Smart Energy and Sustainable Environment

2022, *25*(1): 35-46, https://doi.org/10.46390/j.smensuen.25122.446

A NEW METHOD TO GENERATE VERY LOW GAS FLOWS FOR CALIBRATION OF HYDROGEN PERMEATION MEASUREMENT. PART 1: CONCEPT AND FEASIBILITY TESTING

Nicolae Bidică[*], Narcisa Ghimiș

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:

Generating of very low (and well-known) gas flows meets a wide variety of needs in industry and research (especially in vacuum science and technology), but also among consumers and society (e.g: calibrations of vacuum gauges and leak detectors, leak testing to verify the tightness of many industrial products, assurance of product reliability and performance, environmental and other safety issues, and so on). So far, many methods and devices have been developed for generating very- and ultra-low flows, generally with the goal of setting flow standards for metrology. However, most of them are very complex, so that in case of certain particular research applications, new or adapted methods and devices appear to be more suitable. Such particular case is the calibration of the measurements of hydrogen isotopes permeation fluxes, when very low flows of hydrogen and/or deuterium are injected directly into the permeation cell which is operated in very different modes. This paper presents the concept of a new method proposed to accomplish this purpose, its corresponding mathematical model of flow, as well as the experiments performed to test the feasibility of the method. Briefly, the concept is based on two leaks of constant conductance through which a so-called reference flow is generated, with two stages of pressure reduction, as well as on a variable leak valve through which well-controlled flows (and over a wide range) are generated to be injected into the permeation cell.

Article info:

Received 15 December 2021
Received in revised form
18 March 2022
Accepted 29 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*:*
very low flows of gases, leak, conductance, calibration, permeation, hydrogen

The influence of How to cite: Bidica, N., Ghimis, N., (2022). A New Method to Generate Very Low Gas Flows for Calibration of Hydrogen Permeation Measurement. Part 1: Concept and Feasibility Testing, *Smart Energy and Sustainable Environment, 25*(1), 35-46, 2022, https://doi.org/10.46390/j.smensuen.25122.446.

[*]*Corresponding author: Nicolae Bidica, E-mail: nicolae.bidica@icsi.ro*

1. INTRODUCTION

There are many applications where known small flows of gas are required, for example, in leak detection, calibration of leak elements, low flow metrology, vacuum measurements, or other fields such as gas permeation experiments, where the flow rate could be below 10^{-8} mol/s (~ 10^{-4} Pa·m^3/s), but often this means even several decades below. (Ehrlich & Basford, 1992; Tison, 1993) The simplest way to generate very low gas flows would be by using a single leak element (capillary type preferably), with suitable calibrated conductance, through which the gas flows with rates adjusted by varying its inlet pressure. (Tison, 1993) However, this has a main drawback when wide range of flows are needed: changing the inlet pressure by orders of magnitude will cause changing of the flow regimes, so the conductance of the leak element will need to be measured as a function of pressure, which is a difficult task in practice.

Over time, (Bergoglio et al., 1995; Wangkui et al., 1996; Hedlund & Pendrill, 2006; Hidalgo & de Segovia, 2008; Detian et al., 2011; Detian et al., 2013) but even recently, (Avdiaj & Šetina, 2017; Guo et al., 2019) a multitude of methods have been proposed and devices have been built, with increasing efforts to reduce the lower limit of flow-rates, but also to improve the measurement accuracy of these ultra-low flows. Thus, more and more complex and costly solutions have resulted.

However, most methods and devices have been developed to set flow standards for metrology, while in certain particular research applications, new or adapted methods and devices appear to be more suitable. Such particular case is when the injection of very low flows of hydrogen and/or deuterium directly into the permeation cell is the option for calibration of the measurements of hydrogen isotopes permeation fluxes, in permeation experiments performed in very different modes. (Bidica et al., 2020) An important aspect in this case is that, depending on how the experiment is conducted, very different conditions may occur in the permeation cell, as for example high vacuum - in the case of permeation to vacuum, or a wide range of pressures - in the case of permeation to another gas. Thus, the simplest way of using a single leak (of calibrated conductance) is not appropriate; besides the disadvantage due to change of the flow regimes as the inlet pressure is increased (which leads to pressure dependent conductance), also wide ranges of pressure (instead of high vacuum) at the outlet of the leak would make the task even more difficult. Therefore, a new method has been proposed to accomplish this purpose, and this work presents the concept of this method (including the corresponding mathematical model used to characterize the flow), but also the results of the experiments performed to test the feasibility of the method.

2. THE CONCEPT OF THE METHOD

The concept is based on two leak elements of constant conductance through which a so-called reference flow is initially established, as well as on a variable leak valve through which well-controlled flows (and over a wide range) are eventually generated in order to be injected into the permeation cell. In figure 1 it is presented the flow diagram of the proposed set-up for generating very low flows. Being guided by this diagram, the understanding of the method concept should be easy.

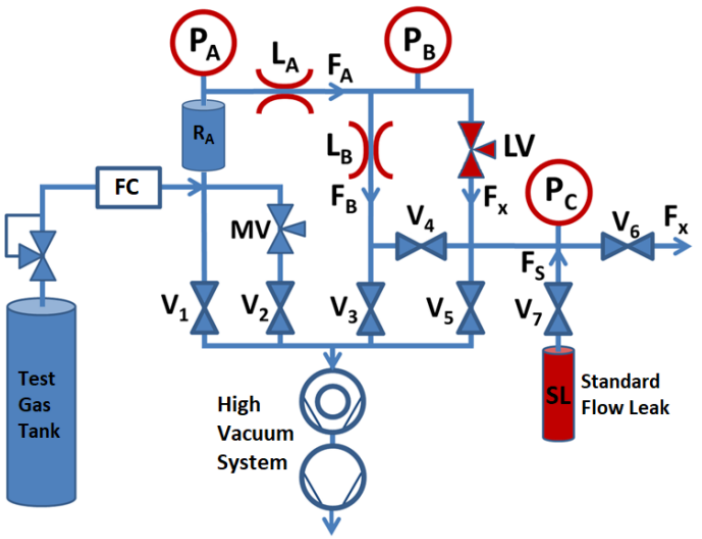

Figure 1. The flow diagram of the proposed set-up for generating very low flows

The key elements, marked by red color in fig. 1, are:

– (L_A) – first leak element of constant conductance C_A;

– (L_B) – second leak element of constant conductance C_B;

– (LV) – variable leak valve for generating very low flows (to be injected into the permeation cell);

– (P_A) – first pressure gauge (of capacitance type);

– (P_B) – second pressure gauge (of capacitance type);

– (P_C) – third pressure gauge (of capacitance type);

– (SL) – standard flow leak (used to calibrate the generated flows).

It is also important to distinguish the following three key compartments (gas chambers):

– Compartment A – consisting of the reservoir (R_A) and its connection tubes with flow controller (FC), isolation valve (V_1), metering valve (MV), the first leak element (L_A), and the first pressure gauge (P_A);

– Compartment B – a pseudo-chamber of a very low volume consisting only of the tubes connecting the first leak element (L_A) with the second leak element (L_B), the variable leak valve (LV), and the second pressure gauge (P_B);

– Compartment C – another pseudo-chamber of low volume consisting only of the tubes connecting the variable leak valve (LV) with the third pressure gauge (P_C) and isolation valves (V_4), (V_5), (V_6), and (V_7).

The test gas is supplied to the compartment A by means of flow controller (FC), and the pressure level – which is measured by the gauge (P_A) – could be maintained constant by adjusting the metering valve (MV) so that the incoming flow from (FC) is balanced by the outgoing flow toward vacuum. Being driven by the constant pressure P_A, a small part of gas from compartment A will also flow through the first leak element (L_A) into compartment B, where the gas accumulates over time but also flows further through the second leak element (L_B) toward vacuum, as being driven by the momentary gas pressure P_B in the compartment B – which is measured by the gauge (P_B).

Note: During this procedure, only the isolation valves (V_2) and (V_3) are opened, while all the other isolation valves, and also the variable leak valve (LV), are completely closed.

As the pressure P_B increases, the gas flow-rate F_A, passing through the leak element (L_A), decreases, while the flow-rate F_B, passing through the leak element (L_B), increases. So, the gas is accumulating into compartment B until the gas pressure P_B reaches steady state. This could be modeled by the following material balance equation for the compartment B:

$$\frac{V_B}{RT}\frac{dP_B(t)}{dt} = F_A - F_B, \tag{1}$$

where: V_B is the volume of compartment B, T is the temperature of gas in compartment B, and R is the ideal gas constant.

According to theory of gas flow at very low pressure (vacuum) through capillary tubes, the two gas flow-rates F_A and F_B depend on the conductances of the two leak elements but also on the pressure drops across each leak, respectively: (O'Hanlon, 2003)

$$F_A = C_A \cdot (P_A - P_B), \tag{3}$$

$$F_B = C_B \cdot P_B. \tag{4}$$

Note: As the outlet end of the leak element (L_B) is connected to high vacuum system, in eq. (4) the pressure drop across (L_B) was approximated with P_B.

Introducing equations (3) and (4) in eq. (1), it results:

$$\frac{dP_B(t)}{dt} = \frac{RT}{V_B}[C_A \cdot P_A - (C_A + C_B) \cdot P_B(t)]. \tag{1'}$$

If the gas flows under molecular flow regime through both leak elements, according to aforementioned theory the two conductances C_A and C_B are independent of gas pressure. Equation (1) can be easily integrated in this case, and its solution has the following form:

$$P_B(t) = P_A \cdot \frac{C_A}{C_A + C_B}\left\{1 - exp\left[-(C_A + C_B) \cdot \frac{RT}{V_B} \cdot t\right]\right\}. \tag{5}$$

The steady state value of the gas pressure P_B, accumulated inside the compartment B, for a given pressure P_A, kept constant inside the compartment A, is:

$$P_B = P_A \cdot \frac{C_A}{C_A + C_B}. \tag{6}$$

This is a consequence of equal flow-rates through both leak elements, $F_A = F_B$, where they are defined in equations (3) and (4). For the next steps, this state of equilibrium will be seen as the "*reference state*"; consequently, the flow through both leak elements will be the "*reference flow*", F^{ref}, and also the gas pressures inside compartments A and B, P_A^{ref} and P_B^{ref}, respectively, which satisfy eq. (6).

Opening the isolation valve (V_4) and closing (V_3) will allow assessing the reference flow-rate, F^{ref}, by measuring the pressure rise in compartment C. Calibration of pressure rise measurement is made when, similarly, the pressure rise in compartment C is measured for the flow-rate supplied by the standard flow leak (SL), i.e. standard flow-rate, F_S.

Note: All the time the entire system has to be kept at constant temperature; this is an essential condition both for constant gas flow parameters (pressures, leak conductances, flow-rates) and for accuracy and reproducibility of pressure rise measurements.

Once the reference flow established, from this point on the goal is to generate the flow rates of interest to be introduced into the permeation cell. This is accomplished by slowly opening of the variable leak valve (LV), then leaving it open at a certain position. The gas will begin to flow through (LV) at a rate, denoted by F_x, which depends on the opening position of the variable leak valve (which can be characterized by a conductance C_x), but also on the gas pressure in compartment B. As a consequence, the pressure in compartment B (which is initialy at the reference value P_B^{ref}) starts to decrease. This could also be modelled by the material balance equation for the compartment B under new conditions:

$$\frac{V_B}{RT}\frac{dP_B(t)}{dt} = F_A - F_B - F_x. \tag{7}$$

Using the expressions of the flow rates through all the leak elements as functions of their conductances and pressure drops, equation (7) becomes:

$$\frac{dP_B(t)}{dt} = \frac{RT}{V_B}[C_A \cdot P_A - (C_A + C_B + C_x) \cdot P_B(t)]. \tag{7'}$$

Integration of this equation with the initial conditions given by the so-called "reference state" (i.e. $P_A = P_A^{ref}$, and $P_B(t_0) = P_B^{ref}$, where both reference pressures satisfy eq. (6)) will lead to the following solution:

$$P_B(t) = P_B^{ref} \cdot \left\{\frac{C_A + C_B}{C_A + C_B + C_x} + \frac{C_x}{C_A + C_B + C_x} \cdot exp\left[-(C_A + C_B + C_x) \cdot \frac{RT}{V_B} \cdot t\right]\right\}. \tag{8}$$

Then, the new steady state value of the gas pressure in the compartment B, that is reached due to the generated flow-rate F_x, will be:

$$P_{B(x)} = P_B^{ref} \cdot \frac{C_A + C_B}{C_A + C_B + C_x}. \tag{9}$$

Because the quantity of interest is not the conductance C_x, but the generated flow-rate F_x established at this new equilibrium, we can replaced C_x by the ratio $F_x/P_{B(x)}$ and the following relation can be obtained:

$$F_x = (C_A + C_B) \cdot \left(P_B^{ref} - P_{B(x)}\right). \tag{10}$$

Thus, the generated flow rate of interest, F_x, is proportional to the difference between two steady state levels of gas pressure measured by the same pressure gauge (P_B)

inside the compartment B. With the hypothesis of constant conductances C_A and C_B (i.e. molecular flow regime), this expression shows us that F_x can be linearly determined by the "*fall*" in the steady state level of pressure P_B, from the reference level, P_B^{ref}, down to a corresponding new level, $P_{B(x)}$. We can denote this "*fall*" in pressure level by $\delta_x P_B$, so that eq. (10) takes a new linear form:

$$F_x = (C_A + C_B) \cdot \delta_x P_B. \qquad (10')$$

Every value F_x of the generated flow-rate can also be assessed by pressure rise measurement in compartment C, if isolation valve (V_5) is closed (and also the other isolation valves, (V_4), (V_6) and (V_7) are kept closed), or it can be supplied to the permeation cell by opening the isolation valve (V_6).

The main goal of this method is to generate known flow rates of hydrogen and deuterium in the range 10^{-13}-10^{-8} mol/s which are necessary to be introduced into a permeation cell, used in experimental studies of hydrogen isotopes permeation, in order to calibrate the permeation flows. Sizing of the leak elements to the appropriate conductances and also choosing the right levels for pressures, in order to match the desired flow range, have to take into account equations of the model described above, especially eq. (10').

Note: Essentially, this method is only based on steady state conditions of flow, so solving time-dependent equations would not have been strictly necessary. However, we chose this more complex approach for some reasons. On one hand, the equations (5) and (8), expressing the time evolution of pressure P_B, could help us in future work to assess the system parameters (especially the volume of the compartment B in accordance to conductances of the two leak elements), and to optimize them, as regarding the time required to reach steady states. On the other hand, these equations will help us to verify the correctness of the molecular flow hypothesis adopted in the above theoretical model, by fitting the experimentally measured (momentary) values of pressure P_B, with the theoretical evolution given by the above equations.

3. EXPERIMENTAL TESTING OF THE FEASIBILITY OF THE METHOD

To test the feasibility of this method, a temporary set-up was assembled as in the flow diagram presented in figure 2, where the two leak elements of constant conductances were replaced by two variable leak valves, (LV_A) and (LV_B), because they provide the higher flexibility needed for the task of these tests. The variable leak valve used for generating the flow-rates *Fx* of interests was denoted as LV_x in fig. 2. All three variable leak valves are of the same type: all-metal gas dosing valve, manually actuated, suitable for UHV applications, maximum conductance with molecular flow of 0.05 l/s, controllable gas flow from $1x10^{-10}$ mbar·l/s to $5x10^{2}$ mbar·l/s, operating pressure $1x10^{-10}$ mbar to $1.5x10^{3}$ mbar.

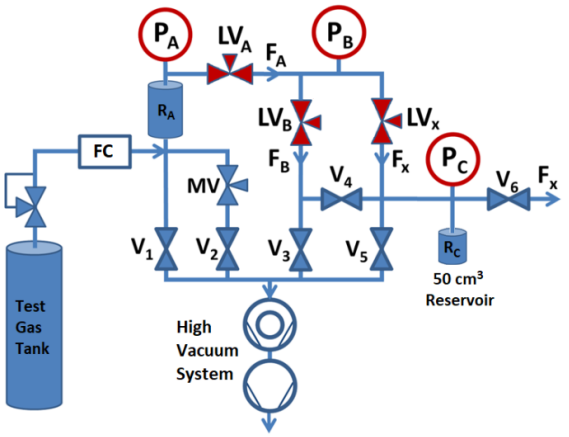

Figure 2. The flow diagram of the set-up used for feasibility testing experiments.

Another difference from the original configuration (fig. 1) is the lack of the standard leak device, which was not available at the time of these tests. However, just measuring the pressure rise in compartment C would have been sufficient for the purpose of these tests to get a relative comparison of flow-rates, so it would not have been strictly necessary to calibrate these flows. In spite of that, the flow controller (FC) used for feeding the test gas was also used to calibrate the pressure rise measurement in compartment C, thus giving a more complete figure on the results of these tests. This flow controller (which was factory calibrated for hydrogen gas) were capable to control flow-rates from 0.014 ml(n)/min (minimum controllable) up to 2 ml(n)/min (i.e. approximately $1x10^{-8}$ mol/s to $1.5x10^{-6}$ mol/s). But because even these quite low flow-rates were higher than the ones purposed to be tested, a reservoir of about 50 cm^3 capacity (R_C) had to be connected to the compartment C.

All the connection tubes, reservoirs and valves were wrapped around with electrical heating tapes and then thermally insulated so that to assure a stable temperature of the entire system (and thus, stable conditions of flow) during experimental testing.

As regarding the pressure measuring devices installed on the testing set-up, all three were vacuum gauges of capacitance type; the gauges (P_A) and (P_B) are identical and have a measuring range from 1 Pa to 10^4 Pa, while the gauge (P_C) has the range between 10^{-3} Pa and 10 Pa.

Before the experimental tests, entire system (i.e. reservoirs, valves and connecting tubes) was heated to 100^{o}C and degassed for about 40 hours under high vacuum, then it was cooled down to 35^{o}C and maintained at this temperature all along experimental testing.

The first step was to "calibrate" the pressure rise measurement in compartment C, by measuring the rate of pressure increase for certain flow-rates of gas provided by flow controller (FC). Thus, the test gas (hydrogen) was fed into compartments A and B, at certain flow-rate values set on (FC), and let it flow into compartment C and toward vacuum, by keeping leak valve (LV_A) fully open, and so the isolation valves (V_3) and (V_4) kept open, while (LV_B) was only slightly open. By restricting the flow through (LV_B), a much higher level of pressure can be realized in compartments A+B compared to compartment C, so that, when pressure in compartments A+B is steady, the measurement of the pressure rise is practically limited to the volume of compartment C only. Flow controller (FC) was set first to 0.015 ml(n)/min. ($1.12x10^{-8}$ mol/s). Leak valve (LV_B) was adjusted so that to create a steady state pressure of about 5000 Pa in compartments A+B; this value was chosen in the middle of the

measuring range of gauges (PA) and (PB) so that there is availability for both increases and decreases of this pressure level for the following procedural steps. That was in the very next steps, the flow-rate set on (FC) was increased to 0.020, then to 0.024, and then to 0.028 ml(n)/min., and the stationary pressure level in compartments A+B increased accordingly until it approached the maximum of the measuring range of the gauges. For each of the four values of the flow-rate set on (FC), and when the pressures P_A and P_B were at steady state, the isolation valve (V_3) was closed, hydrogen gas was accumulating in compartment C and its pressure increase was measured. The measured rates of pressure increase in compartment C, expressed in Pa/s, versus the corresponding flow-rates of hydrogen provided by (FC), expressed in mol/s, are presented in figure 3. The linear regression given by the four points (solid blue squares in fig. 3) was then used to convert every rate of pressure increase in compartment C, measured in units of Pa/s, into units of flow-rate, mol/s (like in case of the distinct point, the empty red circle in fig. 3, which represent a flow-rate rate measurement made in the next procedural step of these experimental tests).

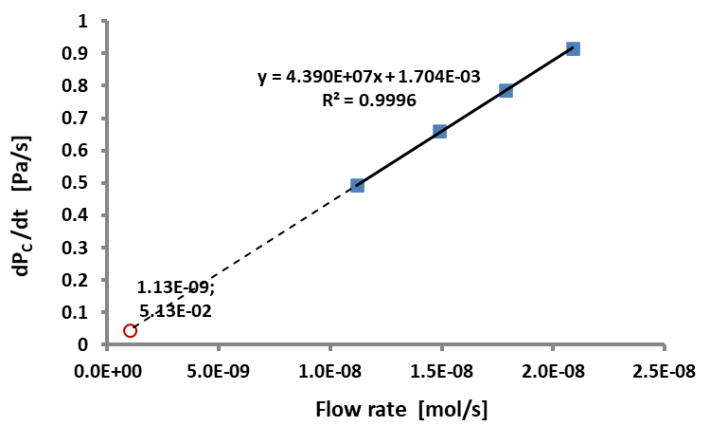

Figure 3. Calibration of the pressure rise measurement in compartment C.

As already mentioned above, the flow-rates purposed to be generated within these tests were much lower than the minimum flow-rate provided by flow controller. Thus, the next goal was to adjust both leak valves (LV_A) and (LV_B) to much lower conductance, so that to create much lower flow-rates of hydrogen gas through them for steady pressures of thousands of pascals in compartment B, and pressure drops across (LV_A) of 1000 Pa at most. To do that, the pressure of hydrogen gas in compartments A+B was decreased back to about 5000 Pa, then the leak valve (V_B) was gradually closed so that the flow-rate through it (assessed by measuring the pressure increase rate in compartment C) was about one order of magnitude lower than the minimum flow-rate provided by (FC). Secondly, the leak valve (V_A) was gradually closed until the pressure of hydrogen gas increased to 6000 Pa and remained stable at this level. During these adjustments of the two variable leak valves, the gas flow rate through them was also gradually decreasing, and thus the metering valve (MV) had also to be gradually opened so that to maintain the desired steady level of hydrogen pressure in compartment A. This procedure proved to be by far the most difficult and time consuming, many other fine adjustments being necessary until true steady state pressure levels in compartments A and B were obtained eventually, as well as a constant flow-rate, at one-tenth of minimum supplied by (FC), for hydrogen gas through the two leak valves. Finally, the following values have been obtained: P_A = 6000 Pa, P_B = 5500 Pa,

for a flow rate $F_A = F_B = 1.13\text{x}10^{-9}$ mol/s, which was calculated from the direct measurement of pressure increase in compartment C ($dP_C/dt = 5.13\text{x}10^{-2}$ Pa/s) and using the calibration curve presented in fig. 3 (also this measurement is represented by the point marked with empty red circle in fig. 3).

The openings of leak valves (LV_A) and (LV_B), which were set-out above, remained unchanged (fixed positions, so constant conductances), and the next experimental test was intended to verify the hypothesis of molecular flow of hydrogen gas through the two leaks, at these fixed conductances and for this pressure range. Thus, starting from the value of P_A = 6000 Pa, the pressure in compartment A was then gradually changed to lower stationary levels (by gradually opening of the metering valve (MV)), down to P_A = 1000 Pa. For each new steady level of P_A, the pressure in compartment B decreased until also new steady level of P_B was obtained, and for each new pair of steady pressures P_A and P_B the resulting flow-rate of hydrogen gas through both leaks (LV_A) and (LV_B) was assessed by measuring the rate of pressure increase in compartment C. Then, the obtained valued of the flow-rates were correlated with the corresponding pressure drops on both leaks, $P_A - P_B$ and P_B, respectively, in order to verify the theoretical linear relations given by equations (3) and (4) from above. These correlations are graphically presented in figures 4 (a) and (b), respectively.

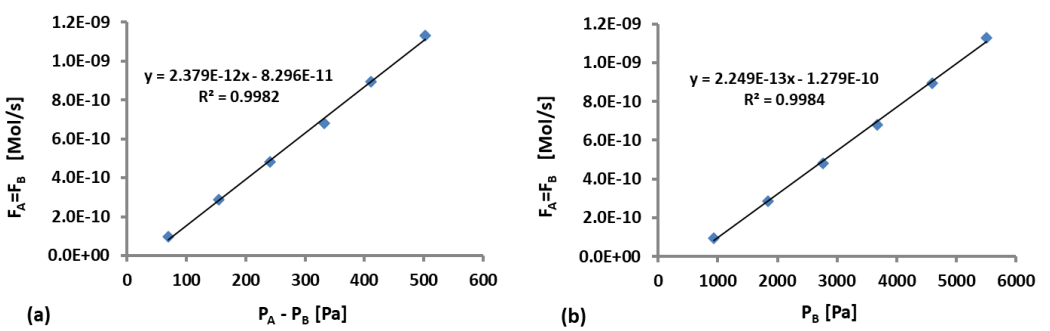

Figure 4. Experimental correlations between the flow-rates through both leaks and the corresponding pressure drops.

A good linearity was obtained for both leaks, which indicate that molecular flow regime could be well approximated with these conditions of flow. This could also be confirmed by the correlation between P_B and $P_A - P_B$, presented in figure 5, which is in a very good agreement with theoretical linear relation given by equation (6) from above.

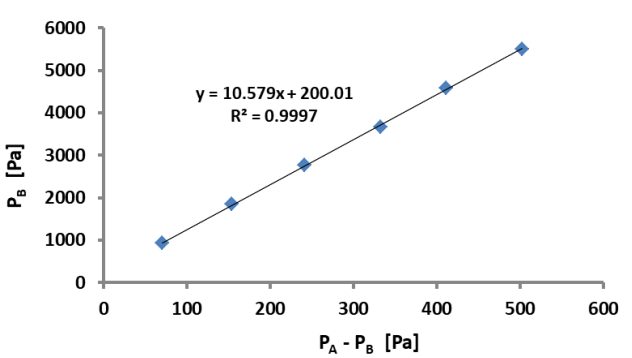

Figure 5. Experimental correlations between the pressure drops on the two leaks.

The slops of the linear regressions of the experimental data presented in figures 4 (a) and (b) determine the conductances of the two leaks, $C_A = 2.379 \times 10^{-12}$ (mol/s·Pa) and $C_B = 2.249 \times 10^{-13}$ (mol/s·Pa), respectively, and their ratio is $C_A/C_B \approx 10.578$, almost equal with the slope of the linear regression of data presented in fig. 5, which also determines the ratio C_A/C_B.

The two variable leak valves, (LV_A) and (LV_B), were kept fixed at the same openings (i.e. the same constant conducatnces) for the next experimental tests where the low flows of interests, F_x, are generated by gradually opening of the variable leak valve (LV_x). However, because small deviations from the linear behavior can be observed in fig. 4 for pressure P_A above 4000 Pa, these tests were performed by setting-up the hydrogen pressure in compartment A (i.e the "reference pressure" P_A^{ref}) at 3000 Pa. Consequently, the steady pressure established in compartment B (i.e the "reference pressure" P_B^{ref}) was 2756 Pa.

First opening of the leak valve (LV_x) was done so that the generated flow, F_x, would produce a „fall" of about 5 Pa in the pressure level P_B (from the reference level P_B^{ref} to the new steady level P_{Bx}, $\delta_x P_B = P_B^{ref} - P_{Bx}$). The experimentally obtained firs "fall", $\delta_x P_B$, was about 4 Pa. The corresponding flow rate F_x was assessed by measuring the rate of pressure increase in compartment C, and then converting it in mol/s units; the resulted value was 7.82×10^{-12} mol/s. The tests were continued in the same manner to produce higher flow-rates values F_x for higher $\delta_x P_B$ (i.e. lower steady levels in pressure P_B). All the obtained results are graphically presented in figure 6, where the generated flow-rates F_x are correlated to the corresponding falls $\delta_x P_B$ in pressure level P_B.

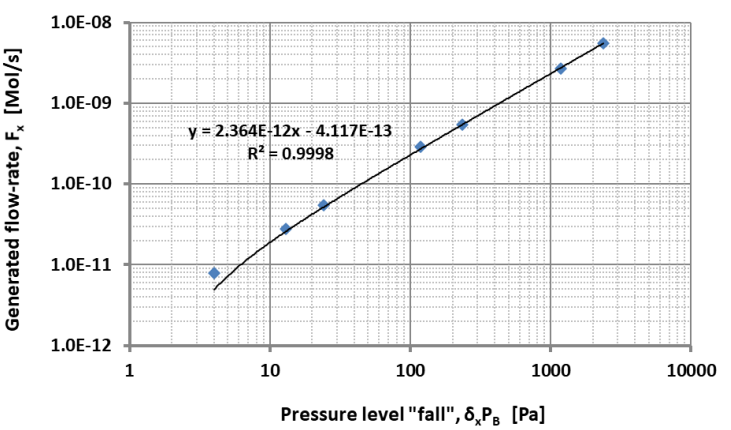

Figure 6. Experimental correlations between the generated hydrogen flows F_x and the corresponding "falls" $\delta_x P_B$ measured for the hydrogen pressure level in compartment B.

These results have shown a good linearity, in very well agreement with equation (10) from above. Also, the slope of the linear regression, 2.364×10^{-12} (mol/s·Pa), which represents the sum of the two constant conductances, $C_A + C_B$, is very close to the sum of their values individually determined above, $C_A = 2.379 \times 10^{-12}$ mol/s·Pa and $C_B = 2.249 \times 10^{-13}$ mol/s·Pa, within the tests performed at for a higher pressure range. The small difference could be explained by the fact that C_A increases slowly at higher pressures.

To further explore the validity of the above described model based on molecular flow hypothesis, but this time for non-steady conditions of flow described by eq. (5), a last experimental test was performed. Thus, the variable leak valve (LV_x) was fully opened in order to evacuate the compartment B, so that the initial condition $P_B(0) = 0$ Pa could be

well approximated. Then (LV_x) was completely closed as fast as possible so that the generated flow-rate F_x could be supposed zero. While the variable leak valves (LV_A) and (LV_B) were kept at the same openings as in previous tests (i.e. the same conductance values are supposed) and the pressure level in compartment A was maintained at the steady level P_A = 3000 Pa, the time evolution of pressure P_B where monitored at every second. The momentary measured values of P_B are presented in figure 7 (the points marked by empty blue squares) in comparison with the theoretically evolution $P_B(t)$ (the red line) which was calculated according to eq. (5) where the following input data were used: P_A = 3000 Pa, C_A = 2.379x10^{-12} mol/s·Pa, and C_B = 2.249x10^{-13} mol/s·Pa – as parameters, and $P_B(0)$ = 0 Pa – as initial condition. This comparison has shown a good agreement of the model prediction (eq. (5)) with experimental data, confirming once again the validity of the model with molecular flow hypothesis.

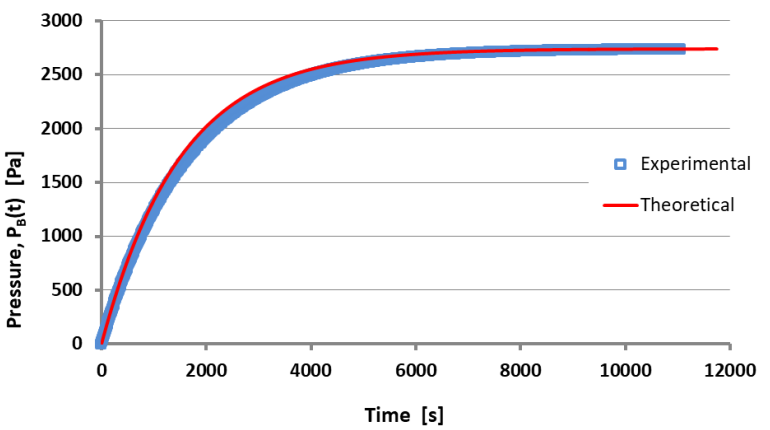

Figure 7. Comparison between time evolutions of P_B as given by experimental measurements (blue empty squares), and theoretical model (red line), respectively.

4. DISCUSSIONS AND CONCLUSIONS

A new method to generate very low gas flows has been proposed in this paper. Its main goal is to generate very low and known flow rates of hydrogen and deuterium in the range 10^{-13}-10^{-8} mol/s which are necessary to be introduced into a permeation cell, used in experimental studies of hydrogen isotopes permeation, in order to calibrate the permeation flows. However, this method can be also used in many other applications.

Briefly the method is based on two leak elements of constant conductance through which a so-called reference flow of gas is established between two steady levels of pressure (measured in two compartments separated by the first leak element), as well as on a variable leak valve (with its inlet connected to the second compartment) through which well-controlled flows (and over a wide range) can be generated. These generated flows can be very well determined by the changes in the steady level of pressure measured in the second compartment. A complete description of this mechanism, and also the associated mathematical model based on molecular flow regime, were presented in this paper.

The feasibility of the method, and also the theoretical model, were experimental tested in many ways (using hydrogen gas) on an experimental set-up where variable leak valves were used instead of the two fixed leak elements on which the concept of the method is based (this providing more flexibility for this kind of testing experiments).

The experimental results of the tests performed within this work are presented in the paper and demonstrate, on one hand, the validity of the theoretical model of flow (based on molecular flow regime), and, one de other hand, that this method can be very well used for generating very low and known flows, with the purpose (at least) of calibration of the permeation flows measurements. The range of hydrogen flow rates generated during the test presented here were approximately from 10^{-11} to 10^{-8} mol/s. However, this range could be extended down to 10^{-13} mol/s by an adequate dimensioning of the conductances of the leak elements and also by decreasing the pressure range, this being the goal of a future work.

Acknowledgments: This work has been carried out within the framework of Core Program, project no. PN-19.11.01.01, contracted with the Romanian Ministry of Research and Innovation.

Funding: This paper was prepared with founding support from Romanian Ministry of Research and Innovation.

Author Contributions: Nicolae Bidica: the method's conceptual, mathematical formalism and the design of the experimental set-up; Narcisa Ghimis: experimental tests and data handling.

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

REFERENCES

S. Avdiaj, J. Šetina, (2017) Flowmeter for very low gas flows of inert gases, *Measurement*, Vol. 111, pp. 384-388.

M. Bergoglio, A. Calcatelli, G. Rumiano, (1995) Gas flowrate measurements for leak calibration, *Vacuum*, Vol. 46, Issues 8-10, pp. 763-765.

N. Bidica, N. Sofilca, G. Popescu, B. Monea, C. Moraru, (2020) Experimental setup and pre-experiment issues for multi-component hydrogen isotopes permeation through metals in non-steady-state, *Fusion Engineering and Design*, Vol. 157, pp. 1-7

L. Detian, C. Yongjun, F. Yan, (2011) Extension of the range of a constant-conductance flowmeter by a flow divider, *Measurement Science and Technology*, Vol. 22, No. 12, pp. 1-5.

L. Detian, C. Yongjun, F. Yan, X. Zhenhua, Z. Lan, (2013)Very low gas flow measurements for UHV/XHV and leak calibration, *Metrologia*, Vol. 50, pp. 15-19.

C. D. Ehrlich and J. A. Basford, (1992) Recommended practices for the calibration and use of leaks, *Journal of Vacuum Science & Technology A*, Vol. 10, Issue 1, pp. 1-17.

E. Hedlund and L. R. Pendrill, (2006) Improved determination of the gas flow rate for UHV and leak metrology with laser refractometry, *Measurement Science and Technology*, Vol. 17, No. 10, pp. 2767-2772.

M. Guo, Z. Ren, Y. Cheng, W. Sun, Y. Li, L. Zhao, M. Dong, (2019) A new high precision, broad range gas micro-flow calibration apparatus, *Vacuum*, Vol. 164, , pp. 428-435.

J.M. Hidalgo and J.L. de Segovia, (2008) Leak calibration by comparison with reference standard leaks, Vacuum, Vol. 82, Issue 11, pp. 1151-1156.

John F. O'Hanlon, (2003) A User's Guide to Vacuum Technology, Third Edition, John Wiley & Sons, Inc., Hoboken, New Jersey.

S. A. Tison, (1993) Experimental data and theoretical modeling of gas flows through metal capillary leaks, *Vacuum*, Vol. 44, No. 11 /12, pp. 1171-1175.

L.Wangkui, Z. Dixin, L. Qiang, L. Shiliang, L. Detian, M. Yang, (1996) A gas flow standard apparatus, *Vacuum*, Vol. 47, Issues 6-8, pp. 519-522.