Smart Energy and Sustainable Environment

2022, *25*(1): 15-22, https://doi.org/10.46390/j.smensuen.25122.444

MICRO-CHANNEL REACTOR FOR LOW LEVEL H_2 OXIDATION WITHIN AN O_2 STREAM: PRELIMINARY TESTS IN VIEW OF INTEGRATION IN A H_2 GENERATOR WITHIN CECE PROCESS

Alina Niculescu[*], George Ana, George Bulubasa, Iuliana Stefan, Ciprian Bucur, Anisia Bornea

National Research and Development Institute for Cryogenics and Isotopic Technologies – ICSI Rm. Valcea, 4 Uzinei Street, P.O. Box Râureni 7, 240050, Rm. Valcea, Romania

Abstract:

CECE (Combined Electrolysis Catalytic Exchange) process is the candidate for low level tritiated water detritiation within any application. The process consists mainly of a H_2 generator, a LPCE (Liquid Phase Catalytic Exchange) column and an O_2 striping column. During operation, tritium is being accumulated within the H_2 generator in the form of tritiated water and the effluent streams (hydrogen and oxygen) show in time an increase concentration in tritium in the form of both tritiated water vapors and gas, which needs to be recovered.

The tritium in the H_2 stream is recovered in a LPCE column, while the tritium in the O_2 stream is recovered in a striping column. In view of lowering the load onto the striping column and to recover the tritium in gas form, a micro-channel reactor is proposed to be included for H_2 oxidation followed by vapor recovery.

The design of the equipment together with the preliminary results of the tests done with H_2 and air will be shown, prior to integration in the CECE process.

Article info:

Received 24 December 2021
Received in revised form
15 February 2022
Accepted 05 March 2022
Available online 04 April 2022

OBSERVATION: The article was presented to the 23rd International Conference "New Cryogenic and Isotope Technologies for Energy and Environment" *EnergEn* 2021, Băile Govora, Romania, October 26-29, 2021.

Keywords*:*
CECE process, micro-channel reactor, LPCE column, mass spectrometry

The influence of How to cite: Niculescu, A., Ana, G., Bulubasa, G., Stefan, I., Bucur, C., Bornea, A. (2022). Micro-Channel Reactor for Low Level H_2 Oxidation within an O_2 Stream: Preliminary Tests in View of Integration in a H_2 Generator within CECE Process. *Smart Energy and Sustainable Environment*, *25*(1), 15-22, 2022, https://doi.org/10.46390/j.smensuen.25122.444.

1. INTRODUCTION

The micro-channel reactor is a small-scale device which becomes the topic of research world due to the many advantages they offer and has an important role in the implementation of the principle of process intensification in the chemical and process

[*]*Corresponding author: Alina Niculescu, E-mail: alina.niculescu@icsi.ro*

industries (Holladay & Wang, 2015; Tonkovich, et al., 2004; Klenov, et al., 2015; Hermes, et al., 2019). The micro-channel reactor is formed of multiple parallel micro-channels, ranging from submillimetre to millimetres, disposed within the reactor, imbued with platinum micro crystals which operate as a catalyst of the reaction of the conversion of hydrogen and oxygen into water.

The Pt based micro-channel catalytic reactor is foreseen to be used in deuterium - tritium pilot plant Tritium Removal System (TRS) from ICSI (Ana, et al., 2019). Also, the equipment represents a very good solution for burning of hydrogen from the oxygen stream coming from the electrolyser within the CECE process under development at ICSI. It is known that CECE process is appropriate for detritiation of low level tritiated water. This process uses a hydrogen generator, a LPCE column and an oxygen striping column. Both gas streams that exit the electrolyser contain tritium, the tritium from the hydrogen stream is recovered in a LPCE column, while the tritium in the oxygen stream is recovered in a striping column. In view of lowering the load onto the striping column and to recover the tritium, a micro-channel reactor is proposed to be included for hydrogen oxidation followed by vapor recovery. The advantages of using the micro-channel reactor are: small thermal inertia, inherent safety, well-controllable reaction conditions, small overall dimensions and the high rate of mass and heat transfer. These advantages allow a very direct control of the reaction temperature, present a very small reactor volume and make them ideal for applications where space requirements are critical (Veser, 2001).

The aim of this paper is to present the design of the micro-channel reactor together with the preliminary results of the tests that were performed for different hydrogen in air concentrations, prior to integration in the CECE process, in order to determine the optimal operation parameters.

The proposed design is used for tritium recovery from streams containing low amounts of tritium, but higher than the limit that can be released into the atmosphere, below 0.5% vol. of tritiated gas.

2. EXPERIMENTAL

2.1. The micro-channel reactor design

The equipment consists of four stainless steel plates with micro-channels drilled on two sides for the inner ones and on one side for the exterior ones (figure 1). On opposite sides, nozzles have been foreseen for gas inlet and outlet. The overall dimensions are 250 mm in length, 70 mm width and 48 mm thick.

Figure 2 shows the actual image of the micro-channel reactor integrated in the experimental setup.

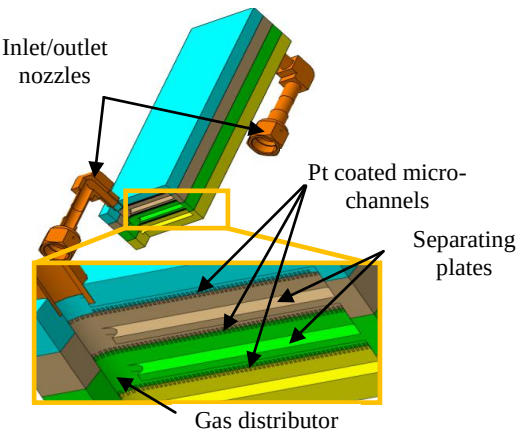

Figure 1. Micro-channel reactor – 3D model

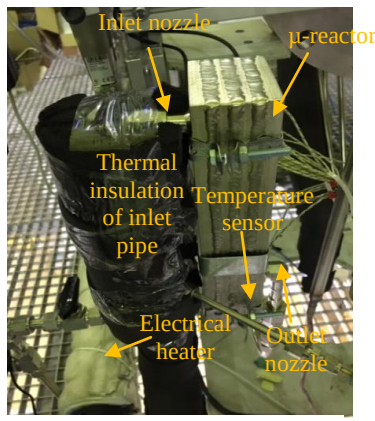

Figure 2. µReactor placed in the experimental setup

The channels, with sections 500 x 1000 µm, have been formed by electrical discharge machining (wire erosion) having a roughness of 0.8 µm. Platinum has been uniformly deposited onto the channels surfaces by thermal vacuum method (VAT) in a layer of 1 µm. The geometrical characteristics are summarized in Table 1.

Table 1. Characteristics of the µ-channels

Channel width	500 µm
Channel height	1 m
Channel length	210 mm
Number of channels	171
Areas with channels	6

For the distribution of the gas inside towards all channels, the equipment shows an inner distributor of gas and separation plates which divides the flow towards the reaction areas. The equipment is required to oxidize up to up to 0.5 percent tritiated hydrogen by volume in 1 std m^3/h oxygen.

2.2. Experimental setup

In view of testing the equipment and commissioning it, the micro-channel reactor has been integrated into an experimental setup as shown in figure 3. The setup has been equipped with instruments for measuring and controlling of flows (FC-M1, FC-M2, FC-M3), the temperature of the inlet gases and also pressures have been monitored along the gas stream (TI 1). Two additional temperature sensors have been placed on the equipment at the inlet (TI 2) and outlet (TI 3) of gases to monitor the shell temperature. Each of the gas lines is provided with valves (V1, V2, V3, V5, V6, V8, V9) and one way valves (NRV-1 and NRV-2).

Prior to any hydrogen tests, the entire system has been checked for leaks with helium. Additionally, to avoid electrostatic charges built-up due to fluid flow friction to the metal walls of the pipework followed by electrostatic discharges which can represent an effective ignition source, the stand has been bonded by means of copper wires and grounded, providing a pathway to drain the charges; all connections are secured against self-loosening, assuring an effective earthing and bonding as EN60079-14 requires.

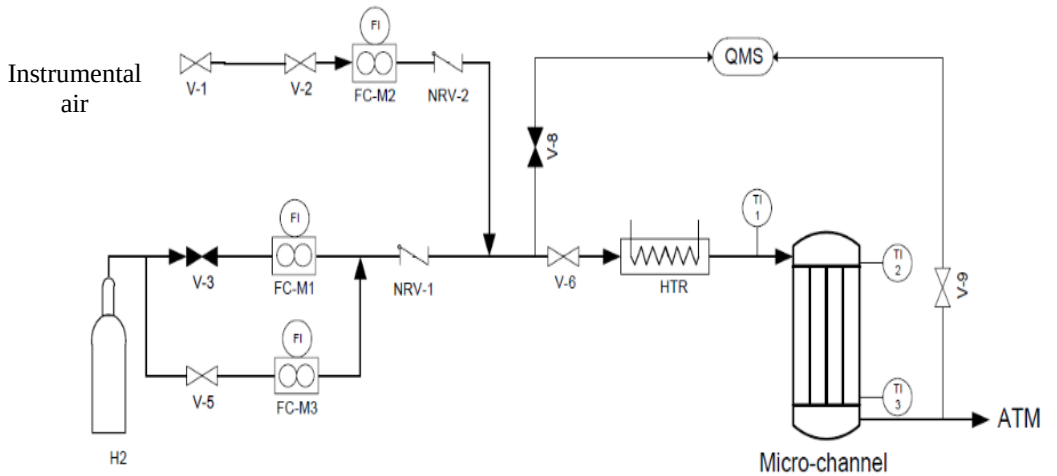

Figure 3. Experimental setup

In order to help initiate the hydrogen/oxygen reaction a heater (HTR) was mounted before the entering in the micro-channel reactor for preheating the feed mixture. The amount of water produced was determined from the difference of flow rates of hydrogen at the inlet and output from the micro-channel reactor and the time of the run. The feed hydrogen flow rate is directly measured and the output hydrogen flow rate is calculated by multiplying the total output flow rate with the measured hydrogen concentration by using a quadrupole mass spectrometer.

3. RESULTS AND DISCUSSION

For experimentally testing of the micro-channel reactor we used different hydrogen in air concentrations in order to determine the induction period and if the heat production from the reaction could be controlled with different mixtures of air and hydrogen. The experiments consisted of three runs with different operating parameters. During each run the air flow rate was kept constant at 1Nm^3/h, while the hydrogen flow rate was variated, yielding different concentrations of hydrogen in air presented in table 2.

Table 2. Parameters of the experimental runs

Run	H_2 feed flow rate Nm^3/h	H_2 feed concentration [%]	Duration h	$H_2O_{calculated}$ g	$H_2O_{collected}$ g
1	0.06	5.66	4	66.72	64.21
2	0.10	9.09	4	33.99	29.78
3	0.10	9.09	3	33.00	28.45

The first experiments were performed using a hydrogen feed flow rate of 0.06 Nm^3/h without heating the gas before entering the micro-channel reactor. The ignition can be observed in the increase of temperatures at the inlet, respectively output of the gas stream. From figure 4 it can be observed that the hydrogen oxygen reaction is initiated at the beginning of the feeding with the gas mixture.

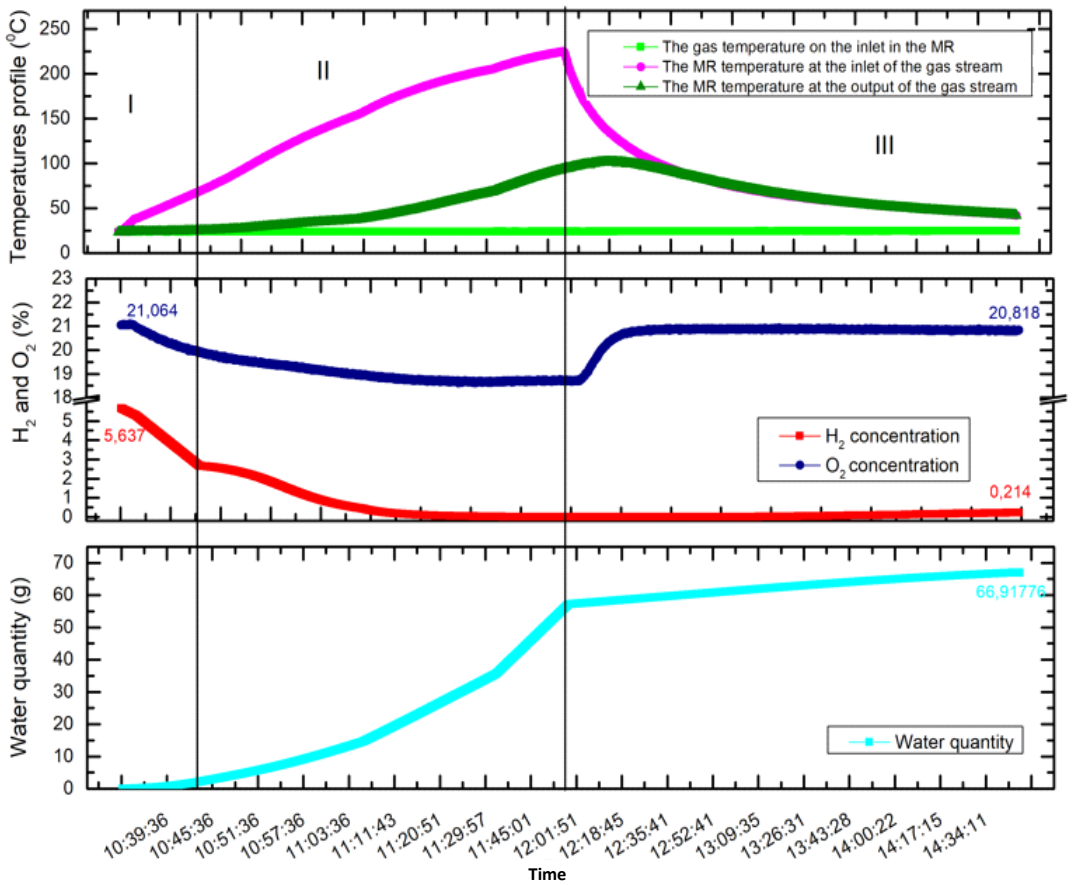

Figure 4. Experiment 1: The temperatures profile of the micro-channel reactor at the inlet and exit gas stream, respectively the calculated water quantity

After about two hours, the hydrogen flow rate was decreased to 0.005 Nm^3/h to avoid overheating of the micro-channel reactor, the temperature in the inlet zone being 225°C. The oxidation reaction continued until the temperature in the inlet zone decreased close to ambient temperature. The water produced during this run was collected, resulting a quantity of 64.21 g, in very good concordance with the calculated amount – 66.72g.

The second and third experiments had the same operating parameters: hydrogen feed flow rate 0.1 Nm^3/h and the gas inlet temperature of 20°C. Similar to the first experiment the hydrogen flow rate was kept constant until the temperature reached 250°C in the gas inlet zone of the micro-channel reactor. The only difference between these two runs consisted of the moment of the ignition that is shown in figures 5 and 6.

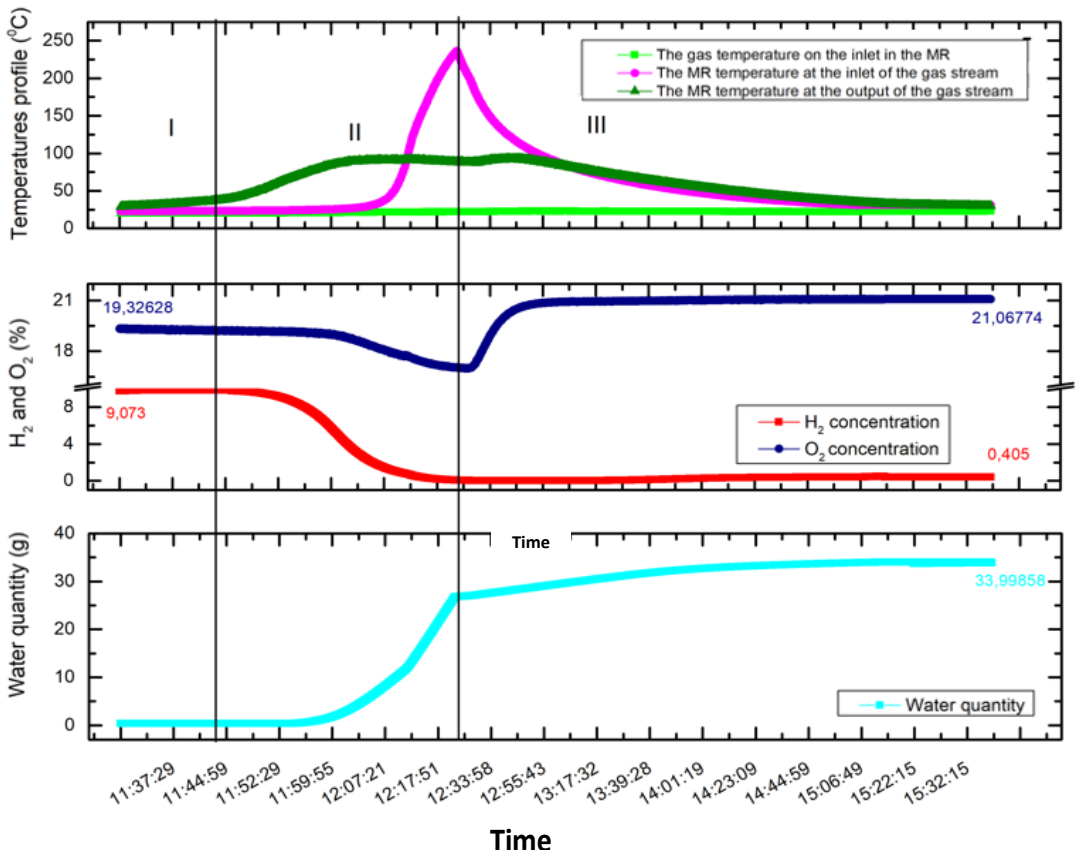

Figure 5. Experiment 2: The temperatures profile of the micro-channel reactor at the inlet and exit gas stream, respectively the calculated water quantity

From figure 6 it can be observed that the ignition took place after 30 minutes from the beginning of the run, confirmed by the increasing of the water amount. In the second zone of graph 6 a steep rise of the temperature at the inlet of the gas stream into the micro-channel reactor is shown. In the third zone the hydrogen flow rate was set at 0.005 Nm^3/h and the hydrogen oxygen reaction was sustained until the temperature in the inlet zone decreased close to ambient temperature. The calculated amount of resulted water was 33.00 g, while the sampled amount was 28.45g.

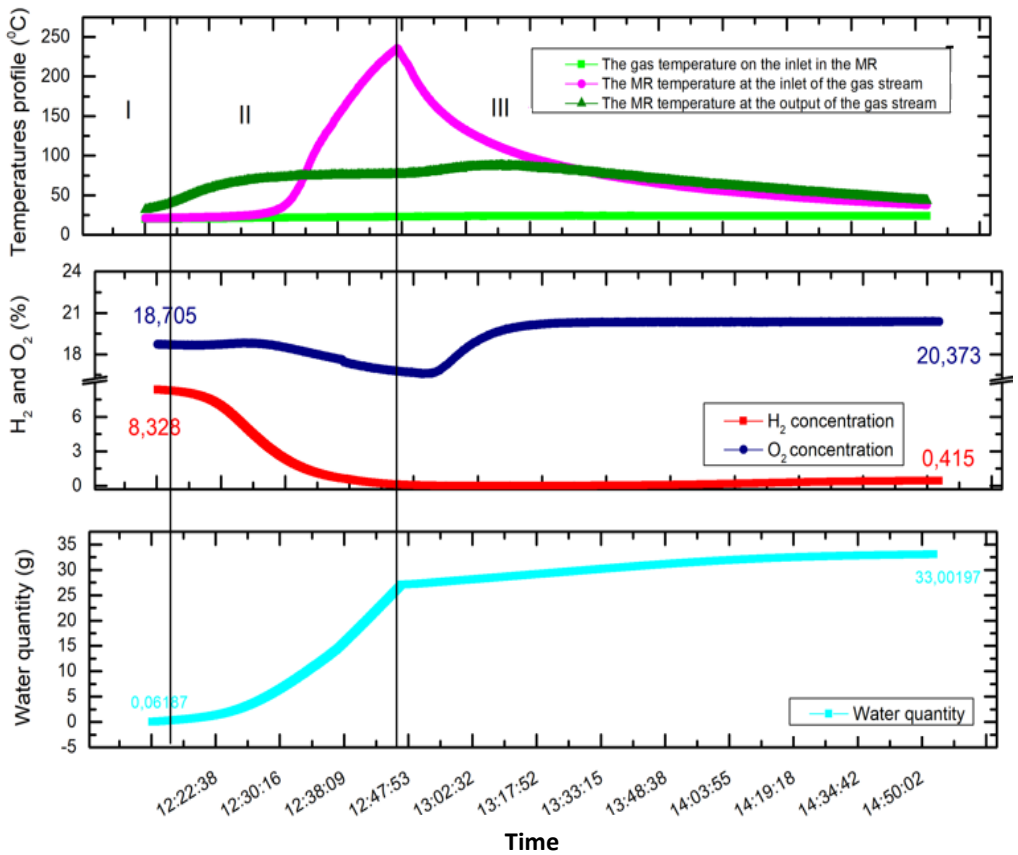

Figure 6. Experiment 3: The temperatures profile of the micro-channel reactor at the inlet and exit gas stream, respectively the calculated water quantity

4. CONCLUSIONS

A micro-channel reactor based on Pt-catalyser used for low level H_2 oxidation within an O_2 stream was shown to be a capable, safe option for conducting catalytic tests with mixtures of explosive gases. The use of small microchannels below the quenching distance for H_2 gas permitted dangerous mixtures of H_2 and O_2 to combine safely in the Pt/Al_2O_3 coated microchannels.

The main conclusion of the all three experiments is that the micro-channel reactor presents very good results for catalysed reaction between hydrogen and oxygen in the range of 5-10% hydrogen in air.

Regarding the repeatability, we noticed that the ignition of the reaction between hydrogen and oxygen started at different moments, probably because of the different ambient temperatures.

In order to obtain better results in the concentrations range of 0-0.5% hydrogen in air that represents the concentration domain of the exit gas from the CECE process, our future research with this system will be improved with mounting of an electrical blanket that will assure the heating of the micro-channel reactor at an adjustable temperature up to 300°C.

Acknowledgments: This work was performed in the framework of Core Program, conducted with the support of the Romanian Ministry of Research and Innovation, project PN 19 11 01 04.

Funding: This paper was prepared in the framework of Core Program, conducted with the support of the Romanian Ministry of Research and Innovation, project PN 19 11 01 04.

Author Contributions: George Ana and Ciprian Bucur concieved the experimental setup. Iuliana Stefan executed the electrical conections for the measurement equipments. Alina Niculescu and Gheorghe Bulubasa . conceived, planned and carried out the experiments, and, also, performed the analytic calculations. Alina Niculescu, George Ana, Gheorghe Bulubasa and Anisia Bornea contributed to the final version of the manuscript. All authors provided critical feedback and helped shape the research, analysis and manuscript.

Conflict of Interest Statement: There is no conflict of interest on this article.

REFERENCES

Ana, G., Pasca, G., Draghia, M., Niculescu, A., Bornea, A., & Zamfirache, M. (2019). Multi-criteria analysis of suitable technologies for the Tritium Removal System in Valcea D-T Pilot Plant. *Fusion Engineering and Design*, vol 146 (A): 1381-1384, doi.org/10.1016/j.fusengdes.2019.02.087.

Hermes, M., Mundhwa, M., Farhad, S., & Hamdullahpur, F. (2019). Catalyst layer design and arrangement to improve the performance of a microchannel methanol steam reforme. *Energy Convers Manag*, vol. 180: 149-161, doi.org/10.1016/j.enconman.2018.10.094.

Holladay, J., & Wang, Y. (2015). A review of recent advances in numerical simulations of microscale fuel processor for hydrogen production. *Journal of Power Sources*, vol. 282: 602-621, doi.org/10.1016/j.jpowsour.2015.01.079.

Klenov, O., Makarshin, L., Gribovskiy, A., Andreev, D., & Parmon, V. (2015). CFD modeling of compact methanol reformer. *Chemical Engineering Journal*, 282: 91-100, doi.org/10.1016/j.cej.2015.04.006.

Tonkovich, A., Perry , S., Wang, Y., Qui, D., LaPlante, T., & Rogers, W. (2004). Microchannel process technology for compact methane steam reforming. *Chemical Engineering Science*, 59(22-23): 4819-4824, doi.org/10.1016/j.ces.2004.07.098.

Veser, G. (2001). Experimental and Theoretical Investigation of Hydrogen Oxidation in a High-Temperature Catalytic Microreactor. *Chemical Engineering Science*, 56(4): 1265-1273, DOI: 10.1016/S0009-2509(00)00348-1.