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MEASUREMENT OF THE CONCENTRATION OF RADIOLYTIC HYDROGEN IN THE CHAMBER OF THE PELLETIZED CRYOGENIC MODERATOR OF THE IBR-2 REACTOR USING GAS CHROMATOGRAPHY

M.V. Bulavin[1,2*], A. Yskakov[1], K.A. Mukhin[1]

[1]Joint Institute for Nuclear Research, Moscow region, Dubna, Russia
[2]Petersburg Nuclear Physics Institute, NRC "Kurchatov institute", Gatchina, Russia

Abstract. *The paper presents the results of measurements of the concentration of gaseous radiolytic hydrogen using gas chromatography, which is produced irradiating the mixture of aromatic hydrocarbons of mesitylene and m-xylene in the chamber of the pelletized cold moderator of the IBR-2 reactor during post-irrradiation heating. As it is shown, the hydrogen concentration in the operating mode of the moderator at 22K in the chamber and a reactor power of 1.6 MW does not exceed 0.13%. After post-irradiation heating, with variations of temperature from 20K to 293K, and zero reactor power, the maximum hydrogen concentration is 22.5%. A conclusion is drawn that such concentration of hydrogen in an inert atmosphere of helium cannot lead to the production of an explosive mixture in the moderator chamber in close vicinity to the IBR-2 reactor core.*

Key words: *Cold moderator, mesitylene, metaxylene, radiolytic hydrogen, gas chromatography, thermochemical detector, oxyhydrogen gas*

1. INTRODUCTION

A mixture of aromatic hydrocarbons of mesitylene ($C_6H_3(CH_3)_3$) and metaxylene ($C_6H_3(CH_3)_2$) (hereinafter – a mixture) [1]-[3] is used as the working substance of the pelletized cryogenic moderator of the IBR-2 reactor [4]-[10]. The mixture is loaded into the chamber in the form of frozen pellets ~ 3.6 mm in diameter, using a cold helium flow at a temperature of 80 K through the supply pipeline, and neutron-physical experiments on the extracted beams of the reactor are carried out at a temperature of ~ 22 K in the moderator chamber and operating pressure throughout the system is 780-800 Torr; in this case the gain of cold neutrons (energy 10^{-5} - 0.0025 eV, wavelength – 25 – 2 A°) from the surface of the moderator ranges up to 14 times [11]. Under such conditions, the pelletized moderator operates during one reactor cycle, which usually lasts 11 days, resulting in an absorbed dose of ionizing radiation of ~ 140-160 MGy. Such high radiation loading leads to significant changes in the chemical composition and properties of the working substance of the moderator, especially in the period of post-irraditional heating of the substance during its scheduled replacement, which can directly affect the service life of the moderator chamber.

The service life of the moderator chamber, in turn, depends on two important factors. The first is the factor of increase in the dynamic viscosity of the mixture after irradiation and heating, due to a change in its chemical composition. For the pelletized cold moderator, under the irradiation conditions mentioned above, the viscosity of the mixture of mesitylene and metaxylene, which is in an inert helium atmosphere, increases to ~ 23 mPa (the viscosity of a pure mixture is 1.15 mPa). At this viscosity value, the irradiated mixture can be quickly and completely drained from the moderator chamber through a special drainage system [12]. At higher absorbed doses of ionizing radiation, an unpredictable stochastic increase in the viscosity of the mixture is possible, right up to production of solid resinous mass and soot, which cannot be removed from the moderator chamber.

The second factor is the rate of radiolytic hydrogen production and its concentration, in the moderator chamber during irradiation and after post-irradiation heating of the working mixture at the end of the reactor operation cycle. It should be noted here that the production of high concentrations of hydrogen (up to 90%) and a rapid increase of pressure in the chamber, as, for example, in the case of a cold solid methane moderator [13]-[15], will not be observed when the mixture based on mesitylene is heated, due to its good radiation resistance [16]-[17]. Also, it is necessary to accurately determine the maximum concentration of radiolytic hydrogen in mesitylene and m-xylene, primarily due to issues of burn up and explosion safety. This concerns the probability of producing an oxyhydrogen gas (4.1 -74.2 vol. % of hydrogen in air) in the cold moderator chamber and piping systems, in particular, in the pipeline for discharging hydrogen into the environment.

Therefore, the main objective of this research is measuring the maximum concentration of radiolytic hydrogen in the pelletized cold moderator chamber

* bulavin85@inbox.ru

and helium exhaust/supply pipelines after post-irradiation heating room at temperature.

During the experimental study, it was necessary to perform the following tasks:

1) use gas chromatography to determine the dependence of the concentration of radiolytic hydrogen produced in the chamber on the radiation dose during full-time operation of the pelletized moderator at a temperature of 22K;
2) determine the dependence of the concentration of radiolytic hydrogen on temperature (within the range 20K to 293K) during post-irradiation heating of the mixture;
3) use gas chromatography technique to measure the maximum concentration of radiolytic hydrogen produced in the chamber and helium supply/exhaust systems after irradiation and post-irradiation heating of the mixture to room temperature;
4) give recommendations on the issue of possible production of oxyhydrogen gas in the pipeline system for removing helium from the cryogenic moderator chamber and its explosion hazard.

2. Experimental setup and results

A schematic of the experimental setup for measuring the concentration of radiolytic hydrogen in the cold moderator chamber at T = 22K is shown in Fig. 1.

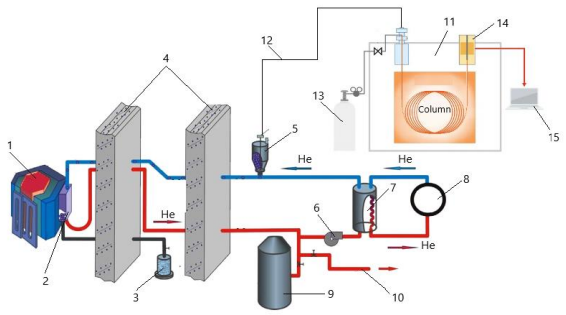
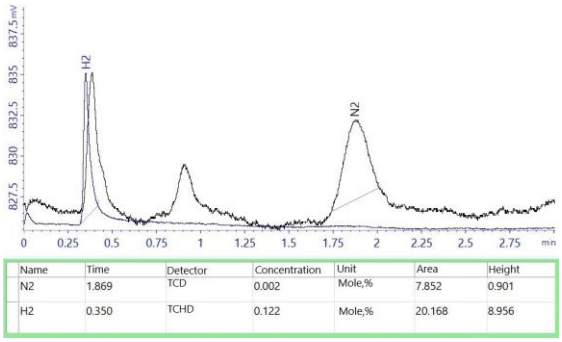


Figure 1. Schematic of the experimental arrangement to determine radiolytic hydrogen : 1 – IBR 2 reactor core, 2 – chamber with frozen pellets, 3 –used mesityline receiver, 4 – biologic shield, 5- device for dosing frozen balls into the helium upply line to the chamber, 6 –gas blower, 7 – heat exchanger, 8 –cryogenic helium plant, 9 – gas holder, 10- pipeline for removing hydrogen into the environment, 11 – Gas Chromatography Equipment (GC hereinafter), 12 – copper tube for transfer gas mixture to Gas Chromatography, 13 – carrier gas cylinder (helium A grade), 14- thermochemical detector (TCHD) 15 – personal computer

The measurement of hydrogen concentration in helium was carried out during the operation cycles of the IBR-2 reactor with a cold moderator. The cycle duration was about 11 days, and the measurement frequency was once a day. After loading the chamber (2) with frozen pellets, the temperature in the chamber decreased to 22 K, and the reactor (1) was put into operation at a nominal power of 1.6–1.8 MW. Measurements were carried out using calibrated "Gasochrome 2000 chromatograph" by Chromatek, specially equipped with a thermochemical detector, with a hydrogen concentration detection range between 0.1 and 25%.GC was calibrated by calibration gas mixture with different percentage of hydrogen and nitrogen. The principle of operation of the detector is based on the shift in the thermal effect during the catalytic burning of the sample. Air is conducted to the detector, to ensure burning of the sample on the catalyst surface when determining low hydrogen concentrations in inert gases.

The selection of gas mixture circulating in the cold moderator system, using a gas blower (6), from the chamber (2) to the heat exchanger (7), was carried out from the dosing device (5), by copper tube (12) to the chromatograph (11 and the thermochemical detector (14). The gas holder (9) is always closed during measurements. The analysis of each sample was carried out for 5 minutes with visualization of measurements in the Chromatek-Analytic program, where peaks with the concentration of hydrogen and nitrogen in helium were displayed (Fig. 2). The numerical value of the measured hydrogen concentration "H2" in percent is also indicated in the Table below the Figure 2. In total, about 40 measurements were carried out.



Name	Time	Detector	Concentration	Unit	Area	Height
N2	1.869	TCD	0.002	Mole,%	7.852	0.901
H2	0.350	TCHD	0.122	Mole,%	20.168	8.956

Figure 2. Chromatogram obtained by measuring the concentration of radiolytic hydrogen using gas chromatograph and the Chromatek-Analytic program during one of the operating cycles of the IBR-2 reactor. The hydrogen concentration (H2 ~ 0.12%) is shown in the table below the graph.

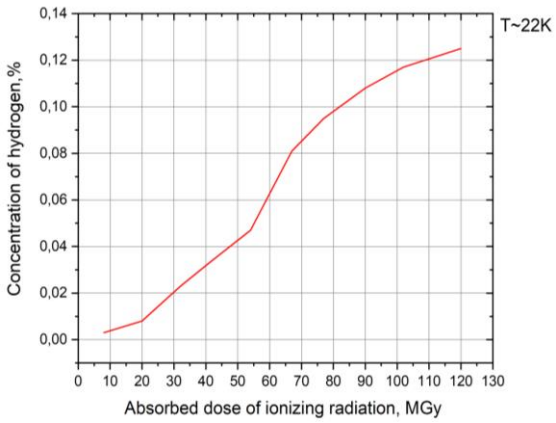


Figure 3. Formation of radiolytic hydrogen in the cold moderator chamber depending on the absorbed dose.

Figure 3 shows the change in the hydrogen concentration as a function of the irradiation time and absorbed dose of the mixture of mesitylene and m-

xylene in the moderator chamber at T = 22K. As we can see from Figure, by the end of the reactor operation cycle and at a power of 1.6 MW (~ 140 MGy), the concentration of radiolytic hydrogen in helium does not exceed 0.123%. This is due to the fact that, at a temperature below the critical value of 33 K, hydrogen is in a condensed state in the moderator matter (i.e., inside frozen pellets), and it is essentially not exhausted [18]. Such low, practically near-zero, concentrations of radiolytic hydrogen in an inert helium atmosphere reduce the possibility of producing oxyhydrogen gas and, accordingly, of ignition or explosion in the chamber, and helium supply/exhaust pipelines of the cold moderator during its operation at a nominal rating power of the reactor.

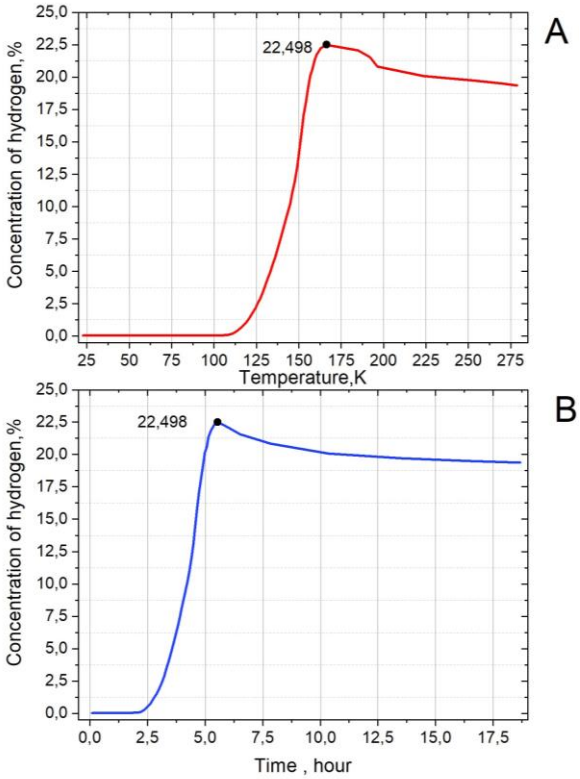
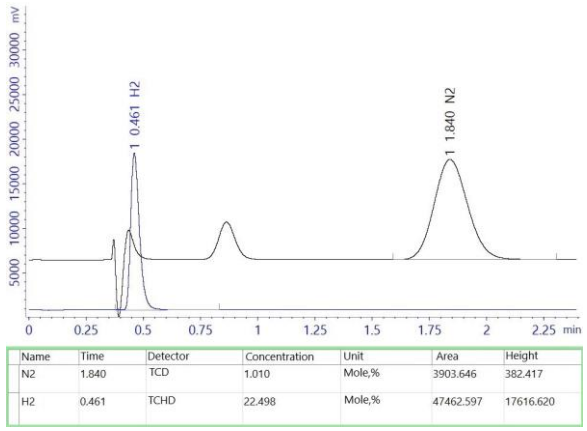


Figure 4. The formation of radiolytic hydrogen during the heating of the system as a function of the temperature (A) and time (B).

The other situation when it is necessary to pay special attention to the production of radiolytic hydrogen in cold moderator systems occurs during the heating of the mixture from 22K to 293K, after irradiation, in order to replace it with a "fresh", unirradiated one. It should be noted that this process occurs during the shutdown of the reactor, when its power is 0 and lasts about 19 hours. During this period, about 240 chromatographic measurements with open gas holder were carried out and the dynamics of the production of radiolytic hydrogen in the moderator chamber was determined (Fig. 4). It can be seen from the Figure 4 that the process of hydrogen exhaust begins at a temperature of 110K and continues until a maximum concentration of 22.5 % is reached (Fig. 5). Above 165K, the increase stops and the hydrogen concentration decreases to about 20%, because there is no fresh production of hydrogen and gas being removed from the system to the GC. This suggests that the production of radiolytic hydrogen in the moderator chamber was determined (Fig. 4).



Name	Time	Detector	Concentration	Unit	Area	Height
N2	1.840	TCD	1.010	Mole,%	3903.646	382.417
H2	0.461	TCHD	22.498	Mole,%	47462.597	17616.620

Figure 5. Chromatogram obtained at the maximum concentration of 22.498% radiolytic hydrogen during warming up of the system to room temperature. The absolute measurement error does not exceed 1% of the final concentration value.

3. Conclusion

The experimentally determined concentrations of radiolytic hydrogen under inert helium cannot lead to the production of oxyhydrogen gas in the moderator chamber, both during its operation at reactor power at a temperature of 22K, and during post-irradiation heating with the reactor out of operation.

In the current operation of the cryogenic moderator, the helium-hydrogen mixture is exhausted after heating through the pipeline for removing helium from the chamber into the environment, while oxyhydrogen gas can product. In order to reduce the possibility of its ignition, it is necessary to ensure the absence of ignition sources near it.

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References

1. S. Kulikov, A. Belyakov, M. Bulavin et al., "Current status of advanced pelletized cold moderators development for IBR-2M research reactor," *Physics of Particles and Nuclei Letters*, vol. 10, no. 2, pp. 230-235, 2013. https://doi.org/10.1134/S154747711302009X
2. E. Shabalin, S. Kulikov, M. Bulavin et al., "The world's first pelletized cold neutron moderator began its operation," *Neutron News*, vol. 24, no. 3, p. 27, 2013. https://doi.org/10.1080/10448632.2013.804363
3. V. Ananiev, A. Beliakov, M. Bulavin et al., "The world's first pelletized cold neutron moderator at a neutron scattering facility," *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atom*, vol. 320, pp. 70-74, 2014. https://doi.org/10.1016/j.nimb.2013.12.006

4. L. Cher, "Organic compounds for cold moderators," *Proceedings of the 14th International Conference on Advanced Neutron Sources*, Starved Rock, Illinois, vol. 2, pp. 241-244, 1998.
   Retrieved from:
   https://www.osti.gov/servlets/purl/1164678
   Retrieved on: May 17, 2020
5. R.D. Taylor, J.E. Kilpatrick, "Entropy, heat capacity, and heats of transition of 1, 3, 5-trimethylbenzene," *The Journal of Chemical Physics,* vol. 23, no. 7, pp. 1232-1235, 1955
   https://doi.org/10.1063/1.1742247
6. T.L. Bauer, B.W. Wehring, K. Unlu, "Study of Neutron Focusing at the Texas Cold Neutron Source. Final Report", *Nuclear Engineering Teaching Laboratory the University of Texas at Austin*, Texas, USA, 1996.
   Retrieved from:
   https://inis.iaea.org/collection/NCLCollectionStore/_Public/28/074/28074813.pdf
   Retrieved on: June 3, 2020
7. B.W. Wehring, K. Unlu, "The University of Texas Cold Neutron Source," *Proceedings of the Second International Seminar on Advanced Pulsed Neutron Sources PANS-II,* Dubna, Russia, 1994.
   Retrieved from:
   https://inis.iaea.org/collection/NCLCollectionStore/_Public/26/041/26041197.pdf?r=1&r=1
   Retrieved on: June 3, 2020
8. D.D. Clark, C.G. Ouellet, J.S. Berg, "On the design of a cold neutron source," *Nuclear Science and Engineering,* vol. 110, pp. 445-454, 1992.
   https://doi.org/10.13182/NSE92-A23917
9. D.D. Clark, "The Cornell University cold neutron beam facility: design features," *Proceedings of the International Workshop on Cold Neutron Sources,* Los-Alamos, USA, pp. 559-563, 1990.
   Retrieved from:
   https://inis.iaea.org/collection/NCLCollectionStore/_Public/23/008/23008249.pdf
   Retrieved on: June 17, 2020
10. M. Leuschner, "LENS: A new pulsed neutron source for research and education," *Journal of Research of the National Institute of Standards and Technology*, vol. 110, pp. 153-155, 2005.
    https://doi.org/10.6028/jres.110.016
11. M. Bulavin, A. Beliakov, A. Verkhogliadov et al., "Gain factor of the pelletized cold neutron moderator at 22K", *Journal of Surface Investigation: X-ray, Synchrotron, Neutron Techniques*, vol. 14, no. 2, 2020.
    https://doi.org/10.1134/S1027451020030040
12. V. Ananiev, A. Beliakov, M. Bulavin et al., "Pelletized cold moderator of the IBR-2 reactor: current status and future development," *Journal of Physics: Conference Series 746* 012031, pp. 1-6, 2016.
    https://doi.org/10.1088/1742-6596/746/1/012031
13. E.P. Shabalin, S. Kulikov, E. Kulagin et al., "Solid Methane Cold Moderator for the IBR-2 Reactor" *Proceedings of the International Workshop on Cold Moderators for Pulsed Neutron Sources Argonne National Laboratory*, Argonne, USA, pp. 73-78, 1997.
    Retrieved from:
    https://www.osti.gov/servlets/purl/1163133/
    Retrieved on: June 15, 2020
14. O. Kirichek, C.R. Lawson, D.M. Jenkins, et al., "Solid methane in neutron radiation: Cryogenic moderators and cometary cryo volcanism," *Cryogenics,* vol. 88, pp. 101-105, 2017.
    https://doi.org/10.1016/j.cryogenics.2017.10.017
15. T.L. Scott, J.M. Carpenter, M.E. Miller, "The development of solid methane neutron moderators at the intense pulsed neutron source facility of Argonne National Laboratory" *Proceedings of the Argonne National Laboratory,* Argonne, USA, pp. 299-304, 1998.
    Retrieved from:
    https://inis.iaea.org/collection/NCLCollectionStore/_Public/46/109/46109622.pdf?r=1
    Retrieved on: June 20, 2020
16. E.P. Shabalin, S. Kulikov, E. Kulagin et al., "Radiation effects in cold moderator materials: Experimental study of accumulation and release of chemical energy," *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 215, pp. 181-186, 2004
    https://doi.org/10.1016/j.nimb.2003.08.026
17. E.P. Shabalin, S. Kulikov, E. Kulagin et al., "Experimental study of spontaneous release of accumulated energy in irradiated ices," *Radiation Physics and Chemistry,* vol. 67, pp. 315-319, 2003.
    https://doi.org/10.1016/S0969-806X(03)00059-8
18. A.A. Beljakov, E.P. Shabalin, E. Kulagin, et al., "First experience of cold moderator operation and solid methane irradiation at the IBR-2 pulsed reactor," *Proceedings of the 16th Meeting of the International Collaboration on Advanced Neutron Sources (ICANS-XII),* Abingdon, USA, vol. 2, pp. 144-155, 1993.
    Retrieved from:
    http://www.neutronresearch.com/parch/1993/01/199301041440.pdf
    Retrieved on: July 13, 2020