03

Control of fluid flow movement in porous medium with NMR-relaxometry method

© V.V. Maikov, S.V. Zhakov, I.V. Byzov, A.A. Mysik

M.N. Mikheev Institute of Metal Physics of the Ural Branch of the Russian Academy of Sciences,
620108, Yekaterinburg, Russia
e-mail: mvv@imp.uran.ru

Received August 11, 2022
Revised August 11, 2022
Accepted October 6, 2022

The possibility of fixing velocities of fluid motion in model porous medium (glass beads) by measuring the transverse relaxation time T_2 by spin echo method is shown. The magnetic field gradient arising due to the difference in the magnetic permeability of the fluid and the porous medium causes a decrease in the average time T_2, wich makes it possible to fix the fluid motion in the absence of an external magnetic field gradient. The addition of magnetic nanoparticles to the fluid under study increases the dependence of T_2 on fluid velocity and increases the sensitivity in determining the permeability of porous medium by NMR-relaxometry.

Keywords: porous media, fluid flow, NMR-relaxometry, magnetic nanoparticles.

DOI: 10.21883/TP.2023.01.55438.200-22n

Introduction

Understanding characteristic properties of the process of fluid motion in porous media is important for the problems of petrophysics, efficiency of oil production, soil and environment analysis, healthcare [1,2]. In petrophysical applications the measurement of fluid velocity in porous media is necessary for both the logging of pay zone permeability by downhole instruments and laboratory core examinations. At the same time it is necessary to provide for measurement of low velocities of fluid motion to ensure reliable permeability logging for real natural porous media. In recent years, also studies were published demonstrating the possibility of affecting the fluid motion in porous media by varying viscosity coefficient of the fluid and the character of surface wetting through addition of nanoparticles with different compositions into the fluid [3–5].

The method of nuclear magnetic resonance (NMR) is rather actively used in petrophysical applications for logging of fluid-saturated porous media. This method is non-destructive and allows for logging of porosity and type of fluid in pores in optically non-transparent media [6]. In addition, NMR allows measurement of fluid motion velocity by changes in the echo phase and amplitude of the NMR-signal [7].

In [8] the influence of fluid motion in porous media on the transverse relaxation time T_2 is shown with measurement of NMR signal by Carr–Purcell–Meiboom–Gill method (CPMG)-sequence) in a magnetic field gradient. Based on results of these experiments an assumption can be made that the observed decrease in T_2 is caused by the emergence of a chaotic velocity component when the fluid moves in a porous medium. As a result, an additional contribution to the spin-spin relaxation takes place similar to the contribution caused by diffusion.

Earlier it was shown [9] that T_2 depends on the concentration of magnetic nanoparticles (MNP) in water, which decrease T_2 in the free volume due to the local gradients of magnetic field created by them. In the case of motion of a hydrogen-containing fluid where MNPs are suspended, an additional decrease in T_2 can be expected if the MNP velocity in the flow is not equal to the fluid velocity. In this case a contribution to the spin-spin relaxation will take place, which will be caused by the additional change in local gradients of magnetic field acting on protons of the fluid. This situation is possible when the fluid moves in a porous medium or in cylinder channels, where fluid velocity depends on the distance to the channel wall [1].

The goal of this work was to investigate the dependence of T_2 on the local gradients of magnetic field arising in porous media due to magnetic susceptibility of the medium and MNPs suspended in the moving fluid. The work was aimed to determine the possibility to improve the sensitivity when determining fluid motion velocity in porous media by the change in T_2 due to addition of MNPs to the fluid, i.e. to find out the possibility to improve the sensitivity when determining permeability of porous samples.

1. Experimental results

In the experiments of this work we used a model porous medium composed of glass balls with a diameter of 0.4 mm. In the course of experiments the balls were placed in a plastic cylinder with a diameter of 40 mm. Length of the region filled with balls was 40 mm. Measurements of T_2 were carried out by a NMR-relaxometer with a Larmor frequency of $f = 2.5$ MHz manufactured by the Institute of Metal Physics of the Ural Branch of the Russian Academy of Sciences. The magnetic field gradient was created using current coils. The experiment scheme is shown in Fig. 1. In the process of experiment volume of the fluid V_o was measured that passed through the sample during a fixed time t and average velocity of fluid in the sample $V = 4V_0/(\pi d^2 t)$ was determined, where d being inner diameter of the cylinder filled with balls.

The NMR-relaxation curve was measured by the Carr–Purcell–Meiboom–Gill method with motionless fluid and in the process of fluid motion at different velocities and different intervals TE between spin echoes. We used distilled water in the experiments. The relaxation time measured on the model sample of glass balls used in the study was $T_2 \sim 1000$ ms for motionless fluid. The magnetic field gradient in the experiments was 40 mT/m, i. e. taking into account the relaxometer receiver band of about 20 kHz, the region of NMR-signal receiving on protons was about 0.01 m along the sample axis.

When carrying out measurements on a porous sample, inversion of the relaxation curve results in a distribution over times T_2, which changes depending on velocity of fluid motion in the sample. With increase in the fluid velocity this distribution shifts towards lower values of T_2 [8,10]. We have taken the geometrical mean time $\langle T_2 \rangle$, as a parameter characterizing fluid velocity in a sample, which was determined for each velocity as follows:

$$\langle T_2 \rangle = \exp\left(\sum_{i=1}^{N} A_i \ln T_{2i} / \sum_{i=1}^{N} A_i \right). \qquad (1)$$

In (1) A_i and T_{2i} are amplitudes and times of relaxation, respectively, derived from the inversion of the experimental

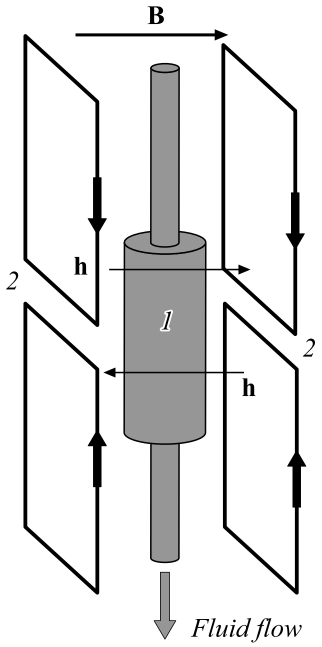

Figure 1. Experiment scheme: *1* — sample, *2* — gradient coils, **B** — field of the NMR-relaxometer magnet, **h** — field of gradient coils.

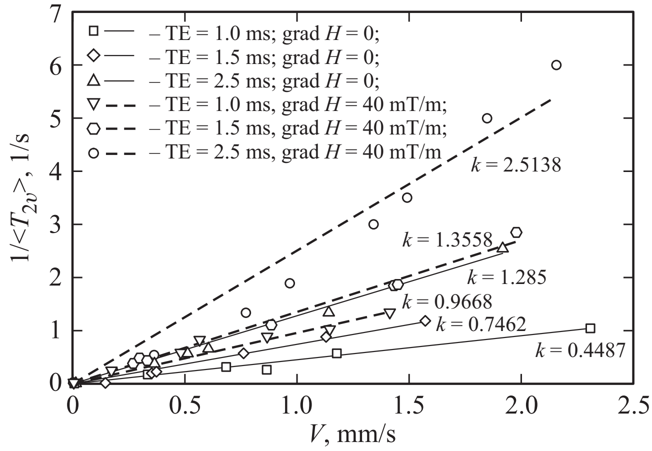

Figure 2. Dependencies of $1/\langle T_{2v} \rangle$ on the motion velocity V without and with magnetic field gradient at different TEs for water.

relaxation curve $R(t)$:

$$R(t) = \sum_{i=1}^{N} A_i \exp(-t/T_{2i}). \qquad (2)$$

Then a contribution to T_2 is calculated, which is caused by fluid motion only:

$$\frac{1}{\langle T_{2v} \rangle} = \frac{1}{\langle T_2(V) \rangle} - \frac{1}{\langle T_2(V=0) \rangle}. \qquad (3)$$

The experimentally observed dependence of relaxation rate $1/\langle T_{2V} \rangle$ on fluid velocity V was close to linear and can be approximately represented in the following form: $1/\langle T_{2v} \rangle = k^* V$. Therefore, when analyzing the experiments, we used parameter k to characterize the susceptibility of the measured value of T_2 to the fluid motion velocity.

Results of the performed experiments can be summarized as follows. Porous medium–balls, fluid–water (Fig. 2). If the external magnetic field gradient is equal to zero (grad H = 0), then coefficient $k = 0.45{-}1.28$ and grows with increase in TE; with an external gradient of 0.04 T/m coefficient $k = 0.96{-}2.5$ and grows with increase in TE as well. The result of the second experiment is similar to that obtained earlier for porous samples [8]. In the case of fluid motion through a porous medium in a magnetic field gradient an additional contribution to the relaxation takes place, being caused by the emergence of a chaotic component of the fluid velocity.

In the first experiment a decrease in T_2 was also observed, although the external magnetic field gradient was equal to zero. This may be caused by the following: first, a change in the magnetic susceptibility at the „fluid–ball surface" interface, which results in the emergence of local gradients of magnetic field near the ball surface. As a result, the unordered motion of fluid in the medium filled with balls leads to emergence of a contribution to the relaxation; second, a decrease in T_2 may be caused by the fact that a fluid volume entered the region of NMR-signal measurement, that was not exposed to the 90-degrees pulse.

To verify the results obtained at a zero gradient of magnetic field, we have measured the dependence of T_2 on the fluid motion velocity in cylinder channels. We used cylinder samples made of plastic by 3*D*-printing method with channels oriented along the longitudinal axis, with diameters of 0.5 and 2 mm. With these diameters of channels and low fluid velocities there is no chaotic component of velocity and flow can be considered laminar.

In these experiments at a zero external gradient of magnetic field the value of k at TE = 1.0, 1.5, 2.5 ms was varied in the range of $k = 0.016-0.022$ for channels with a diameter of 0.5 mm and $k = 0.04-0.044$ for channels with a diameter of 2 mm.

With an external gradient of 40 mT/m and at the same TE $k = 0.021-0.027$ for channels with a diameter of 0.5 mm and $k = 0.046-0.052$ for channels with a diameter of 2 mm, i.e. the dependence of $\langle T_{2v} \rangle$ on the fluid velocity was almost absent. This is indicative of the fact, that at the fluid motion velocities and magnetic field gradient used the decrease in T_2 due to the arriving of fluid not subjected to 90-degrees pulse to the NMR-signal measurement region is negligible. Therefore, a decrease in T_2 when the fluid moves in a zero external gradient of magnetic field may be caused by local gradients of magnetic field created by the balls.

Measurement of magnetization curves of balls using the „Faraday balance" setup manufactured by the Institute of Metal Physics of the Ural Branch of the Russian Academy of Sciences has shown that the specific magnetization of balls in a magnetic field of $H = 60$ mT, corresponding to the field in the magnetic system of the relaxometer used, is $\sigma = 0.00125$ emu/g. As a result the layer of fluid immediately adjacent to the ball surface is in the magnetic field gradient. Thus, even a very small magnetization of the matrix that forms the porous medium results in the observed decrease in time T_2 when the fluid moves.

To verify the influence of magnetic nanoparticles (MNP) on the value of T_2 in a moving fluid, we used carbon-coated nanoparticles of iron, Fe@C, with their surface coated by a non-ionic surfactant (DSPE-mPEG 2000, Lipoid GmbH, Germany) to provide for the aggregative stability in aqueous environment and prevent their interaction with the ball surface. Hydrodynamic diameter of nanoparticles was 150 nm (determined by the method of dynamic light scattering using NanoZS, Malvern Instruments, Great Britain), specific magnetization was 90 emu/g (measured by a vibration magnetometer manufactured by the Institute of Metal Physics of the Ural Branch of the Russian Academy of Sciences).

Value of T_2 measured for the MNP suspension in water, depends on particles concentration to a significant extent [9]. In these experiments we used a concentration of particles of $C = 0.0004$ mg/ml. This concentration results in $T_2 = 500$ ms in the free volume, i.e. it considerably decreases T_2 in comparison with pure water ($T_2 = 2.2-2.5$ s),

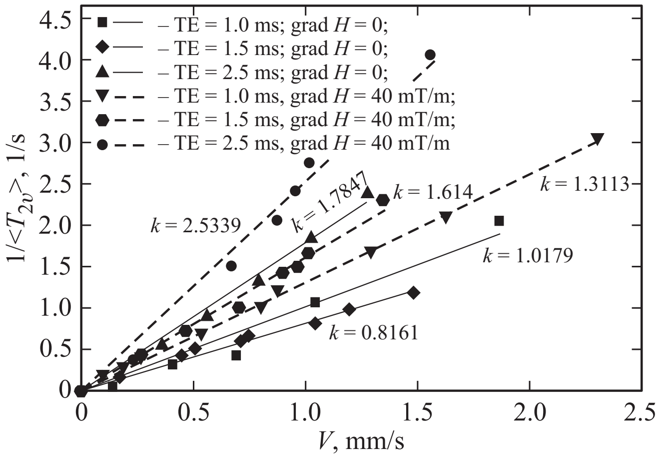

Figure 3. Dependencies of $1/\langle T_{2v} \rangle$ on the motion velocity V without and with magnetic field gradient at different TEs for water with magnetic nanoparticles (MNPs).

which reduces the duration of experiment, but at the same time allows tracking the decrease in T_2 due to the fluid motion.

Fig. 3 shows measured dependencies of $1/\langle T_{2v} \rangle$ on velocity V for water containing MNPs, at different TE, external gradient grad H = 0 and grad H = 40 mT/m. It can be seen, that at grad H = 0, k factor (the slope of straight lines) is $\sim 1.3-1.5$ times higher than that in the first experiment for a fluid without MNP. The inclusion of gradient in this case insignificantly increases coefficient k at low TEs, i.e. in comparison with local gradients created by MNPs, the influence of external gradient in this case is low.

In this experiment the measured value of k factor at TE = 1.0 ms is greater than that at TE = 1.5 ms. Perhaps, it is related to the change in local concentration of MNP in the process of liquid motion.

Fig. 4 shows for comparison the dependencies of $1/\langle T_{2v} \rangle$ on velocity V at a zero external gradient for water and for MNP-containing water. It can be seen, that a significant growth of relaxation rate arises at TE = 2.5 ms.

The decrease in time T_2 arising in the case of MNP-containing fluid motion in comparison with the T_2 for motionless fluid is indicative of the fact that the MNP velocity is not equal to the velocity of carrier fluid when the flow moves in a porous medium. As a result, the emerging difference in velocities between MNPs and fluid molecules leads to an additional change in local gradients of magnetic field in comparison with the case of $V = 0$, an additional contribution to relaxation, and the observed decrease in T_2.

A useful practical conclusion follows from this effect. The presence of MNPs in the fluid flow allows increasing the sensitivity when measuring the fluid motion velocity in a porous medium by the change in T_2, i.e. improvement of sensitivity when measuring the permeability of a porous sample by the NMR-method.

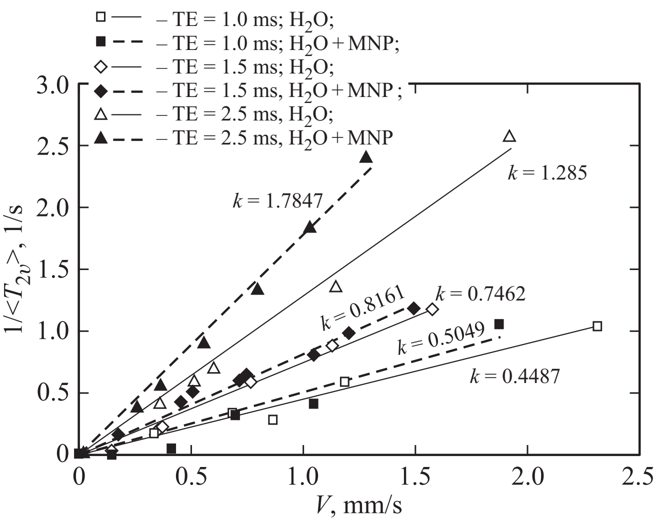

Figure 4. Dependencies of $1/\langle T_{2v}\rangle$ on the motion velocity V at a zero external magnetic field gradient for water and for the water containing MNPs, at different TEs.

Conclusions

1. The MNP velocity in a porous medium is not equal to the velocity of carrier fluid.

2. Fluid motion in a porous medium with a magnetic permeability even slightly greater than unit, which is located in a constant homogeneous magnetic field, causes an easily measured decrease in the transverse relaxation time in comparison with T_2 for motionless fluid. A gradient of magnetic field results in an additional decrease in T_2 in the case of fluid motion in a porous medium. Since the rock that forms many producing oil beds has a magnetic permeability greater than unit, the fluid motion and, as a consequence, the permeability can be measured by the decrease in T_2 when the fluid motion is started as a result of applied external pressure.

3. Motion of a MNP-containing fluid through a porous medium results in an additional decrease in the relaxation time T_2 in comparison with the motion of fluid without MNPs. As a result the use of MNPs can increase the sensitivity of fluid velocity measurement and determination of permeability of porous samples. The possibility of MNP usage to determine permeability needs to be further studied on samples of natural porous media.

Funding

The work was carried out within the „Magnet“ budget topic, State reg. № 122021000034-9

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] A. Donath, A. Kantzas, S. Bryant. Transport in Porous Media, **128**, 459 (2019). DOI: 10.1007/s11242-019-01256-4
[2] E. Fukushima. Annu. Rev. Fluid. Mech., **31**, 95 (1999). DOI: 10.1146/annurev.fluid.31.1.95
[3] M. Almahfood, B. Bai. J. Petroleum Sci. Engineer., **171**, 196 (2018). DOI: 10.1016/j.petrol.2018.07.030
[4] R. Hashemi, N.N. Nassar, P.P. Almao. Appl. Energy, **133**, 374 (2014). DOI: dx.doi.org/10.1016/j.apenergy.2014.07.069
[5] L. Hendraningrat, S. Li, O. Torsæter. J. Petroleum Sci. Engineer., **111**, 128 (2013). DOI: dx.doi.org/10.1016/j.petrol.2013.07.003
[6] G.R. Coates, L. Xiao, M. Prammer. *NMR Logging Principles and Applications* (Halliburton Energy Services, Houston, 1999)
[7] P.T. Callaghan. *Principles of Nuclear Magnetic Resonance Microscopy* (Oxford University Press Inc., NY., 2004)
[8] V. Loskutov, S. Zhakov. Int. J. Heat Mass Transfer, **101**, 692 (2016). DOI: dx.doi.org/10.1016/j.ijheatmasstransfer.2016.05.057
[9] I.V. Medvedeva, S.V. Zhakov, A.V. Revvo, I.V. Byzov, Yu.A. Bakhteeva, M.A. Uimin, A.E. Yermakov, A.A. Mysik. Phys. Metals Metallography, **115** (8), 744 (2014). DOI: 10.1134/S0031918X14080110
[10] M. Chen, J. Dai, X. Liu, Y. Kuang, Z. Wang, Sh. Gou, M. Qin, M. Li. J. Petroleum Sci. Engineer., **184**, 106588 (2020). DOI: 10.1016/j.petrol.2019.106588