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Pulsed Neutron Activation Method**

Per Lindén

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Abstract

The objective of this work was to develop and study the feasibility of a flow-meter, based on the pulsed neutron activation method. It is a non-invasive method with good potential regarding accuracy. However, the ultimate accuracy has not been fully investigated before. If sufficient accuracy is obtained, provided a portable neutron source, the method is well suited for on-site calibration of existing flow-meters.

Two series of flow rate measurements have been performed and analysed. The first series was done under moderately accurate flow calibration conditions to get sufficient confidence in the method and to get indication of the obtainable accuracy. The results were encouraging and further measurements with high accuracy flow calibration were planned.

For high accuracy flow calibration, a dedicated loop was designed and built, and it was used with satisfactory performance. Two models have been used for analysis of recorded data; time weighing methods and a fit to Taylor diffusion theory. The results show that the accuracy in mean flow velocity obtained from used analysis models is in the range of 2-4% for Reynolds numbers greater than 10000. Data recorded from high calibration measurements will also be used for validation of future calculations.

Keywords: pulsed neutron activation, flow rate measurements, single phase flow, pipe flow, fluid velocity determination,

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This thesis is based on the following papers

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Water Flow Measurements with the Pulsed Neutron Activation Method

by

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For high accuracy flow calibration, a dedicated loop was designed and built and it was used with good performance. Two models have been used for analysis of recorded data; one time weighing method and one using a fit to Taylor diffusion theory. The results show that the accuracy in mean flow velocity obtained from the analysis models used is in the range of 2-4% for Reynolds numbers greater than 10000. The recorded data from these high calibration measurements will also be used for validation of future calculations.

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1. Introduction

1.1 General

Flow in pipes is a daily life phenomenon. In pipes, such as a human vein or an oil pipeline, the mass flow rate is often of interest. Depending on many parameters, e.g. pipe diameter or type of fluid, the method of determining the mass flow rate differs. All methods have their benefits and drawbacks and the ultimate method, i.e. a method usable for all flow cases, is not even likely to be invented. This work deals with a method of determining the mass flow rate for water in large pipes, e.g. the feedwater flow in a nuclear power plant, with a high accuracy potential.

Accurate flow measurements are needed in a large number of industrial applications, both for determining the mass transport and for calorimetric measurements of produced heat. The following example is from the nuclear power industry. In order to determine the reactor thermal power, the heat transfer from the reactor to the turbine system has to be known. Therefore, measurements of temperatures and mass flow rates in the turbine system are performed and the heat transfer is calculated with data from these measurements. One dominant parameter in these calculations is the feedwater mass flow rate and, hence, by increasing the accuracy of the feedwater flow it is possible to increase the accuracy of the reactor thermal power.

One important property of a fluid is the viscosity. If there was no viscosity, the velocity of a flowing fluid would be uniform across a pipe section. However, the presence of viscosity in a fluid introduces shearing between adjacent fluid particles that reduces the velocity to zero at the pipe wall. Thus there will be a non-uniform velocity profile in the pipe. For convenience, the Reynolds number is introduced which indicates the ratio of the inertial forces to the viscous forces in the fluid. For low Reynolds numbers the flow is laminar, and can be characterized by parallel layer motion. For laminar flow the velocity profile is parabolic and the mean velocity is proportional to the pressure drop. For high Reynolds numbers, the flow is turbulent and we need to resort to empirical formulae for losses and profiles. It has been found that the mean velocity is proportional to the square of the pressure drop. The turbulent velocity profile is flatter than the laminar one due to turbulent mixing. It becomes increasingly flatter for increasing Reynolds number until all particles, except close to the wall, are moving with the same velocity.

In most practical cases the flow is turbulent and measurements of flow properties are relying on empirical data. In the development of a flow-meter, a large number of measurements and tests must be done to give indications of flow-meter performance. High accuracy demands a detailed understanding of the flow and for the turbulent case, where the flow is characterized by randomness, the analysis of a measurement is a complex problem. Flow measurements are dependent on the theoretical models used. Hence the improvements of both measurements and models walk hand in hand and there is always need for development.

1.2 Existing Methods for Flow Determination

Flow-meters are generally divided into quantity and rate meters and then further subdivided by operating principle. This divides meters into those that measure discrete vol-

umes and those that sense the movement of the fluid. In practice, however, a rate meter is often used to determine the quantity, and sometimes vice versa. Since the market of flow-meters is overwhelming, it is always a delicate problem to find the most suitable one for a specific application.

One can distinguish between two philosophies when performing a flow measurement; to measure local properties or integrating over the pipe cross section. Local measurement techniques, such as pitot tubes, hot wire and laser doppler anemometers, are usually used in research laboratories because of the interest in local flow parameters. For industrial applications, the integrating type of flow meters is usually used.

Since the FlowAct method focus on flow in large pipes with high flow rate, it is interesting to know what competing methods are available commercially. A rough selection comes up with four different types of integrating flow-meters working satisfactorily under these conditions; differential pressure, turbine, electromagnetic and ultrasonic meters. A brief description of the different types, with their advantages and disadvantages follows. For a more detailed discussion Refs. [1] and [2] can be consulted.

Differential Pressure Device

The most common and well known flow-meters are the differential pressure devices, e.g. orifice plates and venturi meters. These meters are well documented, based on long experience and can be constructed from standard design guides. The working principle is to disturb the flow with an abrupt (orifice plate) or a smooth (venturi tube) area change and record the pressure difference caused by the disturbance. Due to poor diffusion in the flow after the device, a high pressure loss is introduced. Further, they are very sensitive to installation effects, such as small upstream distances to pipe bends or valves. The accuracy of these meters is moderate.

Turbine Meters

The revolution of turbine wheels gives a good measure of the flow passing the wheel, provided the bearing drag is small and the blades are well designed. Due to the bearing wear, these meters need frequent calibration and for high accuracy performance, recalibration should be built into the flow system. Since the meter is sensitive to the angle between the fluid and the blade, it is often used in conjunction with a flow straightener for best performance.

Electromagnetic Meters

When a conducting fluid flows in a transverse magnetic field, voltages and currents are generated due to the motion. If the voltage is measured between two electrodes in the pipe wall, it will provide an indication of the volumetric flow rate. Advantages of this method are that the flow is undisturbed, the response is linear and no moving parts are introduced. This type of meters is not as sensitive as the differential pressure devices to installation effects, like upstream pipe bends and valves. The method has also a low sensitivity to upstream conditions. However, for practical reasons, a uniform magnetic field is hard to achieve in the fluid, thus a weight function has to be introduced which indicates the importance of flow in each part of the pipe cross section. The weight function is also affected by an asymmetric flow profile. In many cases the weight func-

tion is difficult to calculate and sometimes it is not possible at all. However, the demand of conductivity excludes measurements of pure water.

Ultrasonic Meters

Ultrasonic meters are, like electromagnetic meters, non-intrusive and fall into three different categories depending on operating principle; transit time meters, correlation meters and doppler meters. However, the doppler meter does not have a high accuracy and in most cases it demands multi phase flows, therefore it is not discussed here any further.

Ultrasonic transit time meters measure the difference in transit time for up and downstream acoustic pulses. Upstream pulses are retarded by the flow and take longer time to reach the other end of the pipe, while downstream pulses are accelerated and take shorter time. From these two values of elapsed time the speed of sound in the fluid and the flow velocity are obtained. It is preferable to obtain the speed of sound in the fluid because the accuracy is dependent on it, and the speed of sound is affected by temperature, flow parameters, etc. Moreover, the accuracy is also affected by a non-uniform flow profile.

The working principle of a correlation meter is to transmit an acoustic wave through the fluid and on a diametral position of the pipe record the transmitted wave. This procedure is performed at two locations displaced in the axial direction. The signals obtained, disturbed by turbulence or a second phase, are then correlated and the time lag in the downstream signal is a measure of the transit time of the disturbance between the two locations. Further, the fluid velocity is determined from the time lag if the distance between the two positions is known. Since the signal is affected along the whole beam path, it is difficult to get a relation between the measured velocity and the mean velocity.

2. The Pulsed Neutron Activation Method

The essence of the pulsed neutron activation (PNA) method is to determine the transit time between two positions for a fluid in a pipe. By activating the fluid with a short neutron pulse and then recording the pass of the induced activity at a position downstream the pipe, a transit time can be estimated. An average velocity of the fluid can be calculated if the distance between the activation and detection points is known. Since the method is density sensitive, it is also applicable on two-phase flow. If a portable pulsed neutron source is available, a measurement based on the PNA method can be done anywhere without any damage on the pipe. Thus, if sufficient accuracy is obtained, the method is well suited for on-site calibration of existing flow-meters.

Flow rate measurements of liquids, solids and slurries by neutron activation were first reported in 1971, where Boswell and Pierce, Ref. [3] showed the feasibility of the method. It has also been used to calibrate and measure liquid sodium flow rates in the secondary system of the Experimental Breeder Reactor II (EBR-II) at the Argonne National Laboratory (ANL), Refs [4], [5] and [6]. Measurements of single and two phase flow have been reported from ANL as well as from Rensselaer Polytechnic Insti-

tute (RPI), Refs. [7], [8], [9], [10] and [11]. PNA technique has also been used at the loss-of-fluid-test (LOFT) facility, Refs. [12], [13], [14] and [15].

A PNA measurement can be divided into three different steps that will be discussed in some detail below. Namely, activation of the fluid, transport of the induced activity and detection of the induced activity at a distance downstream the activation point. In principle, all three steps concern transport of different types of particles. Nevertheless, the time scale or velocity of these processes differ very significantly. The activation (neutron transport) and the detection (gamma transport) are so fast in comparison with the fluid flow that a stationary fluid can be assumed. Thus the task is well defined and can be tackled by standard methods of radiation transport, in particular Monte Carlo methods. The fluid dynamic part is much more complicated and sufficiently accurate quantitative determination of the transport and mixing of the activated component within the flow requires sophisticated methods and careful selection of calculational methods.

2.1 Activation and Detection Profiles

For fluids containing oxygen, e.g. water, the activity is produced by the $^{16}\text{O}(n,p)^{16}\text{N}$ reaction. The resulting activity, in the form of ^{16}N , decays with a high energy γ -photon ($E_\gamma=6.1$ MeV) with a short half-life ($T_{1/2}=7.1$ s). Regarding detection technique, the γ -photons are easy to distinguish from the low energy γ -ray background. Moreover, the fast decay is preferable from the radiation protection point of view. In Ref. [3], activation reactions are suggested for other types of substances.

The cross section (or probability) for the above activation reaction, seen in Fig. 1, has a threshold energy for the incoming neutron. Compared to the scattering cross section for hydrogen and oxygen, the activation cross section is small, and the dominant neutron interaction with the water molecule is thus scattering. Normally, when the neutron undergoes a scattering event, it loses energy and the probability is high that the scattered neutron has an energy below the activation cross section. Hence, a first approxi-

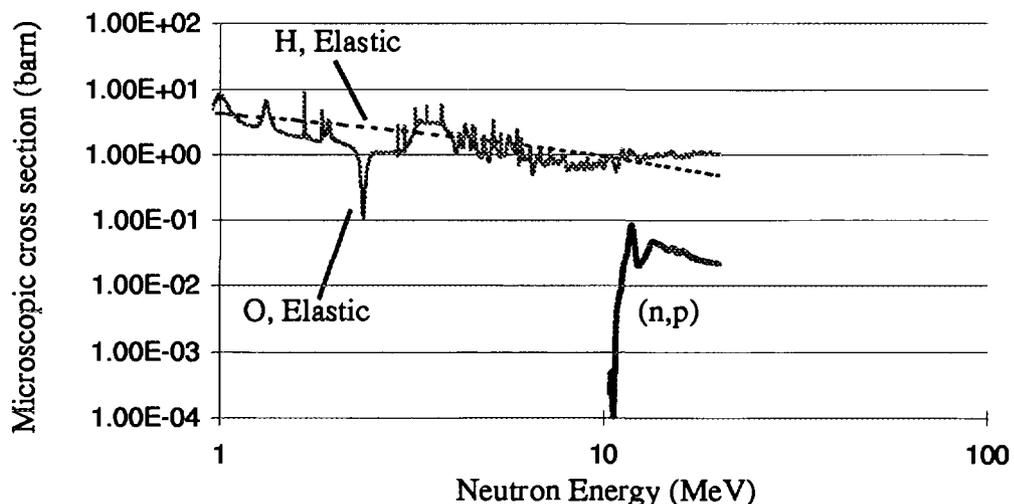


FIGURE 1. Microscopic cross section for $^{16}\text{O}(n,p)^{16}\text{N}$ compared to the elastic scattering cross sections for hydrogen and oxygen. Data from JEF 2.2.

mation for the neutron transport is to neglect the scattering and only consider the

uncollided neutron flux. A similar discussion can be performed for the γ -photon transport if only high energy γ -rays are considered.

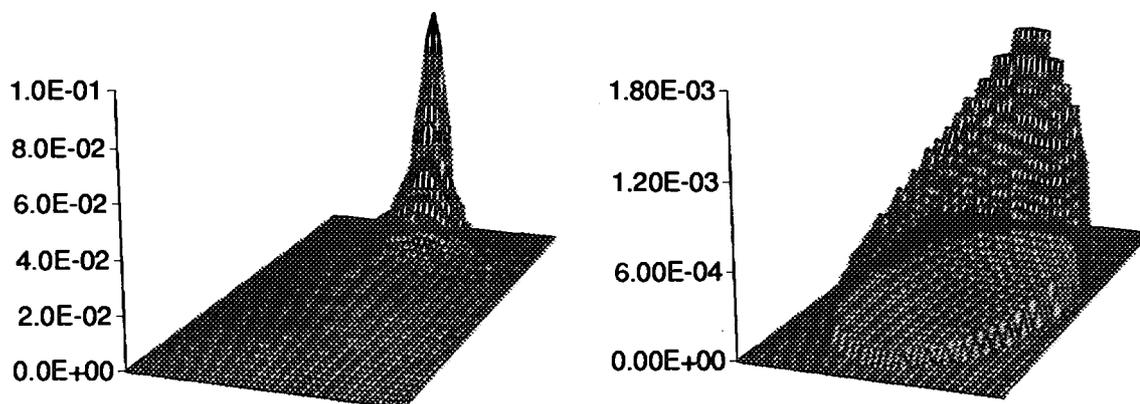


FIGURE 2. PNA activation profiles in a radial plane calculated from the uncollided neutron flux. In the left case, the source is positioned on the peripheral of the pipe and in the right case the distance between the pipe and the source is 1 diameter.

The penetration depths of neutrons and γ -rays are limited by the attenuation in the flowing fluid, hence non-uniform and geometrically non-symmetric activation and detection profiles are obtained. There are certain ways of affecting the shape of these profiles by changing the experimental setup. The axial shape depends on the collimation of the source and the detector, respectively. The radial and/or angular shape is dependent on the source and the detector geometry (usually fixed) and the distance between the source (or the detector) and the pipe. However, the attenuation of the beam from the source into the medium and from the fluid into the detector can not be counteracted. Calculations of activation profiles, using the uncollided neutron flux, are shown as an illustration in Figs 2 and 3.

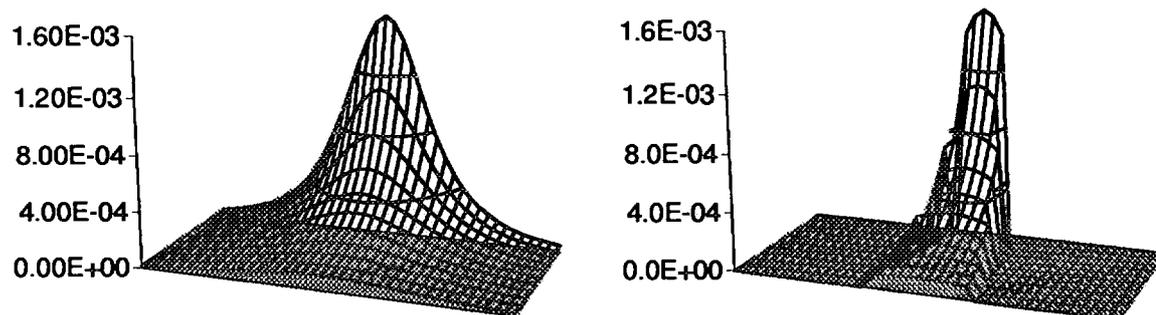


FIGURE 3. PNA activation profiles in an axial plane calculated from the uncollided neutron flux. In the left case the source is uncollimated and in the right case a narrow collimator is used.

A more accurate way of determining these profiles is to perform Monte Carlo simulations, where the scattering of the neutrons and gammas are considered.

2.2 Activity Transport

Because of the asymmetric activation, a detailed understanding of the activity transport is necessary. For single phase turbulent flow, the induced activity is affected by molecular and turbulent diffusion, as well as a velocity profile. Thus theoretical models, considering all these effects, are very complicated and it would be preferable to use a simpler but still accurate model to describe the activity transport. Early works on soluble matter in laminar and turbulent flow, done by Taylor, Refs. [16] and [17], provided the starting point for the developing of such models.

Taylor showed that the centre of diffusion, for a uniformly distributed substance in a pipe, is moving with the mean flow velocity. This is valid, for both laminar and turbulent flow, for large distances ($L/D=50$) from the point where the substance is injected into the flow. Consider a plane, moving at the mean velocity U in the positive z -direction, with a mean concentration $C_m = C_m(z, t)$ of a soluble substance. The diffusion equation for this moving plane is:

$$K \cdot \frac{\partial^2 C_m}{\partial z_1^2} = \frac{\partial C_m}{\partial t} \quad (1)$$

where K is a virtual coefficient of diffusion and z_1 is the center position of the plane: $z_1 = z - U \cdot t$. Assume a uniform concentration distribution induced in a plane at $z = 0$ at time $t = 0$ with A particles per unit area. With the following boundary conditions:

$$\lim_{z \rightarrow \infty} C_m = 0 \quad (2)$$

$$\int_0^{\infty} C_m \cdot dz_1 = A \quad (3)$$

limiting the solution of Eqn (1) to be finite at large distances and conserving the number of particles, the solution of Eqn (1) is:

$$C_m(z_1, t) = \frac{A}{\sqrt{\pi K t}} \cdot \exp\left(-\frac{z_1^2}{4 K t}\right). \quad (4)$$

For the PNA case, the induced substance is radioactive and decays with a decay constant λ , therefore the number of particles induced is decreasing with time. Radioactivity is governed by the well known exponential decay:

$$N = N_0 \cdot \exp(-\lambda \cdot t) \quad (5)$$

where N is the number of particles at time t , given N_0 particles at $t = 0$.

Since the number of induced particles in the pipe is A , corresponds to N_0 in Eqn (5), the conservation of particles is no longer valid and A has to be decay compensated.

Finally, the decay compensated mean concentration in a plane moving at velocity U can be expressed:

$$C_m(z, t) = \frac{A \cdot \exp(-\lambda \cdot t)}{\sqrt{\pi K t}} \cdot \exp\left(-\frac{(z - U \cdot t)^2}{4 K t}\right). \quad (6)$$

Eqn (6) is a first order approximation of a PNA time distribution and the value of K is depending on whether the flow is laminar or turbulent.

3. The FlowAct Project

One requirement for the use of the PNA method is that a pulsed neutron source is available. A permanent neutron generator of type SAMES at the Department of Reactor Physics at Chalmers University of Technology, extensively used for time-of-flight measurements and development of neutron spectrometers for fusion diagnostics, has been used as a pulsed neutron source during these measurements. It can provide a neutron yield of 10^{11} neutrons/s in the D-T mode with a shortest pulse duration of 1 ns. The provided neutron energy is 14 MeV, well above the threshold of the activation cross section (~ 10 MeV).

The first pilot FlowAct experiments and model calculations were done 1994 by Drozdowicz, Ref. [18], and Grosshög, Pázsit, Ref. [19]. The goal of those measurements was to achieve some basic understanding and hands-on experience with the technique. The aim was to investigate the reproducibility and reliability of the FlowAct method. The measurements were performed with already existing equipment and the results from these measurements were positive. A water loop with moderate accuracy flow calibration were used during these measurements and the same loop were also used during the next phase of the FlowAct project.

During 1995, [Paper 1], measurements were performed with dedicated equipment, i.e. two separate counting lines each consisting of a BGO scintillating crystal and computer hardware and software, under more realistic conditions. The aim of these measurements was to investigate the influence of certain ingoing parameters such as the distance between the source and the detectors, neutron pulse duration and reference flow velocity. The results from these measurements were encouraging and provided support for further investigations with a higher degree of flow calibration accuracy.

To meet the new and harder flow calibration requirements, a totally new flow loop had to be designed and built. By the summer of 1996 the new loop was complete and during tests with the loop, the performance seemed promising. The objective of the following measurements, [Paper 2] was to provide high accuracy flow calibration PNA data and, if possible, develop a simple method to extract the mean flow velocity with high accuracy from the data obtained. Time averaging methods, reported earlier Ref. [9], were used to analyze these measurements for comparison.

4. Results

During the preliminary FlowAct measurements, [Paper 1], four parameters affecting the time distributions obtained were varied. Namely, the lower gamma discrimination level, the neutron pulse length, the distance between the source and the detectors and, finally, the reference velocity. With a more accurate flow calibration possibility, the following measurements were more dedicated to investigate the accuracy dependence of the flow velocity determination. Two parameters, the distance between the source and the detector and the reference flow velocity, were varied during these measurements.

Raw data from a FlowAct measurement consist of two separate time distributions recorded at two locations displaced a known distance in the axial direction. Typical raw data can be seen in Fig. 4. From these distributions certain data could be extracted, e.g. average flow velocity. In most PNA measurements, a single detector position is used and the average velocity is extracted from the transit time between the source and the only detector position. With the FlowAct method, more possibilities are open for determination of an average flow velocity, e.g. using an average of the two transit times between the source and both detectors or using the transit time between the two detectors.

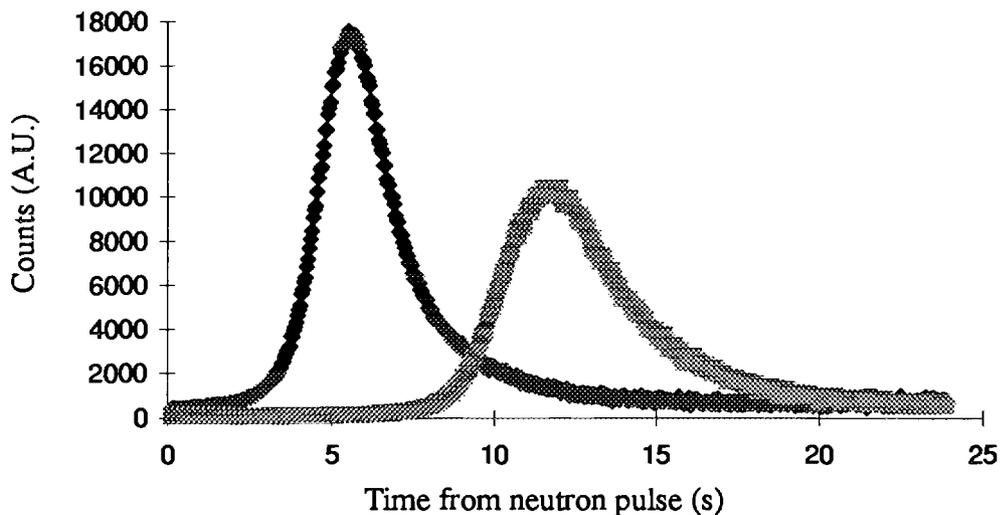


FIGURE 4. Decay compensated FlowAct time distributions for different distances between the activation point and the detector location.

Below a brief discussion of the results obtained during the two series of measurements covered by this thesis follows.

4.1 Moderate Accuracy Flow Calibration Measurements

The measured gamma-ray energy is affected by the setting of the discriminator window. The upper level of the discriminator was set to a value corresponding to 10 MeV gamma-ray energy while the lower level was varied, corresponding to an energy in the range 1-8 MeV, during a series of measurements. In the lower limit of this range, mostly all gamma-rays reaching the detector are counted, thus the fraction of low energy gamma-ray background is high. In the higher limit no counts at all will be registered, due to the limited energy of the decaying activity, and an optimum for the lower

level discriminator is expected. The results from these measurements, considering both the signal-to-background ratio and the variance-to-signal ratio, show that a favourable value of the lower level discriminator is in the range 4-5 MeV. However, one motive for even lower values is to collect as much information as possible during a measurement.

The neutron pulse duration affects the shape and the magnitude of the induced activity profile. The pulse should be sufficiently long in order to yield enough activation, but on the other hand it should be short, compared to the actual transit time, to get a good time resolution. Regarding counting statistics, as long neutron pulse as possible is preferred. In practical cases, it is possible to check whether the pulse is too long by performing measurements with different pulse duration and observe if the width of the time distributions obtained is affected. Such measurements were performed and it was found that the width of the time distributions was not changing when increasing the neutron pulse length from 50 to 100 ms.

During the transport in the pipe the induced activity profile broadens, especially in the axial direction. This broadening, caused by molecular and turbulent diffusion as well as by the velocity profile, can be measured by recording time distributions at various distances from the source. A linear dependence between the width and the distance from the source was observed during such measurements. By assuming the areas of the decay corrected time distributions being constant during these measurements, the influence of the linear background reduction could be checked. It was found that the linear assumption was valid throughout these measurements, except when the detector was located close to the source.

Flow velocity measurements, done with the simple flow calibration loop, were analysed with a simple method for extracting the average velocity, [Paper 1]. The results showed that the relative error in the determination of the average velocity was approximately 4% for Reynolds numbers greater than 10000.

4.2 High Accuracy Flow Calibration Measurements

High accuracy flow calibration demands a facility with stable flow conditions. In tests, the flow stability of the newly designed loop was found to be rather good, better than 0.2% in the mean flow velocity determination. This is in fact better than the design requirements of 0.5%. After solving some start-up problems, the flow loop works well.

Two models have been used in the analysis of these measurements, [Paper 2]. One time averaging technique and one fit to the Taylor theory, Eqn. (6). The time averaging technique used here, described in Ref. [9], is often referred to as the “1/t weighting method” and has been used earlier in a number of PNA measurements, yielding an accuracy in the average flow velocity of 2-3%.

The reproducibility of the FlowAct method was investigated during these measurements. The spread in the relative error of the average velocity measured has been used in the reproducibility study. The results from this study show a spread of 0.6% in the averaged flow velocity, obtained from the “1/t weighting method”. This is in the same order of magnitude as for the flow loop itself and thus satisfactory.

Like in the first preliminary tests [Paper 1], the distance between the pulsed neutron source and the detector was varied during a series of measurements and the relative error in measured average velocity as a function of distance was obtained. A comparison between the two analysis methods used, yields some interesting results. The results from the “1/t weighting method” show both positive and negative relative error values, -3 - +3%, while there are only positive values for the Taylor theory extracted values, 1 - 6%. However, the spread of the results from the “1/t weighting method” is greater than for the fit.

The reference velocity dependence on the relative error was studied in a series of measurements. The shape of the error functions obtained from the two analysis methods is similar and the magnitude of the error is increasing for decreasing velocities. For Reynolds numbers greater than 10000, the magnitude of the relative error for both analysis methods are not greater than 4%. Also here, the spread in the results from the “1/t weighting method” is greater than for the fit. One explanation for this is that the “1/t weighting method” is more sensitive to the background reduction procedure.

In comparison, the accuracy in mean flow velocity using the “1/t weighting method” obtained from these measurements is in agreement with earlier works, 2-3%, Refs. [4] and [14].

5. Summary and Conclusions

Two series of water flow measurements with the pulsed neutron activation method have been performed and analysed. The first series, performed under moderate flow calibration conditions, had the objective of gaining sufficient experience of the method and provide indication of performance. A mapping of some ingoing parameters was done and a simple method determining the mean flow velocity from two time distributions obtained at axially displaced locations was used. The results from these measurements were promising and further measurements were planned.

For high accuracy flow calibration, a dedicated flow loop was designed, built and tested. Providing high accuracy flow calibration, the aim for the next series of measurement was to investigate the potential for high accuracy flow determination using the PNA method. Existing analysis methods were used for comparisons with earlier works and the results were in good agreement. The accuracy dependence on ingoing parameters, such as reference velocity and distance between source and detectors, was investigated using both time averaging methods and Taylor theory. The range of the Reynolds number used indicate that the turbulence is not fully developed. It is expected that for even higher Reynolds number, integrated flow behaviour will become more stable and thus the accuracy, obtained from these measurements, can be seen as a worst case.

The measurements reported in this thesis confirm the expectation that the FlowAct method can be developed into a high-accuracy non-intrusive flow measurement tool. To increase the accuracy of the method, more refined analysis and data processing methods are necessary. Radiative transport simulations in conjunction with advanced flow dynamic calculations, can in the future be used for the development of such methods. If sufficient accuracy is obtained, the non-intrusive performance of the method makes it suitable for on-site calibration of permanently installed flow-meters. It can also be mentioned that the applicability of the FlowAct method becomes better for

higher Reynolds numbers, due to a flatter velocity profile, more intensive mixing, etc. Thus the chances of applying a refined version of the FlowAct method on flows with much higher Reynolds numbers than the present ones seems quite promising.

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