changed to a mixture of nitrogen/hydrogen (component ratio of 90/10); and
5. cooling to operating temperature of  $850^\circ\text{C}$  at a rate of  $1^\circ\text{C}/\text{min}$ .

Experimental studies on the SOFC generating unit were carried out under the following operating conditions:

1. operating temperature set by the high-temperature furnace controller of  $850^\circ\text{C}$ ;

2. composition of the fuel mixture (in terms of dry gas), % by volume: 47.4 CH<sub>4</sub>, 52.6 CO<sub>2</sub>;

3. flow of fuel mixture components, cm<sup>3</sup>/min: 86 CH<sub>4</sub>, 95.5 CO<sub>2</sub>;

4. composition and flow of the oxidizing mixture: 2.5 dm<sup>3</sup>/min air;

5. relative humidity of the fuel mixture: from 12 (when entering the mode) to 20% (during tests).

### EXPERIMENTAL

Figure 2a shows the current-voltage and power characteristics of the SOFC generating unit as a dependence of voltage  $U$  and specific power  $P$  on current  $I$ . Air was used as an oxidizer at a flow rate of 2.5 dm<sup>3</sup>/min, and hydrogen was used as a fuel at a flow rate of 0.6 dm<sup>3</sup>/min. The relative humidity of the gases was equal to 12%, the output power was equal to 35.75 W at an assembly voltage of 1.4 V, and the specific power was equal to 0.22 W/cm<sup>2</sup>. Peak power was not achieved due to the current limit of 30 A; the power at this current was equal to 40 W.

Taking into account the expected steam reforming of the methane contained in the landfill gas in the membrane-electrodeassembly (MEA) at 850°C, the composition of the fuel mixture was determined, % (by volume):

H <sub>2</sub>	59.5
H <sub>2</sub> O	22.3
CO	13.0
CO <sub>2</sub>	5.1
CH <sub>4</sub>	0.1

This composition is close to thermodynamic equilibrium, which is typical for nickel catalysts, % (by volume):

H <sub>2</sub>	59.5
H <sub>2</sub> O	22.5
CO	13.0
CO <sub>2</sub>	5.0
CH <sub>4</sub>	0

At the same time, the specific volumetric fuel consumption for SOFC (H<sub>2</sub> + CO) is 172 cm<sup>3</sup>/min per 1 MEA, which requires landfill gas consumption in terms of dry gas of 90.8 cm<sup>3</sup>/min, i.e., 86.0 cm<sup>3</sup>/min CH<sub>4</sub> (47.4% by volume) + 95.5 cm<sup>3</sup>/min CO<sub>2</sub> (52.6% by volume). Air (2.5 dm<sup>3</sup>/min) was used as an oxidizing agent. The relative humidity has been increased to 20%.

Figure 2b shows the current-voltage and power characteristics of the SOFC generating unit when using model landfill gas as a fuel. One can observe that

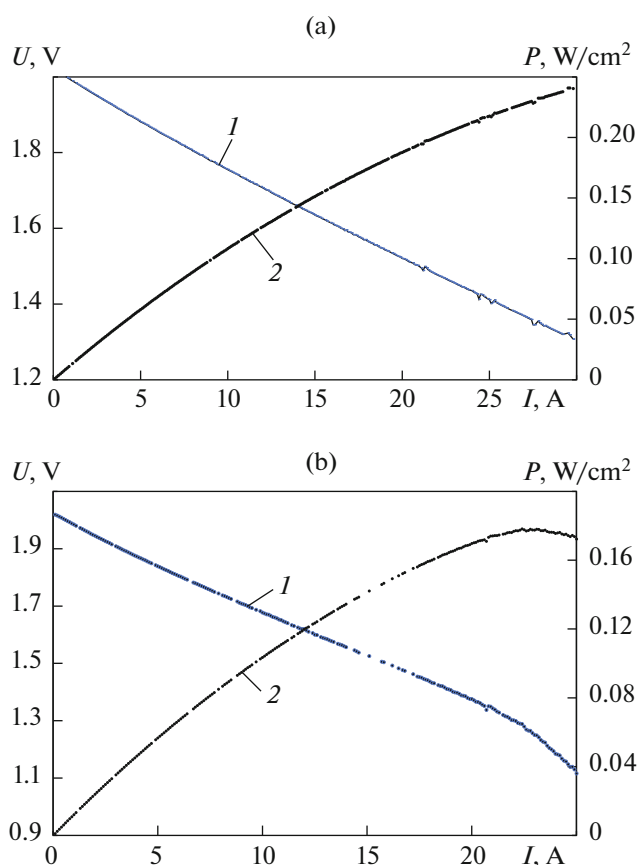


Fig. 2. Current-voltage and power characteristics of an assembly of two SOFCs using (a) pure hydrogen and (b) landfill gas as fuel. 1— $U$ ; 2— $P$ .

the power at an assembly voltage of 1.4 V was approximately equal to 27.0 W, and the specific power was 0.17 W/cm<sup>2</sup>. At a current of 23.1 A, these indicators were equal to 28.8 W and 0.18 W/cm<sup>2</sup>, respectively. The gas composition was measured using a gas-analysis system. The content of methane in the exhaust gases was 0.01% (by volume), which indicates a high degree of its utilization. Thus, we can conclude that a device with a power of 1 kW consumes approximately 6.3 dm<sup>3</sup>/min of fuel in terms of dry gas.

In order to check the possibility of studying the electrochemical performance, the hodographs of the impedance spectra for each of the MEAs that make up the SOFC generating unit were studied under open circuit conditions at various compositions of fuel mixtures. Figure 3 shows the impedance hodographs of one of the SOFC MEAs included in the generating unit when hydrogen and landfill gas are used as fuel. The results show that the main contribution to the total internal resistance of the cell under study is made by the processes occurring on the SOFC electrodes. The ohmic resistance hardly changes when changing the fuel used.

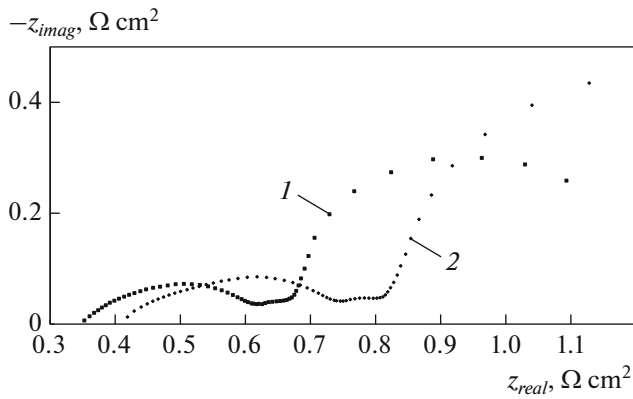


Fig. 3. Impedance spectrum of one MEA using (1) pure hydrogen and (2) landfill gas as fuel.

Despite the fact that the experimental results obtained gave us the opportunity to proceed to quantitative assessments of the technical potential of landfill gas application in SOFCs, additional studies of performance time stability are required, which will be done in further works.

#### ASSESSMENT OF THE GROSS LANDFILL GAS POTENTIAL IN RUSSIA

To estimate the potential annual release of landfill gas from Russian MSW landfills (gross potential), several assumptions were made. The technique for determining methane emissions from MSW landfills,

$$E_{\text{CH}_4} = \left( MSW \times MCF \times DOC \times DOC_f \times F \times \frac{16}{12} - R \right) (1 - OX), \quad (1)$$

where  $MSW$  is the total amount of wastes delivered to the landfill per year;  $MCF$  is the methane flux correction factor, reflecting the depth of the waste bedding in the landfill body (typical value is 0.6);  $DOC$  is the proportion of potentially decomposing organic matter (determined by the wastes composition);  $DOC_f$  is the proportion of organic matter that actually decomposes (typical value 0.77);  $F$  is the methane proportion in the gas generated at landfills (typical value 0.5);  $16/12$  is the conversion factor for C content to  $\text{CH}_4$  content;  $R$  is the consumption of utilized methane (in the calculations of the authors it is assumed to be 0); and  $OX$  is the oxidation number (usually 0).

For assessing  $DOC$ , the morphological composition of  $MSW$  in the Moscow oblast was used (Fig. 4). The composition of decaying organic matter waste was determined from the carbon content in paper and textile materials (40%), food waste (15%), non-food waste from parks and gardens (17%), and wood waste and straw (30%). The key characteristic for assessing the yield of landfill gas and methane in its

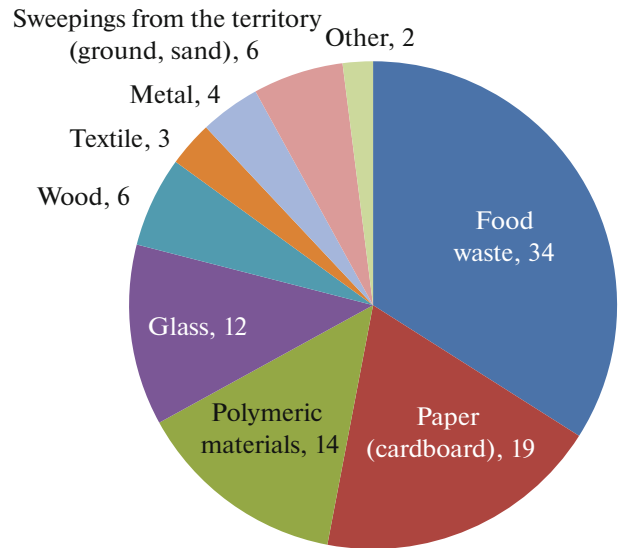


Fig. 4. Morphological composition of municipal solid waste [17].

described in [15] and tested in [16] was taken as the basis for quantitative assessments of the gross potential of landfill gas and methane in its composition. It was assumed that the annually generated landfill gas volume is proportional to the amount of MSW delivered to landfills annually (i.e., the annual volume of waste reaches the active methanogenesis stage). Thus, the annual methane emission, thousands t/year, from the MSW landfill can be estimated as

composition is the annual generation of municipal solid waste. Waste-generation rates vary in different regions of Russia and depend on the season; however, the following average rates were adopted at 50% humidity for generalized estimates [18]:

1. for urban residents: 1.2 kg/(person day);
2. for rural residents: 0.52 kg/(person day) (it is assumed that in rural areas food waste is used to feed pets and poultry and is not included in the waste).

Landfills in rural areas are much smaller and dispersed. In practice, degassing systems for these landfills are rarely used. Therefore, calculations of the methane yield, as a high-calorie component of landfill gas, were carried out only taking into account the urban population of Russia as of January 1, 2021 [19]. The total amount of landfill gas generated annually in Russia amounted to  $8800 \times 10^6 \text{ m}^3/\text{year}$ , including  $4200 \times 10^6 \text{ m}^3/\text{year}$  of methane (2722000 t/year). Using the same method, the volumes of landfill gas and methane in its composition were estimated for large Russian cities with a population of at least



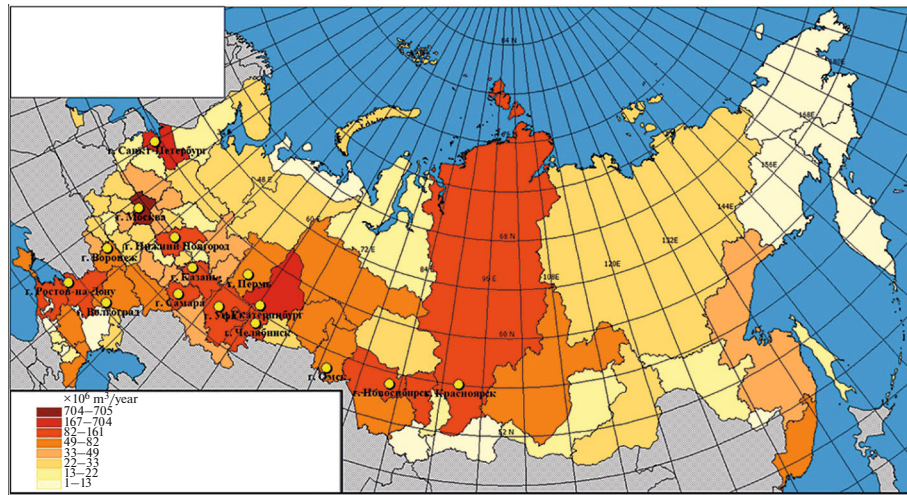


Fig. 5. Gross potential of methane in the landfill gas composition for the subjects of Russia.

100000 people (117 cities in total) [19]. The total amount of landfill gas generated annually in large cities of Russia amounted to  $5490 \times 10^6 \text{ m}^3/\text{year}$ , including  $2600 \times 10^6 \text{ m}^3/\text{year}$  of methane (1685000 t/year). Depending on the urban population distribution within the region, administrative centers generate from 30 to 100% of MSW volumes and, accordingly, landfill gas and methane. The estimation results are shown in Fig. 5, which shows the distribution of the volumes of generated methane without detailing its distribution over the territory of the subjects.

#### ASSESSMENT OF TECHNICAL ENERGY POTENTIAL IN CASE OF SOFC APPLICATION FOR LANDFILL GAS UTILIZATION

To estimate the potential production of electricity from landfill gas, the following assumptions were made:

a cogeneration SOFC-based power plant is installed at the MSW landfill. The two-stage landfill gas-purification system is applied (based on water cooling and sorption of impurities on activated carbon);

both thermal and electrical energy are fully utilized by grid and the nearest settlement heating system;

thermal energy output (0.47 kW h/(kW h) (e)) and its consumption for its own needs  $\varphi = 7.7\%$  of the output for power supply of water cooling units, booster compressor, supercharger correspond to the results of the DEMOSOFC project [11];

the specific consumption of landfill gas  $q$  for the production of 1 kW h (e), according to the experimental data obtained at the Institute of Solid State Physics (Russian Academy of Sciences) is  $0.4 \text{ m}^3/\text{h}$ .

The expression for the power output  $W_e$  for specific polygons and regions can be written as

$$W_e = qE \frac{1}{\omega} \left(1 - \frac{\varphi}{100}\right), \quad (2)$$

where  $\omega$  is the methane's share in the landfill gas composition and  $E$  is landfill gas output.

Despite the fact that, during the gross potential of MSW landfill assessment, the yield of both methane and landfill gas was calculated, landfill gas yield was taken into account in the assessments of the technical potential, and  $q$  value was determined for a mixture of gases, taking into account possible diffusion restrictions from carbon dioxide. The expression for thermal energy  $W_h$ , taking into account the data of the DEMOSOFC project, can be represented as [11]

$$W_h = 0.47W_e. \quad (3)$$

The results of calculations using formulas (2) and (3) showed that the available electrical energy when using SOFC can be 3278 million kWh/year and thermal energy can be 1512 million kWh/year for all MSW landfills in Russia. Of these, 2045 million kWh/year and 930 million kWh/year are in large cities. As expected, the technical potential is concentrated in large urban agglomerations, which simplifies the task of the generated electrical and thermal energy utilization.

#### CONCLUSIONS

(1) The SOFC stack operation on model landfill gas without an external methane steam-reforming unit was experimentally demonstrated. Further it makes possible the effective conversion of natural gas and other methane-containing mixtures.

(2) The annually generated landfill gas technical potential for the territory of Russia is 3278 million kWh/year of electric energy and 1512 million kWh/year of thermal energy in the case of SOFC application for its utilization.

(3) The electric power of individual power plants is 1.0–1.5 MW, which opens up wide opportunities for the domestic SOFC market development within the framework of programs for energy efficiency and reclamation of MSW landfills.

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#### CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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