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8 Abstract: Renewable energy (RE) sources like wind and solar power have been introduced to 9 power systems to make the energy market more sustainable and environmentally friendly. 10 For this reason, the supply-demand mismatch in power systems is more severe than before, 11 in terms of both frequency and magnitude. It is therefore necessary to reinforce the 12 capability to make supply and demand adjustments in power grids. Here, we clarified the 13 dynamic characteristics of biogas plants with combined heat and power (CHP) systems, that 14 represent controllable RE power sources, and evaluated the possibility of using these systems 15 as a new resource for supply-demand adjustments in the power grid. Additionally, the 16 properties of the exhaust gas from the investigated CHP engines were measured as an 17 environmental evaluation. We considered real-world biogas CHP systems operating in both 18 Germany and Japan. Based on the results, biogas CHP systems have the potential to 19 contribute to a long-term power equivalent adjustment such as tertiary control reserve 20 (TCR) in Germany. However, these systems required several minutes between starting and 21 stopping. Furthermore, increased emissions of methane and formaldehyde were measured 22 when the CHP systems were starting up. We found that an operation with as few starts and 23 stops as possible is therefore desirable.

24

25 Keywords:

26 Biogas, Combined heat and power (CHP), Load response, Power control, Emissions

27

28 1. Introduction

29 With the introduction of renewable energy (RE) power sources-whose output depends

30 greatly on weather conditions-maintaining a real-time supply-demand balance becomes

31 increasingly challenging. Due to such operational difficulties, ten electric power companies

32 have been granted regional monopolies in Japan. In August 2011, the Special Measures Law

concerning the procurement of RE electricity by electric utilities was enacted, while on July 33 34 1, 2012 a, "Feed-in Tariff Scheme for Renewable Energy" (FIT), requiring electric utilities 35 to procure electricity generated using solar, wind, hydro, geothermal, and biomass for a fixed period at a fixed price set by the government, was introduced [1]. Power supply facilities 36 37 were greatly affected by the Great East Japan Earthquake of March 11, 2011. Hence, there 38 are growing expectations in Japan for a power source that can reduce greenhouse gas 39 emissions while simultaneously ensuring energy security. The Japanese Ministry of 40 Economy, Trade and Industry has also announced a policy to increase the ratio of RE power sources in the power supply mix by 2030 from about 22% to 24% [2]. Hence, the 41 42 supply-demand mismatch is more severe than before in terms of both frequency and 43 magnitude, and it is necessary to reinforce the supply-demand adjustment capability. Until 44 now, supply-demand adjustments have been resolved by either increasing or decreasing the 45 output of conventional power sources, such as thermal power plants, but the power generation capacity of such sources has decreased, and therefore the supply-demand 46 47 adjustment capability of the power system has decreased. [3]. Additionally, conventional 48 power plants, such as coal-fired plants, have an inferior dynamic performance and long cold 49 start-up times of up to 48 hours [4]. In recent years the stabilization of power grids by 50 controlling battery storages has been studied [5][6].

51 In many European countries the challenge of managing the fluctuations from renewable 52 energy sources, such as wind power, has already been addressed. As far back as 2002 a study 53 in Denmark investigated a self-supply strategy by managing the fluctuation from wind 54 power with combined heat and power (CHP) units and selling excess electricity to 55 neighboring countries [7]. In Germany and other parts of Europe, where FITs have been developed and RE power is increasing, CHP systems, such as biogas plants, are already 56 57 being used to regulate capacity in transmission lines. In Germany, additional adjustment 58 capacity is procured by the grid operator through auctions a day or week in advance. The 59 power to be procured consists of three levels: primary control reserve (PCR), secondary 60 control reserve (SCR) and tertiary control reserve (TCR) that are required to supply electricity within 30 s, 5 min and 15 min, respectively. German biogas CHP systems can 61 62 participate in this market, typically as SRC or TCR, with operators of the power control pool 63 managing individual plants and dealing with grid operators. Since the biogas plant would be 64 remotely controlled by the pool operator, it is necessary to respond quickly to increasing 65 power demand, and to shut down the CHP system when grid power becomes excessive [8]. 66 When a CHP system responds to a supply-demand target as an adjustment capability, 67 responsiveness (speed) and adjustment capability (control range) are important parameters. 68 Many studies have been published regarding the optimization of operational strategies, for 69 example for the day-ahead spot market [9], by increasing the power [10] and storage 70 capacity of biogas plants [10, 11], by integrating heat pumps with CHP units [12] or by utilizing local resources ahead of central power plants [13,] or the profitability, by using 71 72 thermal storage and Organic Rankine Cycle (ORC) technology [14], by simulating the 73 effects of providing positive secondary control energy with biogas plants [15], by comparing 74 operation of plants using solid, liquid and gaseous biofuels and the legislative and market 75 conditions in Germany [16], by simulating different operational scenarios of a biogas plat 76 participating in the spot market and control energy reserves [17] or by conducting an 77 economical and ecological assessment based on different biogas plant configurations [18], of 78 CHP systems. In a previous study a demand response program for an industrial customer 79 was implemented using stochastic programming to solve short-term self-scheduling 80 problems for CHP systems and enable them to participate in the power market more 81 successfully [19]. In a different study the same authors investigated different demand 82 response programs to serve the power and heat demands of the customer with minimum cost 83 [20].

84 Another study has clarified that a CHP system can follow various load fluctuations in a 85 microgrid [21]. Aghaei et.al. [22] investigated optimal operation strategies for CHP based microgrids regarding minimizing operational costs. The study shows that by implementing 86 87 an optimized demand response program the operational costs could be reduced considerably 88 [22]. A further study showed the necessity of precise assessment of reserve capacity of CHP 89 units when managing load fluctuations in the power grid, when operating such units in 90 conjunction with district heating networks [23]. These studies show that extensive work has 91 been done regarding optimizing operational strategies of various CHP systems. However, all 92 these evaluations have used simulations; there are few characteristic evaluations based on 93 measurements using actual CHP systems. Furthermore, they do not address the specific 94 demands of rural biogas CHP systems. Regarding actual on-site measurements Tappen et.al. 95 [24] investigated the efficiency of 8 biogas CHP system over a long period of time (40,000 96 to 60,000 operating hours). They have confirmed that the continuous operation of biogas 97 CHP systems under a partial load is accompanied by a significant decline in electrical 98 efficiency [24].

- 99 In this study, we clarified the dynamic characteristics of biogas CHP systems that represent
- 100 controllable power sources among RE systems and evaluated the possibility of load response
- 101 to utilize biogas CHP systems as a new supply-demand adjustment resource to supplement
- 102 the output of naturally fluctuating power sources in rural Japan, where the management of
- 103 load fluctuations in the power grid is very difficult [25].
- 104 The emphasis of the on-site measurements was on the load response of the biogas CHP

105 systems. Additionally, we measured the exhaust gas emissions during transient operation to 106 evaluate the environmental impact of part load and load response operation. In previous 107 studies both the greenhouse gas emissions, such as carbon dioxide (CO_2) and methane 108 (CH_4) , [26, 27, 28, 29] and the pollutant emissions, such as nitrous oxides (NO_X) or carbon 109 monoxide (CO) [26, 29, 30] have been investigated with on-site measurements at nominal 110 load. When regarding GHG emissions the CHP exhaust emissions were identified as the second major source of methane emissions at biogas plants, after open storage of digestate 111 112 [26, 28]. Considering the pollutant emissions under current Japanese law, biogas CHP 113 systems do not fall under the category of fuel stipulated in the Air Pollution Control Law, so 114 emissions are unregulated. However, in Europe such emissions regulations exist [31] and 115 particularly in Germany the current standards for CHP systems with an electric rated output 116 of over 1 MW were enforced recently [32]. A study showed, that when investigating 10 117 biogas CHP systems in Italy, the investigated biogas plants were within prescribed limits, as well as the German limits, for NO_X and CO over one year of monitoring, while transient 118 119 operation was not specifically analyzed [27]. However, there are still very few studies 120 regarding the environmental impact of part load and transient operation of biogas CHP 121 systems. Hijazi et.al [33] investigated the specific greenhouse gas (GHG) emissions of 122 biogas CHP units in flexible operation. They found that the specific GHG emissions under 123 part load are higher than at full load due to the decrease in efficiency of the CHP unit at part 124 load [33].

125 This study is necessary to utilize various resources in future power systems, with the 126 viewpoint of increasing naturally fluctuating power sources, while maintaining stable 127 operation of the power grid in rural areas of Japan. Additionally, the environmental impact 128 of the management of fluctuations in the power grid is of the utmost importance when 129 regarding the future operation of biogas CHP systems. This paper intends to bridge the gap 130 between the proven technologies and practices in Germany and the current state in rural 131 Japan, while considering the importance of environmentally friendly solutions and 132 sustainable operation. Being a global issue, it is imperative to secure appropriate adjustment 133 capacity efficiently and effectively by expanding the introduction of natural power sources, 134 while utilizing currently available infrastructure, such as biogas plants.

135

136 2. Methodology

137 Nine biogas CHP systems from Japan and Germany generating electricity using biogas 138 produced by fermentation were targeted. The raw material for the biogas was a mixture of 139 corn feed and pig/dairy cow manure in Germany and dairy cow manure in Japan. Since the 140 CHP systems were practical ones operating on farms in both countries, the properties of biogas used in each CHP system were not uniform. The methane concentration of the biogas on the German biogas plants ranged between 55% and 65%. All German biogas plants implemented desulfurization of the biogas. At most small traces of H₂S were detected in the biogas. Additionally, two natural gas CHP systems were included for comparison to the biogas CHP systems. These units were operated with so-called H-Gas (high-calorific gas), which had a methane concentration of over 94% at the time of the measurements. Table 1 gives an overview of the investigated CHP systems.

- 148
- 149

Table 1 Overview of Biogas CHPs and Natural gas CHPs

Category	tegory Biogas CHPs*1								Natural gas CHPs*2		
No.	G BG 1	G BG 2	G BG 3	G BG 4	G BG 5	J BG 1	J BG 2	J BG 3	J BG 4	G NG 1	G NG 2
Nominal Electric Power(kW)	200	549* ³	255	901	400	600	50	100	160*4	250	20
Output range				50%-10	0%					50%- 100%	25%- 100%
Year of Commissioning	2011	2006	2016	2018	2010	2015	2016	2015	2018	2019	2016
Country*5	G	G	G	G	G	J	J	J	J	G	G
Catalyst*6	OC	TPC	OC	OC	OC	Unkn own	ос	OC	OC	ос	3way Catalyst

150

151 *1 Biogas CHPs: Biogas combined heat and power systems

152 *2 Natural gas CHPs: Natural gas combined heat and power systems

153 *3 CHP unit runs at a reduced power output of 500 kW

154 *4 CHP unit runs at a reduced power output of 150 kW

155 *5 G: Germany, J: Japan

156 *6 OC: Oxidation Catalyst, TPC: Thermal Post-Combustion

157

158 The load responsiveness depending on the output, as well as the startup and shutdown

159 characteristics were investigated for all CHP systems. Fig.1 shows the measurement setup.

160 An electric power logger was installed after the generator of each CHP system. Using a

161 PW3360¹ clamp-on power logger, voltage (V), current (A), and active power (kW) were

- 162 measured at intervals of 1 s.
- 163



164 165

166

Fig.1 Measurement Setup

- 167 The startup response time $\Delta t_{startup}$ was defined as the interval between the start command
- (0 s) and when the CHP system reached nominal load (100%). This interval can be split intotwo phases: the synchronization phase and the load ramp (Fig.2).



170 171

172

173 The step response time Δt_{step} was measured as the interval between giving the step command 174 and the CHP system reaching the specific load (Fig.3). The command value was changed in

175 10% steps starting from nominal load and ending at 50% of nominal load, before increasing 176 the power output stepwise back up to nominal load. Based on the step response time Δt_{step} 177 and the value of the load step $\Delta load_{step}$, the step response speed or ramp m_P could be 178 calculated [34].

179

$$180 \quad \Delta load_{step} = (P_1 - P_2) * P_{nominal} \tag{1}$$

181
$$m_P =$$

 $\Delta load_{step}$

 Δt_{step}



182 183

184

Fig.3 Step response time

185 The environmental impact of the five German biogas CHP systems and two natural gas 186 CHP systems were also analyzed. The concentration of components from the exhaust gas of the CHP systems were measured using a Fourier Transform Infrared (FTIR) CX40002 187 188 spectrometer. Gas was sampled continuously from the exhaust pipe of the CHP systems 189 downstream from the exhaust gas aftertreatment with a heated probe (Fig. 1). The exhaust gas was supplied to the measuring device using a heated flexible tube that was constantly 190 heated to 180 ° C to prevent water precipitation in the device. Residual oxygen in the 191 exhaust gas was measured using a PMA10³ paramagnetic oxygen analyzer and was measured 192 dry, i.e., when cooled to 5° C and water vapor is precipitated and removed from the sample. 193 During the step response and startup/shutdown experiments, concentrations of CO, NO_X , 194 195 CH₄), formaldehyde and sulfur dioxide (SO₂) in the exhaust-gas were measured. These 196 values represent dry exhaust-gas by subtracting measured amounts of water vapor from 197 exhaust-gas. For the step response tests the concentrations of exhaust-gas components were 198 analyzed as mass concentrations (mg/m^3) and referred to a residual oxygen concentration of 199 5 Vol.-% as specified by the German Emissions Standard TA Luft [35]. During the startup

(2)

and shutdown tests the direct volumetric exhaust-gas concentrations were analyzed. These results were not referenced to 5 Vol.-% residual oxygen to prevent a distortion of the measurements due to the varying residual oxygen content during the startup and shutdown phases. All measurements were performed downstream form the exhaust gas catalyst. The catalysts were in variable state of conditions due to the investigated biogas plants being real-world plants in actual operation. No in-depth inspections of the catalysts or measurements were made during the measurement campaign.

- 207
- 208 3. Results and discussion
- 209 3.1 Biogas CHP system performance
- 210 3.1.1 Electric and thermal outputs efficiency

211 Fig. 4 shows the values of electric efficiency and thermal output efficiency of each CHP

212 system at nominal load as published in the manufacturers data sheets.





Fig.4 Electric, Thermal output and overall Efficiency of the CHP systems

215 *1 Biogas CHPs: Biogas combined heat and power

216 *2 Natural gas CHPs: Natural gas combined heat and power

217

218 CHP systems, with a high electrical output, were more efficient than smaller CHP systems,

219 with the exception of "J-BG-4", a state-of-the-art biogas CHP system with a high electrical

220 output efficiency. "J-BG-1" had an extremely low thermal output efficiency at 23.0%, due to

the manufacturer designing this unit considering the low heat utilization potential on site.

While "G-BG-3" and "G-BG-5" both reached high thermal output efficiencies of 50.2%. In general, the thermal output efficiency is not only dependent on the technology used for the CHP system, but also on the intended application of the heat. The thermal output efficiency depends on the heat utilization. The average overall efficiency (electrical output + thermal output) of German and Japanese CHP systems was 84.9% and 78.4%, respectively. "G-NG-1," a natural gas operated unit, uses a turbocharged lean burning engine. The second baseline system, "G-NG-2," is a naturally aspirated and stoichiometrically operated.

229

230 3.1.2 Startup dynamic characteristics

Fig. 5 shows the load characteristics during the startup process of the CHP systems.



233

232

Fig.5 Start-up response

234 *1 BG: Biogas CHP

- 235 *2 NG: Natural gas CHP
- 236

This represents the time that elapsed from the start command given to each device until it reached the rated output. "G-BG-1," "G-BG-5," and "G-BG-2" took 72 s, 124 s and 211 s, respectively, until the rated output was reached. This positively affected the stability of the power output. Although it would be possible for these systems to contribute to TCR and SCR, a PCR supply from a central power plant or storage battery would still be required.

"G-BG-4" and "G-BG-3" showed slower response times during the startup process than the other German systems reaching rated loads in 324 s and 310 s, respectively. Moreover, they show increased power output fluctuations in two separate situations: During the startup process for both CHP systems; and, before the engine of "G-BG-3" could reach stable ratedoperation.

All Japanese biogas CHP systems took more than 400 s to reach the rated output from the start command. "J-BG-2" has a better startup responsiveness than "J-BG-4" and they reached their rated loads in 247 s and 456 s, respectively. It was observed that regardless of

the size of a CHP system, the output initially increased to about 40–50% of the rated output

- after startup, and then, after a short stable operation at this load, continued increasing until
 it reached the respective rated output.
- 253 "G-NG-2," being a relatively small engine, reached the rated output at a faster rate, i.e.,

within 116 s, compared to "G-NG-1" (299 s). "G-NG-1," which was issued with settings to

255 provide a SCR on the German power control market, reached the rated output in under the 256 required 5 min limit, but as slowly as possible to provide the most stable startup process

- 257 possible.
- 258
- 259 3.1.3 Shutdown dynamic characteristic
- 260 Fig. 6 shows the load characteristics when the CHP systems are shutdown.



261 262

263 *1 BG: Biogas CHP

264 *2 NG: Natural gas CHP

265 This indicates the elapsed time from the issuance of a stop command during rated operation

- 266 until the output reaches 0 kW. After the generator connection to the grid is cut, the engines
- 267 of the CHP systems have a 30 s to 60 s cool down phase in idle operation that was not

268 included in the evaluation. Three general response characteristics could be defined from the

269 experimental test once the stop command is issued: (1) The output of the stop characteristic

- decreases linearly. (2) The output decreases linearly to about 30–70% of rated output, then
- 270 decreases linearly. (2) The output decreases line
- after a short period that load decreases to 0 kW at a faster rate. (3) The output decreases linearly to about 10% of rated output and after a certain time is reached at this load, the
- 273 CHP shuts down.
- 274 For "G-BG-5", "G-BG-4", "G-BG-2" and "J-BG-2", the behavior classified in (1) is 275observed. The output decreases at a constant rate for these systems. This is not suitable for 276 applications requiring high load responsiveness. The time required for the power output of 277 these systems to reach 0 kW was 101 s, 162 s, 163 s and 120 s, respectively. The result of "J-BG-1" is a stop characteristic based on an automatic shutdown which occurs when the 278 279 level of the gasbag of the biogas plant reaches a certain value. This CHP system does not 280 include a manual stop mechanism, so only startup and automatic shutdown responses could be assessed. "G-BG-3" and "J-BG-4", show a behavior classified in (2). They adopt a control 281 282 program that does not generate a sudden load drop, even when stopped to protect the 283 generator. The time required for the power output of these systems to reach 0 kW was 64 s and 83 s, respectively "J-BG-4" had an output fluctuation: A short stoppage time of 59 s, 22 s 284 285 and 2 s from the stop command to 50%, 30%, and to 0 kW of the rated output, respectively, 286 were realized. "G-BG-1", displayed a behavior classified in (3). It reduced its load from the 287 stop command to 10% of the rated output in only 39 s, but continued operating at this load 288 for about 2 min. This CHP system uses an asynchronous generator, i.e., it is always 289 connected to the grid when operating even in the 2 min cool down phase at 10% load. Such 290 a generator responds quickly during startup, and can be suitable for a quick shutdown, if 291 only 90% of its capacity is defined as control reserve.
- It was observed that the response speed of the shutdown was faster for smaller CHP systems.
 The shutdown characteristics defined by (1) protect the CHP system from ramp load
 fluctuations best, followed by those defined by (3) and (2).
- "G-NG-2" had a quick shutdown time of 109 s due to the small size of the engine while
 "G-NG-1" took 26 s to shut down, but still representing a short shutdown time, considering
 the size difference of the engine.
- 298
- 299 3.1.4 Step response characteristics
- 300 Fig. 7 shows the rate of change when the value of issued command is increased by
- 301 increments of 10% starting from minimum output of 50%. The results represent the mean
- 302 values from the start command until the unit reaches the new set-point. The electric output
- 303 was measured in 1 s intervals.



304



Fig.7 Step responses (Increase output by 10%)

- 306 *1 BG: Biogas CHP
- 307 *2 NG: Natural gas CHP
- 308

309 Fig. 8 shows the rate of change when output values are decreased by factors of 10% starting

- 310 from rated output.
- 311



- 314 *1 BG: Biogas CHP
- 315 *2 NG: Natural gas CHP
- 316

312 313 317 Fig. 7 and 8 help us evaluate the response characteristics of the biogas CHP systems. The

318 horizontal axis depicts the ratio to the rated output (utilized output) and represents the

319 operating state (output before change) at the time of issuing the command. The vertical axis

320 is the output change rate (%/s) based on the rated capacity of each CHP system. Fig. 7 and

- 321 8 show that the larger the absolute value of output change rate for each 10% output step, the
- 322 better the response characteristics.
- As per Fig. 7, when the output was changed from the 90% state to the rated output (A), there was no clear difference in response speed among all CHP systems which was found to be between 0.11 %/s and 0.65 %/s. The German CHP systems showed high response characteristics with increasing output (B). The German systems achieved an average response speed of 0.50 %/s to 0.69 %/s, while the Japanese systems achieved 0.13 %/s to 0.26 %/s. The Japanese systems showed a stable response speed regardless of output band.
- 329 Of the German CHP systems, "G-BG-5" showed a particularly fast response characteristic
- 330 when the output was changed from the 60 % (0.97 %/s) to 70 % (1.01 %/s) (C). "G-BG-1"
- and "G-BG-4" generally showed stable response characteristics regardless of the operatingstate at the time the command was issued (D).
- It can be seen from Fig. 8, that the Japanese systems took longer to respond than German systems when the output was decreased (E). The average response speed over all 10 % steps was found to be between -0.27 %/s and -0.14 % /s. The values for the German systems were between -1.05 %/s and -0.46 %/s. "G-BG-1", "G-BG-4" and "G-BG-2", which exhibited stable response characteristics when the output was increased, also exhibited a stable response characteristic when it was decreased. These CHP systems had a fast response characteristic (F).
- 340 "J-BG-2", "J-BG-3" and "J-BG-4" are German-manufactured CHP systems, but this test 341 confirms that the systems do not show the same high response and flexibility as the German
- 342 CHP systems. The average response speed of natural gas CHP systems of the same
- 343 manufacturer ("J-BG-2" to "J-BG-4") is low (0.17 %/s). "G-NG-1" was set at the minimal
- responsiveness setting to provide SCR on the power control market in Germany.
- 345 We focused on biogas CHP systems of the same manufacturer with different models,
- 346 "G-BG-2" and "G-BG-4," and compared the response speed of output change (Table 2).
- 347
- 348

Table2 compared the response speed of output change

			(unit: %/s)		
_		G BG 2	G BG 4		
Output width		2006	2018		
-50)%	-0.59	-0.57		
-20%		-0.61(80%)* ~-0.59(100%)*	-0.59(80%)* ~-0.55(100%)*		
-10%	the fastest	-0.59 (70%, 80%) *	-0.54 (100%) *		
	the latest	-0.68 (100%) *	-0.60 (60%) *		
10%	the fastest	0.65 (90%) *	0.66 (50%) *		
	the latest	0.23 (50%) *	0.53 (90%) *		
20	%	0.59(60%)* ~0.60(80%)*	0.56(60%)* ~0.57(80%)*		
50	%	0.58	0.56		

349

350 351

* State during operation

With increasing output, the speed of the response of "G-BG-2" was 0.65 %/s at its fastest 352 point at a 90% output state. In contrast, the response was the slowest (0.23 %/s) at 50 % of 353 354 rated output. "G-BG-2" showed a high flexibility for small output changes near the rated 355 output but tended to have low flexibility near the minimum output regardless of the width of 356 the output change. On the contrary, "G-BG-4" had the fastest response speed at the lowest 357 output of 0.66 %/s, showing high flexibility; it also showed a response speed of 0.53 %/s from 90% output state to rated power, which is more than twice that of "G-BG-2" 358 359 (0.23 %/s). When output decreased, "G-BG-4" was generally faster than "G-BG-2," and the 360 response speed was almost constant regardless of the operation state. Thus, "G-BG-4" was 361 found to have improved flexibility during partial load operation compared to "G-BG-2." This 362 is consistent with the fact that the concerned company has recently been aggressively 363 pursuing development to improve the responsiveness and flexibility of power generation 364 output.

Our results correspond well with a previous study by Bär et.al. [36]. They investigated the potential of balancing intermitted power from PV with biogas CHP units. The study concluded that maximum ramp rates of 0.66 %/s and minimum ramp rates of 0.33 %/s should be considered to compensate fluctuations in PV power depending on the distribution grid [36].

The natural gas CHP system "G-NG-1" showed very consistent response times for all load increments with values ranging between 0.16 %/s and 0.20 %/s. The response times are

372 similarly consistent with decreasing load. "G-NG-2" showed very high responsiveness for

both increasing and decreasing load (50% and 60%). The responsiveness of this CHPsystem decreased as it got closer to full load.

375

376 3.2 Environmental Impact Assessment of load response

377 Exhaust-gas composition is influenced by the catalytic method for exhaust-gas purification installed in each CHP system [19]. Only "G-BG-2" adopted a TPC system. "G-NG-2" is a 378 379 stoichiometric operated engine and therefore uses a three-way catalyst. All other biogas 380 CHP systems used oxidation catalyst systems. To evaluate the effect of the load response on 381 the exhaust emissions, steps of 10% load were observed. The measurements represent value 382 of the exhaust-gas components for each operating condition. CO emissions were very low 383 when the oxidation catalyst was new, and the operational load of the CHP system did not 384 affect the emissions (Fig. 9).





386

Fig.9 The CO emissions during step responses

- 387 *1 BG: Biogas CHP
- 388 *2 NG: Natural gas CHP
- 389

This result is supported by those of the CHP systems "G-BG-1," "G-BG-3," and "G-BG-4," which were equipped with an oxidation catalyst (Fig. 4). However, "G-BG-5" had the highest CO emissions at the minimum output (50% load operation) because of the old oxidation catalyst. At partial load, the conditions in the engine (e.g., cylinder pressure and temperature) are not as high as at the rated output operation, resulting in an incomplete 395 combustion of engine-fuel and higher CO emissions in exhaust gas. When the oxidation 396 catalyst was older its performance decreased, resulting in higher CO emissions, as seen with "G-BG-5." At the rated load, CO emissions were the lowest due to complete combustion of 397 398 the engine fuel. The thermal post combustion system (TPC) maintains low CO emissions 399 regardless of the operating state of the CHP system. The CHP system operating a three-way catalyst ("G-NG-2") showed consistently low CO emissions at the rated load and a small 400peak at partial load. The performance of this kind of catalyst is influenced by the air-to-fuel 401 402 equivalence ratio of the engine, which can deviate slightly when adjusting the load. All 403 examined CHP units were within the regulatory limit for CO emissions when operating at 404 full load [35]. Only "G-BG-5" exceeded the limit at partial load due to the old oxidation 405 catalyst. NO_X emissions were highest during rated output operation regardless of the catalyst type when no NO_X control system was installed (Fig. 10). 406



407 408



409 *1 BG: Biogas CHP

410 *2 NG: Natural gas CHP

411 This is due to the "thermal-NO_X" mechanism, where NO_X forms due to high combustion 412 temperatures in the engine. Although the catalytic method of an oxidation catalyst does not

413 affect NO_x emissions, CHP systems "G-BG-1", "G-BG-3" and "G-BG-5" have NO_x control

414 systems installed. The emissions were controlled at approximately 500 mg/m³ regardless of

415 operating conditions. Here the excess air ratio of the engine is controlled based on the

- $\label{eq:stability} 416 \qquad \text{measured NO}_X \text{ in the exhaust-gas. A study showed that the excess air ratio has a direct effect}$
- 417 on the NO_X emissions. However, while increasing the excess air ratio results in a decrease in
- 418 NO_x emissions, it comes with a loss of efficiency for the CHP system [30].
- 419 The TPC system ("G-BG-2") showed almost constant NO_X emissions regardless of the
- 420 operating conditions due to the use of a NO_X control system. "G-NG-2" exhibited the lowest
- 421 NO_X emissions since it used a three-way catalyst. All CHP units with NO_X control systems
- 422 ("G-BG-1", "G-BG-3" and "G-BG-5") were within the limit of the emissions standard of
- 423 $500 \text{ mg/m}^3 \text{ NO}_X$ [19] while those with none exceeded the limit.
- 424 The CH₄ emissions in the biogas CHP units employing oxidation catalyst systems were
- lowest during rated output operation and increased with reduced output due to incompletecombustion engine-fuel (Fig. 11).





Fig.11 The CH4 emissions during step responses

- 429 *1 BG: Biogas CHP
- 430 *2 NG: Natural gas CHP
- 431

432 The oxidation catalyst has no influence on CH_4 emissions resulting in the high emissions at 433 partial load. In TPC systems, extremely low CH_4 emissions were observed regardless of the 434 operating conditions. "G-NG-2" showed very low CH_4 emissions due to a naturally aspirated 435 engine with low power output efficiency resulting in high exhaust-gas temperatures, at 436 which the three-way catalyst has optimal operational temperature to convert CH_4 emissions with a higher performance. There are currently no emissions standard for methane
emissions in Germany. However, an emissions standard for total hydrocarbons will be
introduced in 2025 for stationary combustion engines with an electric output of over 1 MW
[32].

- 441 In CHP systems with new oxidation catalysts, formaldehyde emissions were generally low
- 442 regardless of the operational load (Fig. 12). All CHP systems were within in prescribed limit
- 443 of 20 mg/m³ [32], apart from "G-BG-5" which exceeded the limit, especially at part load.



444 445

Fig.12 The formaldehyde emissions during step responses

- 446 *1 BG: Biogas CHP
- 447 *2 NG: Natural gas CHP
- 448

449 The old oxidation catalyst used by "G-BG-5" was not as effective at reducing these 450 formaldehyde emissions. Here, higher formaldehyde emissions were observed, especially at 451 partial load, due to the incomplete combustion of engine fuel. The TPC system showed extremely low formaldehyde emissions regardless of operating conditions. Formaldehyde 452 453 emissions of "G-NG-2" were very low for all operational loads. No clear relationship was 454 observed between SO_2 emissions and power output. SO_2 depends on the sulfur content in biogas, which is thought to have been affected by the desulfurization capacity before flowing 455 456 into the engine of the CHP system.

457 From these results, a TPC system is very effective for the exhaust-gas of biogas CHP units

regarding CO, CH₄ and formaldehyde. Additionally, NO_x can be controlled using a Lambda
control system. However, such a TPC system has high investment and operating costs, and
high demand for installation space.

- 461 An oxidation catalyst is effective for CO and formaldehyde, because these catalysts have
- 462 been optimized for operation due to existing German emissions standards. It affects NO_X
- 463 and CH₄ emissions only slightly at higher temperatures, however the lower investment and
- 464 operating costs of such a catalyst make it an attractive option if there is no emission standard
- for CH₄. However, the results show that the performance of oxidation catalysts decreases over time resulting in higher CO and formaldehyde emissions at the end of its lifespan.
- 467 Three-way catalysts have a high potential for converting CO, NO_X, CH₄ and formaldehyde.
- 468 However, these types of catalysts can only be applied to stoichiometric operated engines and
- 469 require exact control of the air-to-fuel equivalence ratio of the engine.
- 470

471 3.3 Environmental Impact Assessment of startup and shutdown

- 472 The startup and shutdown of the engine had an impact on the exhaust emissions of biogas
- 473 CHP systems. Parameters were unchanged from the standard stationary operations.
- 474 In terms of the CO emissions of the biogas CHP systems during startup, no peak was
- observed for the systems with new oxidation catalysts ("G-BG 1" and "G-BG-3") (Fig. 13).



476

477 Fig.13 The CO emissions during startup and shut down 478 *1 BG: Biogas CHP 479 *2 NG: Natural gas CHP 480 481 However, the system with the old oxidation catalyst ("G-BG-5") had a distinctive maximum 482 during the startup process. During shutdown of the engine peaks, CO emissions were observed for both the new ("G-BG-3" and "G-BG-4") and the old ("G-BG-5") oxidation 483 484 catalyst systems. Only one CHP system with a new oxidation catalyst ("G-BG-1") showed 485 no maximum during shutdown. The natural gas CHP unit "G-NG-1" showed consistently 486 low CO concentrations during startup and shutdown due to the installation of a brand-new 487 oxidation catalyst. The other natural gas CHP system with a three-way catalyst, "G-NG-2," showed a peak in CO at startup but none during shutdown. Similar behavior was observed 488 489 for the biogas and natural gas CHP systems in terms of CH4 and formaldehyde emissions 490 (Fig. 14 and 15).



491

492

- Fig.14 The CH4 emissions during startup and shut down
- 493 *1 BG: Biogas CHP
- 494 *2 NG: Natural gas CHP





496

Fig.15 The formaldehyde emissions during startup and shut down

497 *1 BG: Biogas CHP

498 *2 NG: Natural gas CHP

During engine startup, all systems with oxidation catalysts showed a brief maximum of CH₄
and formaldehyde emissions before decreasing during normal operation. On shutdown,
distinctive peaks in CH₄ could be seen but formaldehyde emissions were not as high.

502 The operation of the TPC system ("G-BG-2") demonstrated differing behavior in terms of 503 CO, CH_4 and formaldehyde emissions. The TPC system was in standby operation on engine

startup. Hence, high concentrations of CO, CH_4 and formaldehyde could be measured. The TPC system began operation towards the end of the engine startup process resulting in decreasing concentrations of CO, CH_4 and formaldehyde in the exhaust-gas. The natural gas system "G-NG-2" showed low CH_4 emissions during startup and shutdown.

508 The NO_X emissions are not dependent on the oxidation catalyst system or fuel used 509 (Fig. 16).



510

511

Fig.16 The NOx emissions during startup and shut down

- 512 *1 BG: Biogas CHP
- 513 *2 NG: Natural gas CHP
- 514

The emissions can differ between biogas CHP systems with NO_X control systems 515 ("G-BG-2", "G-BG-3" and "G-NG-1") and those without ("G-BG-1" and "G-BG-5"). The 516 517 NO_X emissions of all engines rose steadily until rated operation was attained. The CHP 518 systems without NO_X control systems then stayed at this level during stationary operation. 519 With installation of a NO_X control system, the lambda of the engine increases after reaching 520 rated operation until the NO_X emissions reach 500 mg/m³. During shutdown, the NO_X 521 emissions reduce steadily regardless of a NO_X control system being installed. The CHP 522 system "G-NG-2" uses a three-way catalyst that can only be used for stoichiometric operated 523 engines and can also reduce NO_X emissions. Small peaks in NO_X were seen during startup 524 and shutdown of "G-NG-2" due to some residual oxygen in the exhaust-gas that reduces the 525 performance of the three-way catalyst.

 SO_2 emissions were observed during both, startup and shutdown of the engines but were observed to be independent of the startup or shutdown. Low concentrations of SO_2 were

528 measured regardless of the load when sulfur concentrations were observed in the biogas

- 529 while no SO₂ concentrations were measured for either natural gas CHP system.
- 530 The above results show that a TPC system can reduce CO, CH_4 and formaldehyde emissions 531 effectively. However, high concentrations of the exhaust components could be measured 532 during the startup and shutdown process before the TPC system was in full operation. Cold 533 startup processes of the TPC were not considered for this study. The TPC systems generally 534 have startup processes that take several hours when initially starting operation after longer 535 shutoff periods. In this study a "warm" startup was considered when the engine was shutoff 536 for only some minutes with the TPS system in standby operation.
- 537 Oxidation catalysts can help reduce CO and formaldehyde emissions during startup and shut 538 down but have little influence on CH4 emissions. The older the oxidation catalyst, poorer is 539 its performance. NO_X emissions are generally high during startup. If a NO_X control is installed, then its concentrations are decreased after startup. The shutdown process does not 540influence NO_X emissions significantly. If a stoichiometric operated engine is considered, a 541 542 three-way catalyst can not only reduce CO, CH_4 or formaldehyde emissions during 543 stationary operation, but also that of NO_X. However, such a catalyst performs poorly when 544 small amounts of residual oxygen are present in the exhaust-gas, e.g. during startup and 545shutdown due to the sensitive nature of the lambda control. The emissions were generally 546 found to be lower or equal for a three-way catalyst in comparison to an oxidation catalyst.
- 547

548 4. Conclusions

549 Assuming that a biogas CHP system is used as a new supply-demand adjustment resource 550 for mitigating output fluctuations in variable renewable energy, it can contribute to a 551 long-term adjustment power equivalent, such as SCR or TCR in Germany. The current state 552 of the art is suitable for SCR with start-up and shut-off times of under 5 minutes. In the 553 current Japanese power market, responsiveness and flexibility of biogas CHP systems are not 554 essential, but in the future, the requirements for adjustment capability will be categorized 555 based on responsiveness [37]. However, to operate efficiently, it is necessary for the biogas CHP system to equip a biogas bag with the capacity to supply power to restore the control 556 reserve. Many biogas plants in Japan do not have adequate gas-bag capacity due to the 557 558 FIT-specification power production strategy, so an upgrade is essential to provide adjustment capacity. Since the emission of CH4, formaldehyde, and SO2 increases when a 559 CHP system starts and stops, gas-bag capacity should be designed to reduce output 560 561 fluctuations. Furthermore, as the use of RE power sources continues to expand in the future, 562 there is a concern that the supply-demand adjustment capability of the entire power source 563 system will be insufficient. To improve the operability of the biogas CHP system, other 564 strategies must be developed, such as increasing the response speed of output changes and

565 lowering the minimum output to increase the range of output changes.

566 It is important to clarify the effects of differences in operational methods (normal/flexible 567 operation) through measurement and analysis of operational data of biogas CHP systems 568 that are connected to the grid for commercial operation. The Global Warming Potential 569 (GWP) of flexible operation should be investigated further due to an increase of CH_4 and 570 formaldehyde during startup and shutdown.

- In the case of the German energy system, where frequency control by biogas CHP systems is already common practice, the potential for providing further ancillary services need to be investigated. These services include voltage control, provision of reactive power, congestion management, and black start capabilities. The need for such services is expected to increase as the share of renewable energy in the grid rises. Biogas presents an ideal renewable energy resource in this regard as it can easily be stored in large quantities—unlike other resources such as solar and wind energy.
- 578
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- 586
- 587 Footnotes
- 588 1. Manufactured by Hioki Electric Co., Ltd.
- 589 2. Manufactured by Gasmet Technologies Oy.
- 5903. Manufactured by M&C TechGroup Germany GmbH.
- 591
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