

Jury Member Report – Doctor of Philosophy thesis.

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PhD Program: Petroleum Engineering

Title of Thesis: Application of LF-NMR measurements for characterization of unconventional using machine learning

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Name of the Reviewer: Prof. Dmitriy Potapenko

I confirm the absence of any conflict of interest

(Alternatively, Reviewer can formulate a possible conflict)

Date: 10-08-2022

The purpose of this report is to obtain an independent review from the members of PhD defense Jury before the thesis defense. The members of PhD defense Jury are asked to submit signed copy of the report at least 30 days prior the thesis defense. The Reviewers are asked to bring a copy of the completed report to the thesis defense and to discuss the contents of each report with each other before the thesis defense.

If the reviewers have any queries about the thesis which they wish to raise in advance, please contact the Chair of the Jury.

Reviewer's Report

Reviewers report should contain the following items:

- Brief evaluation of the thesis quality and overall structure of the dissertation.
- The relevance of the topic of dissertation work to its actual content
- The relevance of the methods used in the dissertation
- The scientific significance of the results obtained and their compliance with the international level and current state of the art
- The relevance of the obtained results to applications (if applicable)
- The quality of publications

The summary of issues to be addressed before/during the thesis defense

The work of Strahiniva Markovic is focused on development of the evaluation methods for recovery of high viscosity hydrocarbon resources with special focus on development of heavy oil and bitumen reservoirs. Specific problems covered in Mr. Markovic Ph.D. thesis include the use of low-magnetic field NMR for assessment of viscosity of the reservoir hydrocarbons and the saturation of formation fluids in the wide range of temperatures. In his work Mr. Markovic proposes a new workflow for assessment of the mentioned parameters which utilizes machine learning algorithms and proves its advantage over similar potential alternative methods.

The work of Mr. Markovic includes three logically-linked parts focused on three different components of the announced problem: Heavy oil viscosity assessment using low magnetic field NMR data; improved oil viscosity characterization using low magnetic field NMR and supervised machine learning; and using the machine learning technique of choice and low magnetic field NMR data for defining water saturation in Canadian oil sands. The thesis goes into details of the theoretical background of the proposed workflow and provides experimental evidences of its applicability for practical purposes. Obtained results demonstrate that application of this methodology may enable significant improvement in monitoring of the heavy oil/bitumen recovery during EOR processes and establishing proper control of the development of such reservoirs.

The thesis is very well written with an excellent logical structure behind it. Obtained results are properly supported by implementation of the methods of statistical analysis which demonstrates the systematic focus of the performed research and contributes to the reliability of the developed conclusions.

The candidate meets all formal criteria for the Ph.D. work. He has three paper publications in high-ranked international scientific journals (Q1/Q2 category), with one more publication under review, one patent application and two international conference proceedings. In addition to that the candidate has two non-thesis publications relevant to the field of the study.

Some of my minor non-essential comments to the thesis of Strahiniya Markovic are listed below. I hope that these comments, if they are accepted by the author, will help with further clarification of the background and emphasizing the importance of the performed work.

- Page 20, the last two sentence. Suggesting to re-phase as “The special attention is paid to development of new workflows and models for predicting oil viscosity and water saturation which are two key factors for recovery of hydrocarbons.”
- Page 25 - last 6 rows and page 26, first 12 rows. Suggesting to re-work the description of the theory of the NMR technique with the special focus on the difference between the magnetic moments of the individual protons and the behavior of the total magnetic moment of the system of such individual magnetic moments (spins) in the external magnetic field without and upon the irradiation. I think it would make sense to add the description of the corresponding Hamiltonian equation with explanation of the forbidden and the allowed energetic transitions in the system. Then, after providing the description of the NMR theory, it would also make sense to properly describe the theory behind the echo-type NMR measurements as briefly proposed in the corresponding comments to the manuscript.
- Page 30. I Recommend to clearly define what “low-field NMR” means in the scope of the performed work. Typically, low-field NMR is considered to take place then the strength of the magnetic field is comparable with the spin-spin hyperfine interaction constant. If different criterium is used, I would strongly suggest to emphasize it.

- As a refinement option I would suggest the author to do an additional stylistic check of the text of the manuscript (not the grammar which in my opinion is at the way much better level than I saw on most the reviewed publications.). I see that the meaning of some of the statements in the text may be further improved though their simplification and the use of a bit different vocabulary. Some of the suggestions can be found in the text of the thesis which is I am submitting along with the current review. However, this comment is extremely minor, and does not impact the overall quality of the manuscript as well as the quality the performed work. Considering the current timeframe, I am leaving it solely to the author's discretion.
- Overall, I am very impressed with the quality of the performed work and the prepared manuscript. I wish Mr. Markovic all his best in his further research and I am looking forward for the chance to get familiar with his further works.

All of these mentioned points should be considered solely as recommendations for further improvement, not as a criticism. Provided comments do not reduce the scientific value of the present work, which Mr. Markovic undoubtedly demonstrated.

Overall, the topic of the dissertation is relevant to its content. The methods employed are relevant to the dissertation subject. Obtained results are novel and have significant value. The thesis's results have been published in high-ranked peer-reviewed journals.

To conclude, Mr. Markovic definitely demonstrated his ability to perform research work and to apply the results of his research in practice. Mr. Markovic's thesis certainly satisfies the requirements of the Ph.D. program at Skoltech and I strongly recommend the Skoltech Ph.D. committee to grant this degree upon the review of the provided thesis.

Provisional Recommendation

I recommend that the candidate should defend the thesis by means of a formal thesis defense

I recommend that the candidate should defend the thesis by means of a formal thesis defense only after appropriate changes would be introduced in candidate's thesis according to the recommendations of the present report

The thesis is not acceptable and I recommend that the candidate be exempt from the formal thesis defense

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Skolkovo Institute of Science and Technology



Curtin University

Application of LF-NMR measurements for characterization of unconventional
hydrocarbons using machine learning

Doctoral Thesis

by

STRAHINJA MARKOVIC

JOINT DOCTORAL PROGRAM IN PETROLEUM ENGINEERING WITH SKOLKOVO
INSTITUTE OF SCIENCE AND TECHNOLOGY AND CURTIN UNIVERSITY

Supervisors:

Professor Alexey Cheremisin

Professor Reza Rezaee

Moscow - 2022

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I declare that the work presented in this thesis was carried out by myself at Skolkovo Institute of Science and Technology, Moscow, and Curtin University, Perth. To the best of my knowledge and belief this thesis contains no material previously published by any other person except where due acknowledgment has been made. This thesis contains no material which has been accepted for the award of any other degree or diploma in any university.

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A handwritten signature in cursive script, reading "Strahinja Markovic".

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Abstract

Heavy oil and bitumen resources are the most abundant hydrocarbon energy source worldwide. However, thermal enhanced oil recovery (EOR) methods are frequently applied to enhance their mobility and production due to their high viscosity. In addition, owing to the chemical dissimilarity of oils and various temperatures these oils are exposed to, as well as LF-NMR equipment limitations, the commonly used models fail to perform at a satisfactory level, making them impractical for use in heavy oil and bitumen reservoirs, and in environments with large temperature **oscillations** (e.g., mechanical systems). Information about the distribution of oil viscosity within the reservoir can be used to **help the management of the** thermal EOR **project**. Nuclear magnetic resonance (NMR) downhole tools provide a non-destructive way to determine the oil viscosity without recovering samples (core or produced oil) from the **wellbore**.

A new analytical NMR viscosity model was developed and tested on a suite of 23 Canadian heavy oils recovered from **different** reservoirs. The model was based on two NMR parameters – T_2 logarithmic mean and relative hydrogen index. Subsequently, the model was tested on a single bitumen sample at **a** temperature range from 26-200 °C. Results were compared to nine well-known NMR viscosity models **from** the literature, and in both cases, the **enhanced** model **scored** the most favorable statistical scores in this study. Furthermore, a simple model optimization procedure was presented, employing nonlinear least squares (NLS) regression. Experiments were carried out at temperatures corresponding to those in the hot steam injection EOR treatments. The same methodology can be extended for use in cyclic solvent injection (CSI), where the NMR model can detect oil viscosity changes when the solvent vapor dissolves into ~~the~~ oil.

To **make the viscosity prediction even more successful**, we developed a framework that combines supervised learning algorithms with domain knowledge for synthesizing new features using only one NMR parameter – the T_2

logarithmic mean. Two principal methods were considered, support vector regression (SVR) and gradient boosted trees (GBRT). Models were trained using the experimental data from our previous studies and literature data combining conventional oils, heavy oils, and bitumens from various reservoirs in Canada and the USA. The models' performance was compared against four other intelligent algorithms and four well-known empirical NMR models against which the SVR and GBRT-based models achieved the highest statistical scores. The proposed framework can also be applied to determine other physicochemical properties of oils by LF-NMR, where supervised learning is usually impractical due to the limited volume of data.

Finally, water saturation determination is among the most challenging tasks in petrophysical well-logging, which directly impacts the decision-making process in hydrocarbon exploration and production. However, quantification of oil and water volumes is problematic when their NMR signals are not distinct. We developed two machine learning frameworks to predict relative water content in oil-sand samples using LF-NMR spin-spin (T_2) relaxation and bulk density data to derive a model based on Extreme Gradient Boosting. The NMR T_2 distributions were obtained for 82 Canadian oil-sands samples at ambient and reservoir temperatures (164 data points). The actual water content was determined by Dean-Stark extraction. The statistical scores confirm the strong generalization ability of the feature engineering LF-NMR model and indicate that this approach can be extended for the improved *in-situ* water saturation evaluation by LF-NMR and bulk density measurements.

Publications

1. S. Markovic, J. L. Bryan, A. Turakhanov, A. Cheremisin, S. A. Raj Mehta, A. Kantzas. In-situ heavy oil viscosity prediction at high temperatures using low-field NMR relaxometry and nonlinear least squares. *Fuel, Volume 260, 2020.*

DOI: 10.1016/j.fuel.2019.116328

2. A. Askarova, A. Turakhanov, S. Markovic et al., Thermal enhanced oil recovery in deep heavy oil carbonates: an experimental and numerical study on a hot water injection performance, *Journal of Petroleum Science and Engineering, 2020.*

DOI: 10.1016/j.petrol.2020.107456

3. S. Markovic, J. L. Bryan, V. Ishimitsev, A. Turakhanov, R. Rezaee, A. Cheremisin, A. Kantzas, D. Koroteev, S. A. Mehta. Improved oil viscosity characterization by low-field NMR using feature engineering and supervised learning algorithms. *Energy & Fuels, 2020 34 (11), 13799-13813*

DOI: 10.1021/acs.energyfuels.0c02565

4. S. Markovic, J. L. Bryan, R. Rezaee, A. Turakhanov, A. Cheremisin, A. Kantzas, D. Koroteev. Application of XGBoost model for *in-situ* water saturation determination in Canadian oil-sands by LF-NMR and density data. *Scientific Reports, under review.*

Conferences and conference proceedings

1. S. Markovic, A. Turakhanov, A. Cheremisin, J. L. Bryan, S. A. Raj Mehta, A. Kantzas. Evaluation and prediction of heavy oil viscosity at high temperatures - application of low-field NMR measurements. *World Heavy Oil Congress & Exhibition, Muscat, Oman, 2018.*

2. S. Markovic, J. L. Bryan, R. Rezaee, A. Cheremisin, A. Kantzas. In-situ water saturation by LF-NMR and supervised learning - application to Canadian oil sands. *EAGE Geotech, London, United Kingdom, 2022.*

DOI: 10.3997/2214-4609.20224021

Patents

1. S. Markovic, A. Cheremisin, S.A. Mehta et al., RU Patent 2021111887, Method for thermal enhanced oil recovery tests on whole core samples, 2021.
2. Method for *in-situ* water saturation oil-sands by LF-NMR and density data and XGBoost algorithm; (**under preparation**).

Non-thesis publications, relevant for the field

1. A. Ivanova, A. Orekhov, S. Markovic et al., Live Imaging of Micro and Macro Wettability Variations of Carbonate Oil Reservoirs for Enhanced Oil Recovery and CO₂ Trapping/Storage, *Scientific Reports, Nature, 2022*
DOI: 10.1038/s41598-021-04661-2
2. E. Tokareva, I. Tkachev, G. Sansiev, G. Fedorchenko, A. Ivanova, P. Grishin, S. Markovic et al., Study of the process of hydrophobization of carbonate rock with organic acids (Russian), *Neftyanoe khozyaystvo-Oil Industry, OnePetro, 2022*
DOI: 10.24887/0028-2448-2022-3-73-76

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List of Symbols, Abbreviations

LF-NMR	Low-field nuclear magnetic resonance
OOIP	Original oil in place
EOR	Enhanced oil recovery
TE	Echo spacing
COD	Coefficient of determination
RMSE	Root mean square error
MAE	Mean absolute error
MSLE	Mean square logarithmic error
MAPE	Mean absolute percentage error
MaAE	Maximum absolute error
XGB	Extreme gradient boosting - Xgboost
FE	Feature engineering
FS	Full spectrum
XGB-FE	Extreme gradient boosting model based on feature engineering
XGB-FS	Extreme gradient boosting model based on full T ₂ distribution
DS	Dean-Stark
DS-w	Water content by Dean-Stark
DS-o	Oil content by Dean-Stark
L1	Lasso regression
L2	Ridge regression
CPMG	Carr-Purcell-Meiboom-Gill pulse sequence
SNR	Signal-to-noise ratio
CANOVA	Continuous analysis of covariance
MI	Mutual information
BO	Bayesian optimization
LOOCV	Leave-one-out cross-validation
X-ray CT	X-ray computed tomography
BSS	Blind-source signal separation
ANN	Artificial neural network(s)

SL	Supervised learning
PLS	Partial least squares
HI	Hydrogen index
RHI	Relative hydrogen index (mass)
RHI _v	Relative hydrogen index (volume)
SRM	Structural risk minimization
ERM	Empirical risk minimization
LAD	Least absolute deviation
GS-CV	Grid-search cross-validation
RBF	Radial basis function
SV	Support vector
SVR	Support vector regression
SVM	Support vector machine(s)
GBRT	Gradient boosted regression trees
MLR	Multiple linear regression
K-NN	K nearest neighbors
DT	Decision trees
RF	Random forests
NLS	Non-linear least squares
ODR	Orthogonal distance regression
GOR	Gas oil ratio
VAPEX	Vapor extraction
SAGD	Steam-assisted gravity drainage
ISC	In-situ combustion
DTS	Distributed temperature sensing
CSS	Cyclic steam stimulation
FID	Free induction decay
NE	Number of echoes within CPMG sequence
NT	Number of trains within CPMG sequence
TW	Wait-time within CPMG sequence

ILT	Inverse Laplace transform
BBP	Bloembergen-Purcell-Pound NMR relaxation model
RHOB	Bulk density log
NPHI	Neutron porosity log
FFI	Free-fluid index
DSE	Debye-Stokes-Einstein model
ASTM	American Society for Testing and Materials
PEEK	Polyether ether ketone
M-L	Marquardt-Levenberg

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Chapter 1 INTRODUCTION

In this work, we studied unconventional reservoirs, namely oil sands, heavy oils, and bitumens. These hydrocarbons represent the most abundant unconventional hydrocarbon resources worldwide. Due to their high viscosity, thermally enhanced oil recovery (EOR) methods are often required for their production, followed by extensive refining processes for their downstream distribution. Therefore, the breakeven price for their production is very high, which stimulates the major oil producers to constantly invest in developing new technologies to decrease their exploration and recovery prices. In petrophysical well-logging, this involves optimizing and developing new methods for the improved *in-situ* characterization of these hydrocarbons.

In light of the foregoing, I focused my research on advancing conventional and establishing new analytical and data-driven methods for *in-situ* characterization of heavy oil and bitumen resources, primarily from LF-NMR measurements. The heavy oil viscosity and reservoir fluid saturation distributions are two principal factors considered for the recovery of these resources. Therefore, this thesis aims to develop new workflows and models for predicting oil viscosity and water saturations from LF-NMR data.

1.1. Heavy oil and bitumen resources

Bitumens and heavy oils usually form deposits in shallower geological settings, where due to the cold temperatures and a lack of the caprocks, they are being subjected to bacterial biodegradation. The depth of these deposits usually does not exceeds 4 km, while the temperatures are most often lower than 80 °C¹. In such conditions, the biodegradation process spans over geological periods, where hydrocarbon-degrading bacteria use lighter oil fractions for the metabolic processes, which gradually reduces the viscosity, initial mass, and gas-to-oil ratio (GOR) of the oil.

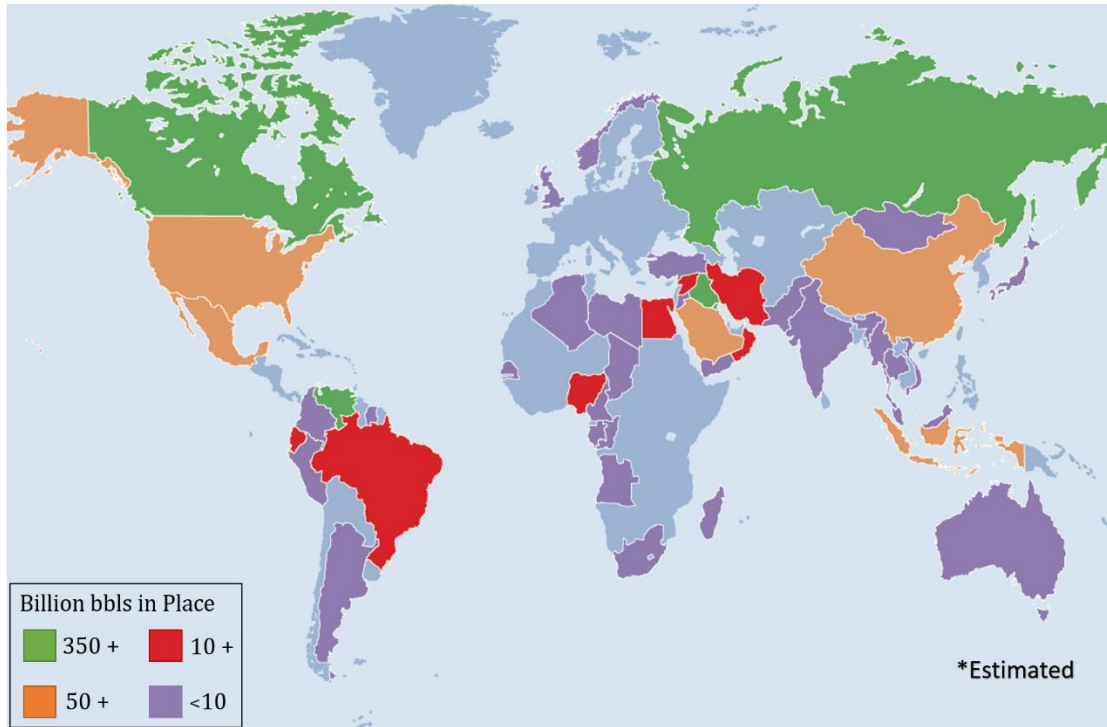


Figure 1: Estimated worldwide heavy oil reserves by Country (copyright Schlumberger).

Heavy oil and bitumen are among the most abundant hydrocarbon resources worldwide. They are principally different from other hydrocarbon resources because of their high viscosity, density, and increased concentration of heavy components such as asphaltenes, resins, and wax. Along with these components, heavy oils and bitumens often contain various heavy metals, sulfur, and nitrogen. Due to these **properties**, their economic value is considerably smaller **than** their lighter counterparts, as their recovery and refinement require **the application** of

costly technologies and time-demanding processing. However, their large in-place volumes and high oil market prices allow their profitable extraction and refinement, so major oil companies **are acquiring** licenses for their development as an alternative to ~~the diminishing~~ conventional oil resources. Studies estimate that heavy oil and bitumen reserves today amount to about 55% of the total world reserves (nearly 9 trillion barrels), while the most extensive deposits were found in Canada, Venezuela, and Russia¹.

1.2. Enhanced Oil Recovery (EOR) methods for heavy oil fields

As heavy oils and bitumens have distinctively high viscosities, their recovery from the reservoir rocks is often **aided** with the use of various thermal Enhanced Oil Recovery (EOR) techniques such as hot steam injection or in-situ combustion ~~via injection wells~~, where the heat exchange process reduces oil viscosity in the formation of interest². In heavy oil reservoirs, the viscosity may vary up to a hundred times in vertical and horizontal directions. Therefore information about the spatial distribution of viscosity can affect not only the well placement and injection or production rates but mathematical reservoir simulations as well³.

In literature, the EOR methods are generally divided into two groups: cold and thermal production. The cold production methods are economically and technically convenient recovery methods; however, they are limited to shallow deposits since they involve open-pit mining. Waterflooding was also occasionally used with some success, but in fields where oil viscosity was not more than 100 cP, the sweeping efficiency of the waterfront reduces with an increase of viscosity, most notably due to the viscous fingering¹. Vapor-assisted extraction (VAPEX) is another cold production method that proved efficient. It is based on miscible solvent vapor injection into parallel stacked horizontal wells. The miscible solvent vapor is injected into the upper well, where the vapor chamber is produced around the well. This causes the dilution of solvent vapor into surrounding heavy oil and bitumen, which leads to viscosity reduction and drainage of the

hydrocarbons to the extraction well beneath, with the help of gravity. This process is energy efficient since it is not based on thermal exchange and does not require the infrastructure for generating hot steam or water.

In thermal methods, on the other hand, increasing the temperature in the well or reservoir causes viscosity to decrease, thus improving the heavy oil and bitumen mobility. For instance, in the steam-assisted gravity drainage (SAGD), the placement of the injection and producer wells is almost identical to the one in the VAPEX method; however, instead of the solvent vapor, hot steam is injected. After the hydrodynamic linking between wells is obtained by the initial steam treatment of both wells, the injection of steam is continued in the injection well, where hydrocarbons are affected by the steam expansion. The temperatures of this treatment can reach over 200 °C, thus initiating a highly mobile gravitational drainage of bitumen and heavy oil towards the underlying producing well. In addition to the SAGD, cyclic steam stimulation (CSS) is used, where one well is used for injection and production. In this approach, hot steam is injected into the targeted formation and left to soak up and warm the oil, followed by the production cycle. Another well-known thermal method is in-situ combustion (ISC) or fireflooding, where an oxidizing gas (i.e., air) is injected into the formation, which causes the ignition of heavy oil in-situ. In this set-up, the portion of the hydrocarbon's heavier components is utilized as a fuel for further propagation of the combustion front, where exothermal reaction heats the surrounding rocks and thus lowers the oil viscosity. An added benefit of ISC is that the heavier components are being consumed due to the thermal cracking, which results in upgraded oil.

The success of the EOR project in heavy oil and bitumen deposits is primarily dictated by the initial viscosity of the hydrocarbons and their spatial distribution in the field. In steam EOR, for instance, monitoring of the steam front is often performed using periodic or permanent four-dimensional (4D) seismic surveying by collecting pressure and fluid saturation data within the field⁴. Monitoring based

on logging is also commonly implemented by fiber-optic distributed temperature sensing (DTS), which provides constant temperature monitoring along the wellbore. NMR measurements have been considered for monitoring the fluid saturation changes and wettability alteration assessing^{4,5}. If this technology is used for reservoir monitoring, NMR logging could also be used as a continuous, non-invasive **option** for monitoring viscosity variations and changes **due to** temperature changes in the reservoir.

1.3. NMR theory review

1.3.1. Spin-lattice (T_1) and spin-spin (T_2) relaxation

The hydrogen atom in its core contains a single proton with a positive charge (H^+). Protons have spins and they exhibit magnetic behavior, which is why the orientation of their spins can be **manipulated by introducing** an external magnetic field. For instance, if a magnetic field (B_0) is introduced to the H^+ proton-containing system, the proton spins will tend to align along the direction of the B_0 field ~~due to their magnetic field~~. The quantum theory posits that the protons will **reconfigure to a low or high energy state in such a setup**. ~~At this point~~, the difference between the number of protons in high and low energy states will generate the total or bulk magnetization M_0 that NMR tools can measure.

$$M_0 = N \frac{\gamma^2 \hbar^2 I(I+1)}{3(4\pi^2)kT} B_0 \quad (1)$$

where N is the number of protons **for** unit volume, I is the quantum spin number, T is the temperature in Kelvins, and k and \hbar are Boltzmann's and Planck's constants, respectively. The polarization of protons is not immediate but instead grows by a specific time constant. This constant is called spin-lattice or longitudinal relaxation time (T_1). As the polarization is exponential, and under the assumption that the polarization orientation transpires along the z-axis in 3D space, then:

$$M_z(t) = M_0 \left(1 - e^{-\frac{t}{T_1}}\right) \quad (2)$$

where t is polarization time, and $M_z(t)$ is polarization magnitude along the z-axis at the time t . The T_1 time represents a moment at which ~63% of the magnetization is reached. At three T_1 , about 95% of magnetization is reached.

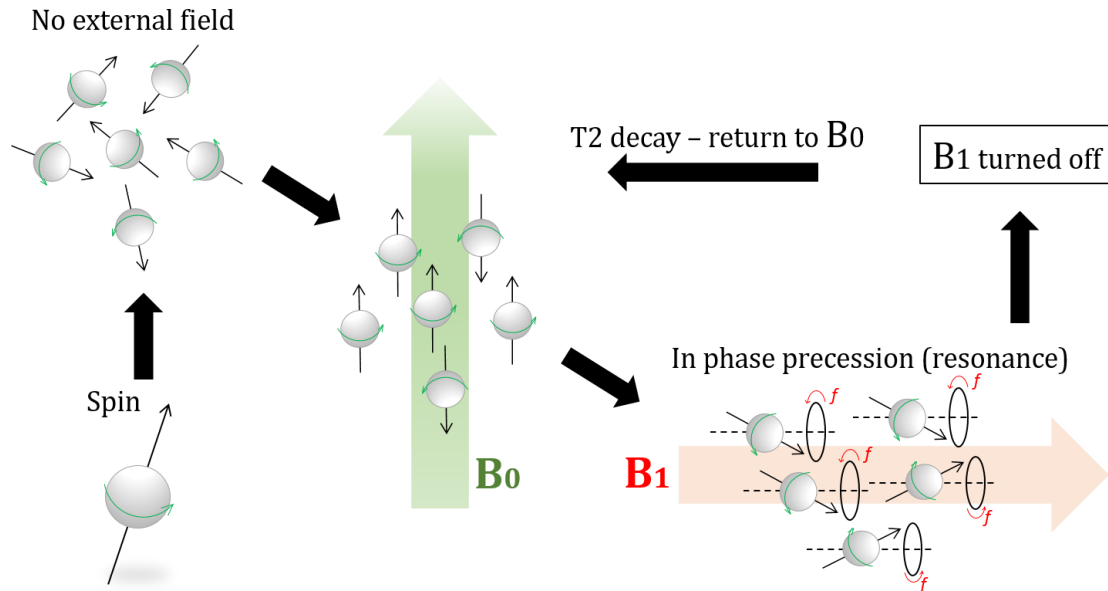


Figure 2: Flowchart of a typical NMR relaxation experiment, illustrating the proton polarization steps

For nuclear magnetic resonance to occur, it is necessary to perform the pulse tipping of the protons. While protons are polarized along the B_0 , an additional short radio-frequency oscillating pulse (B_1) is applied to the system. The B_1 Larmor frequency (f) must correspond to the Larmor frequency of the spins to achieve the resonance. The tipping angle (θ) is defined as:

$$\theta = \gamma B_1 \tau \quad (3)$$

where τ is the period when the B_1 is applied to the system. The tipping angle can be controlled by both B_1 and τ . In practice, the first B_1 pulse tips the proton system into the perpendicular plane (x, y) relative to the B_0 (z), thus $\theta = 90^\circ$. At this point, the protons are precessing about B_0 and are in phase, and the NMR device can detect their signal. However, immediately as the B_1 stops, the protons start to dephase due to the B_0 magnetic-field inhomogeneity and molecular tumbling,

usually at an exponential rate. Therefore their signal decays at the particular time constant (T_2^*) and is called free induction decay (FID).

Fortunately, the dephasing due to the B_0 inhomogeneity can be reversed by adding the B_1 pulse ($\theta = 180^\circ$) after τ time elapsed from the first pulse ($\theta = 90^\circ$). The second pulse flips the protons, which reverses the dephasing process, meaning that the protons will return to the same phase after τ time elapsed from the second pulse. This subsequent pulse is referred to as a 'refocusing pulse.' When spins return to the same phase, the 'spin echo' signal is produced. The NMR device can be configured to produce a series of refocusing pulses, thus generating a series of spin echoes. This series or sequence of refocusing 180° pulses is known as Carr-Purcell-Meiboom-Gill (CPMG) pulse sequence, while the recorded sequence of echoes is called the 'spin echo train' ⁶.

Although the CPMG pulse sequence can reverse the effect of B_0 field inhomogeneity, the dephasing of spins due to molecular tumbling and diffusion is irreversible. Since the dephasing is mainly due to the interaction of spins, the decay of magnetization in the horizontal plane (x, y) is called spin-spin relaxation, or transversal relaxation, as the magnetization is in the transversal plane relative to B_0 . The time constant associated with the decay rate is T_2 relaxation time.

$$M_{xy}(t) = M_{xy}(0) e^{\frac{-t}{T_2}} \quad (4)$$

where $M_{xy}(0)$ is magnetization magnitude at $t=0$. According to Equation 4, for one T_2 time constant, the magnitude of M_{xy} will drop to $\sim 37\%$ of its initial value, and after three T_2 constants to $\sim 5\%$.

1.3.2. CPMG pulse sequence configuration and NMR data processing

The CPMG pulse sequence consists of a 90° initial pulse that tips the polarized protons into an x, y-plane, followed by several refocusing 180° pulses. The quality of the obtained CPMG decay data, its inversion, and the quality of interpretation

are strongly dependent on the configuration of the CPMG sequence. The principal controlling parameters include the time between two pulses or echo spacing (TE), the polarization time or waiting time (TW), the number of pulses or the number of echoes (NE), and a number of CPMG trains (NT).

Setting an NMR tool to a short echo spacing (TE) will influence the signal-to-noise ratio (SNR) twofold; first, the density of spin-echoes within a train will increase; second, the echoes will be recorded earlier. Consequently, this leads to increased SNR. However, the experiment time will increase proportionally when the number of echoes (NE) increases. The same is true for the number of trains (NT) and waiting time (TW). It should be noted that TW should be configured according to the sample or reservoir interval and the purpose. If the goal is to characterize heavy oil and clay-bound water, the TW can be decreased since protons will return to equilibrium much faster than pure water or light oil.

The representation of the decaying echo train in the time domain is usually obtained by the Laplace Inverse Transform (ILT)⁷, whereas the output of a T_2 distribution is obtained (Figure 3). However, the inversion of the NMR signal represents the ill-posed problem since minor perturbations (i.e., noise) in the measurements can substantially impact the T_2 distribution form, that is, the stability of the solution. A stable and unique solution can be obtained if the inversion of the signal is performed numerically. In such a case, the representation of the T_2 decay is achieved from echo-fitting. To simplify the echo-fitting, the discretization of the T_2 decay signal can be performed, where the predefined number of discrete T_{2i} relaxation times correspond to individual exponential decay. Then, the set of echo trains can be expressed as a system of linear equations where each equation corresponds to an individual echo train. Since the fitting is performed to the sum of multi-exponentials, the stable solution can be obtained by solving non-negative least squares. The standard approaches are Lawson-Hanson and Tikhonov regularization⁸.

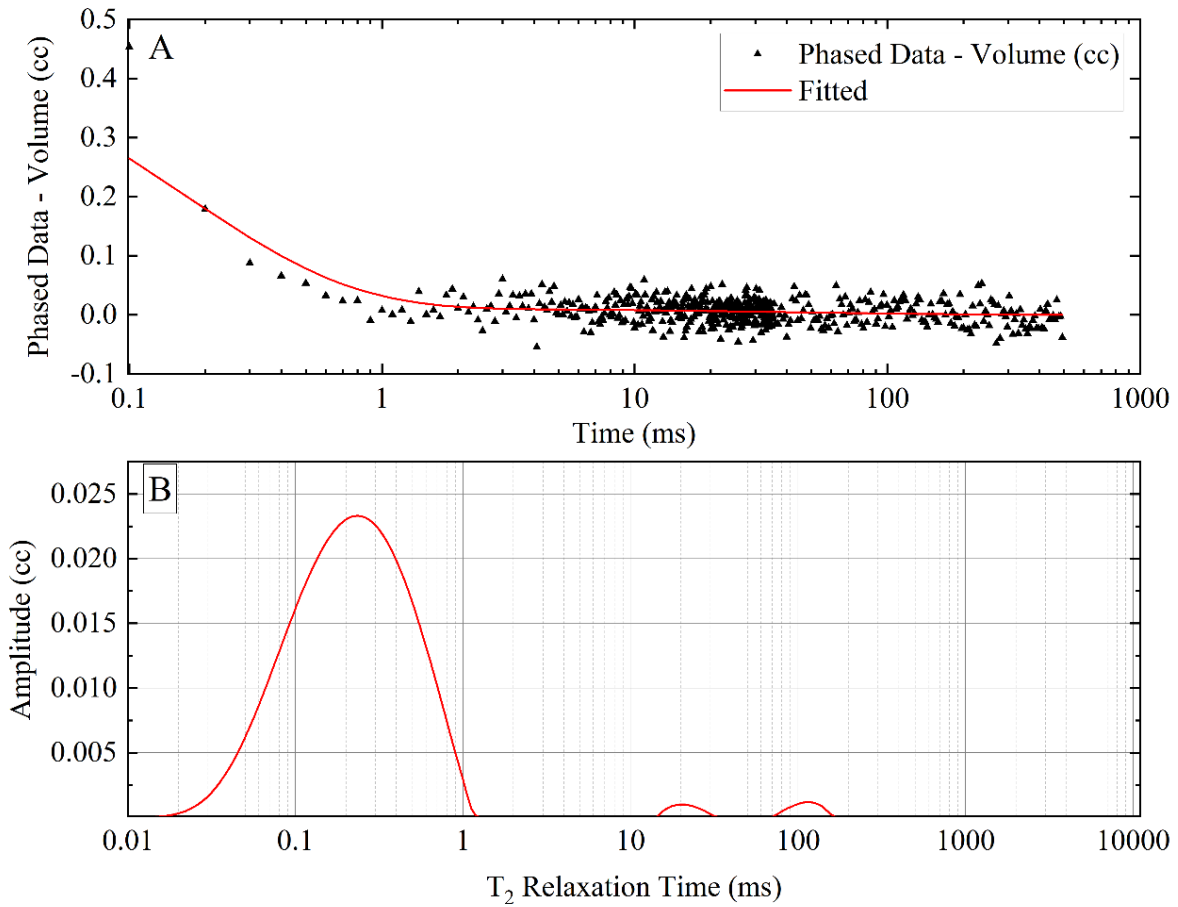


Figure 3: (A) Raw T_2 NMR relaxation data and (B) T_2 data after inversion.

1.4. Oil viscosity

1.4.1. Conventional measurements of oil viscosity

Viscosity is a physical fluid property that reflects the amount of internal friction of a fluid. In other words, the fluid's viscosity indicates the magnitude of the resistance to flow. Mobilizing a fluid requires a certain amount of force, and the rate of change of the induced deformation can be measured in function of time. In fluids, since viscous forces control the flow velocity, we measure the flow velocity for the applied amount of force or applied amount of shear stress. Bryan et al., in

their work, link the relationship of rheological viscosity measurements with Eyring's theory of viscosity, which is also consistent with the Bloembergen-Purcell-Pound (BPP) NMR relaxation model^{9,10}. As the fluid undergoes shearing deformation, the rate of shearing is proportional to the applied shear stress (Equation 5).

$$\tau = \eta \cdot \gamma \quad (5)$$

where τ presents the amount of shear stress applied to the fluid, γ stands for the shear rate, and η is the fluid viscosity. Based on the Eyring's theory of viscosity, molecules in fluids are structured in lattices, while intermolecular space remains vacant but not spacious enough for other molecules to shift through readily¹¹. However, if force is applied, the molecules will reconfigure their positions until the vacant space becomes large enough for another molecule to enter. Eyring proposed an analytical model for such behavior (Equation 6)

$$\eta = \left(\frac{\delta}{a}\right)^2 \cdot \frac{N\hbar}{V} \cdot e^{\left(\frac{\Delta G_0}{RT}\right)} \quad (6)$$

where δ is intermolecular layer distance, a is the distance between the vacant space and a molecule, N is Avogadro's number, \hbar is Planck's constant, and V is a fluid's molar volume G_0 activation energy, R is the universal gas constant, and T is the absolute temperature. Heavy oils and bitumens comprise long chain-like molecules, cyclic paraffin, and branches originating from heavy components. To mobilize these molecules, more activation energy is required (G_0) since the attractive forces of surrounding molecules hinder the movement of molecules attempting to occupy the vacant space. In rheological measurements of heavy oils by a cone and plate viscometers, this resistance to flow translates to high shear stresses. From Equation 6, it can be observed that the temperature is in an exponential relationship with viscosity. If we apply heat to the heavy oil, the heavy components will gain more energy while the intermolecular distance will increase, enabling the less restricted motion of molecules. This is also consistent with rheological viscosity measurements, where for fixed shear stress and with an increase in temperature, we observe increased shear rate⁹. Equation 6 can be rewritten by substituting constants (Equation 7).

$$\eta = A \cdot e^{\left(\frac{E_a}{RT}\right)} \quad (7)$$

where A is a constant and E_a is the viscous free energy of activation. In this Arrhenius-type equation, the values of E_a and A vary for different oil samples. These variations are associated with molecular weight variation of different oil components and their chemical composition and structure¹¹. These variations may be substantial since the composition of heavy oils and bitumen, in particular, can be significantly different, indicating that derivation of the general viscosity model is challenging.

1.4.2. Oil viscosity by LF-NMR measurements

The oil is a blend of a diverse range of liquid hydrocarbons with inconsistent molecular structure¹². When it has a higher proportion of complex high molecular weight compounds such as asphaltenes and resins, oil viscosity will be higher, signifying that viscosity reflects oil's chemical complexity¹³. This natural inconsistency of oil compositions elicits a constant demand for the development of new techniques for their efficient characterization. In recent years, the wave of innovation has led to the application of **low-field nuclear magnetic resonance** (LF-NMR) tools to characterize hydrogen-bearing liquids due to their ability to rapidly convey a series of contactless, non-invasive experiments.

To relate a fluid viscosity to NMR relaxation, it is necessary to comprehend and model the molecular interactions. Equations 6 and 7 demonstrate that viscosity can be expressed without macroscopic flow or shearing (Equation 5). These findings are consistent with a theoretical Bloembergen-Purcell-Pound (BPP) NMR relaxation model, associated Debye-Einstein-Stokes spherical molecules model, which anticipates different rotational correlation times (τ_c) for various molecule sizes^{10,14,15}. The correlation time represents the mean time required for a molecule to rotate one radian and is a crucial parameter for determining microviscosity. It is also a fundamental component of the BPP relaxation model.

Based on this relationship, one can study the association of T_2 relaxation with viscosity¹⁵. The physics of this relationship is discussed in section 2.2 (Fuel).

As previously mentioned, the T_2 relaxation distribution after mathematical inversion can be represented in a time domain. Distributions of T_2 relaxation between different fluids can be compared using a mean T_2 distribution time, such as T_2 logarithmic mean (T_{2lm}). When fluids are measured in a bulk state, the primary relaxation mechanism will be bulk relaxation (T_{2B}), which is due to the energy exchange of the H spins and diffusion. Straley et al.¹⁶ and Coates et al.⁶ have experimentally shown that the T_{2B} is proportional to the ratio of temperature (T) and viscosity (η):

$$T_{2B} \propto \frac{\eta}{T} \quad (8)$$

Since the relaxation times of light oils, water and gases have long relaxation times, and high viscosity fluids such as heavy oils have short relaxation times, the T_{2B} and T_{2lm} can be correlated with oil viscosity¹⁶⁻¹⁸. Although this relationship is used as a foundation for nearly all existing NMR viscosity models, it only works well for the light and medium viscous oils ($\sim 1-800$ cP)¹⁹ composed of lighter hydrocarbons with a more straightforward chemical structure. In the case of heavier oils, the T_2 relaxation deviates from the classical BPP model, and the relationship described in Equation 8 alters significantly.

In the past 30 years, many analytical NMR viscosity models have been proposed for characterizing crude oil. However, the reports in the literature show inconsistency in the prediction accuracy of these models due to three main reasons: use of the light oil NMR models for the prediction of heavy oil viscosity^{16,18,20,21}, use of models (including heavy oil models) without prior tuning for a given reservoir or a suit of oils, and due to use of ambiguous mathematical procedures for model tuning^{3,22,23}. Moreover, there were several attempts to develop a “universal model” for *in-situ* heavy oil viscosity prediction aiming to estimate viscosity in the formations with weak prior knowledge about the oil properties²²⁻²⁵. These models have default parameters derived for heavy oils from

a particular oil field. However, when applied to different heavy oils, they generate significant prediction errors, in some cases over a factor of three²³. To develop a universal analytical NMR viscosity model for systems with oils of various compositions would be contradictory to Debye-Stokes-Einstein's findings stating that different correlation times are expected for different molecule sizes¹⁴. However, for the order-of-magnitude *in-situ* viscosity evaluation, existing analytical models can be improved to a degree where more reliable estimates can be utilized to optimize the decision-making process for viscosity variation monitoring during EOR projects.

Although LF-NMR technology has been proved to be a viable tool for observing differences in variable viscosity oils, numerous constraints arise as a consequence of not only the embedded chemical complexity of oils and limitations of LF-NMR devices but also from analytical tools and models used for the interpretation of experimental results^{23,25-29}. Since the former two are technologically challenging to change, one can attempt to improve the analytical tools and frameworks using new mathematical approaches. In such circumstances, the supervised learning (SL) methods have been proven helpful in developing more reliable mathematical models in many relevant fields such as fuel processing, petrophysical studies of porous mediums, and oil viscosity monitoring equipment in mechanical systems³⁰⁻³⁴.

1.4.3. LF-NMR oil viscosity measurements in other industrial fields

The application of NMR viscosity models is not relevant only for petrophysical well-logging. One potential application is in fuel processing, where there has been a surge for the last few years in the development of fast methods for the characterization of petroleum fractions by LF-NMR³⁵. Among many studied physicochemical properties, the oil viscosity showed to be of the principal importance in determining the rate of interaction with fuel during combustion processes in internal combustion engines^{36,37}. In these studies, the LF-NMR predictive models were typically derived using multivariate calibration with

partial least squares (PLS) regression or artificial neural networks (ANN), which proved efficient. However, in nonlinear datasets, the reports in the literature show that PLS did not provide satisfactory accuracy, whereas ANN tended to overfit the data, thus leading to poor model generalization^{38,39}. In the LF-NMR examination of petroleum fractions, this nonlinearity can occur due to their chemical intricacy, leading to the degradation of model forecasting performance¹⁴.

Moreover, in mechanical systems (tribosystems), viscosity reflects the oil's capacity to render the sufficient thickness of the lubricating film between the surfaces exposed to friction. In order to efficiently buffer the rate of machinery wear, the oil selection is made under the speed-load and temperature conditions of the system⁴⁰. The prevention of malfunctions in tribosystems is usually performed by monitoring oil viscosity, where its relative increase may indicate excessive oxidation or contamination of the oil by other fluids. In contrast, its decrease may indicate the beginning of a thermal cracking process, occurring at high temperatures⁴¹. In earlier studies, LF-NMR measurements were proposed as an alternative to conventional monitoring approaches that involve direct-contact instruments based on vibration, acoustic, and micro-displacement methods³⁴. In circumstances where these instruments **would be** difficult to utilize, the LF-NMR tools **could** be used instead for non-invasive, real-time viscosity monitoring^{34,42,43}. As the operating conditions of these systems may lead to significant oil viscosity fluctuations, the robust data-driven or analytical NMR model could be used to measure the fluctuations accurately and, in that manner, help in the early detection of equipment failure.

1.5. Water saturation

1.5.1. Water saturation by resistivity measurements

One of the primary purposes of petrophysical formation evaluation is the quantification of hydrocarbon and water saturations. To properly evaluate the volume of hydrocarbons, it is necessary to determine water saturation beforehand, since generally:

$$S_o = 1 - S_w \quad (9)$$

where S_o is oil saturation and S_w is water saturation. Conventionally, in well-logging practice, the water saturation was determined using resistivity logs, and depending on the reservoir type, resistivity data would be used in combination with other standard logs such as density (RHOB) and neutron (NPHI)⁴⁴. The widely used empirical model for water saturation estimation in conventional hydrocarbon reservoirs was developed by Archie⁴⁵ (Equation 12).

$$F = \frac{R_o}{R_w} = \frac{a}{\varphi^m} \quad (10)$$

$$I_r = \frac{R_t}{R_o} = \frac{1}{S_w^n} \quad (11)$$

$$S_w = \sqrt[n]{\frac{aR_w}{R_t\varphi^m}} \quad (12)$$

In Equation 10, F is a formation factor, R_o fully water-saturated rock resistivity, R_w brine resistivity, φ fractional porosity, a and m are tortuosity coefficient and cementation exponent, respectively. In Equation 11, I_r is the resistivity index, R_t is rock resistivity, R_o is the resistivity of fully water-saturated rock resistivity, S_w is fractional water saturation of the formation, and n is the saturation exponent. Finally, Equation 12 presents Archie's water saturation model, which is obtained by combining Equations 10 and 11. The m , n , and a are known as rock resistivity parameters. While most of the parameters can be obtained from conventional logs, particular attention is required to calibrate a , m , n . These are obtained from laboratory-controlled resistivity tests and subsequent least squares regression.

Archie's equation was successfully used in systems with simple, uniform pore space saturated by water⁴⁶. However, issues arise in reservoirs with large amounts of clay-bound and capillary-bound water, strong variations of salinity with depth, and formations containing clays or conductive minerals such as pyrite^{47,48}. This is also true for Canadian oil-sands⁴⁹. In these terms, the presence of bound water and clays will cause the underprediction of OOIP, while the variable salinity can cause either overprediction or underprediction of OOIP.

1.5.2. Water saturation by LF-NMR measurements – T₂ cutoff approach

Since the LF-NMR tools measure the response H⁺ protons of the fluids and are therefore lithology-independent, most of the issues relevant for resistivity logging can be avoided. Another advantage of NMR measurements is differentiating between irreducible water saturation (capillary and clay bound water) and producible fluids. The primary assumption is that larger pores are saturated by producible fluids, where the flow can occur in the presence of pressure gradient, while smaller pores contain fluids trapped by capillary forces or are bound within the lattice of clay minerals. If this condition is true, a T₂ cutoff value (location in T₂ distribution) can be defined, separating the T₂ distribution to signals corresponding to clay-bound, capillary-bound and producible fluids. Integration of the separated regions can be performed and related to producible and bound fluid volumes. Straley et al.,¹⁶ were the first to empirically identify the universal T₂ cutoffs for clay-bound water at 3 ms in conventional sandstones. The cutoff value was determined by comparing the clay-bound water calculated from the ratio of cation exchange capacity and pore volume (Q_{ve}) with cumulative T₂ distribution porosity, using sandstone core plugs from 45 American and European oilfields. To evaluate the producible porosity or free-fluid index (FFI), it was necessary to perform NMR T₂ measurements on core samples in two states – cleaned and fully saturated state (S_w 100%) and after centrifuging to the

irreducible water saturation (S_{wirr}). These experiments were performed for the suit of 86 sandstone samples, and the universal cutoff was found to be at 33 ms¹⁶.

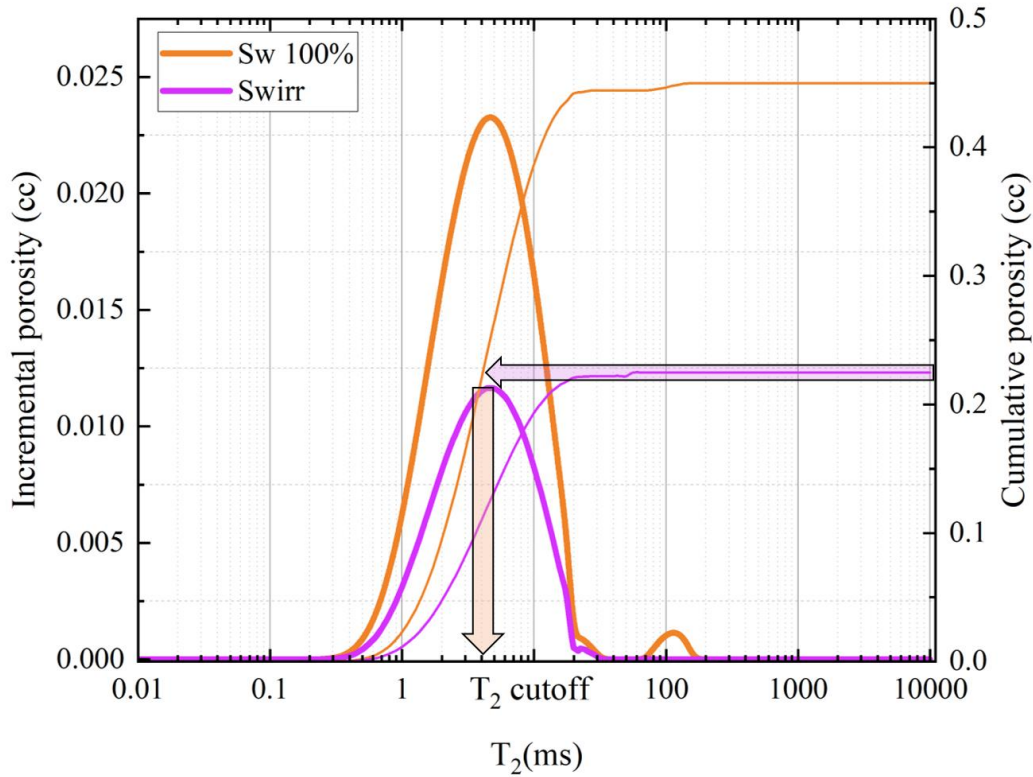


Figure 4: An example of the determination of T_2 cutoff value by LF-NMR and centrifuge. The orange curve presents the T_2 distribution of the 100% water-saturated sample (S_w 100%). The purple curve presents the T_2 distribution of the sample centrifuged to irreducible water saturation (S_{wirr}).

Although these universal cutoff times work for conventional sandstone reservoirs where porosity and permeability are generally uniform, many recent studies have shown that T_2 cutoff values vary dramatically for reservoirs of other lithologies, such as shales, carbonates, oil-sands, coals, and tight sandstones⁵⁰. In addition, even if the T_2 cutoffs are determined experimentally, it is not recommended to use a single T_2 cutoff for the same well or oilfield since the T_2 distribution can vary drastically both vertically along the well and laterally, which would potentially cause erroneous estimation of OOIP⁵⁰.

1.5.3. Water saturation by LF-NMR measurements in oil-sands

The T_2 cutoff determination for oil-sands is even more problematic for two principal reasons. First, the centrifuging to S_{wirr} cannot be adequately performed since the sand exposed to severe centrifugal forces will lose its structure and native filtration properties, rendering the subsequent NMR experiments inadequate. Second, the heavy oil and bitumen in oil-sands have a fast T_2 relaxation time and produce a signal in the same region as capillary and clay-bound water, causing a significant overlap. One of the well-known approaches that had considerable success in addressing this effect is based on NMR spin-spin relaxation (T_2) distribution peak deconvolution⁵¹. The concept behind this approach is that viscous bitumen relaxes faster in an NMR distribution than surface-bound water, so early T_2 signals are attributed to bitumen, and later T_2 signals correspond to the water saturation in the rock.

Assuming that the oil-sands are largely water-wet, water will generally be found in the corners of connected sand grains and potentially as a thin film over the grain surface. The principal relaxation mechanism of hydrogen protons in high viscosity oils and bitumen would be bulk relaxation, while water would strongly influence surface relaxation, with bulk relaxation playing a minor role in the water T_2 values. Bulk relaxation and surface relaxation times of water and oils are unique for the most part, that is, the oil molecules generally relax quicker relative to the water molecules. When the NMR T_2 distribution contains discrete oil and water responses (Figure 5A), a simple cutoff method can be applied to separate their amplitudes and quantify their volumes.

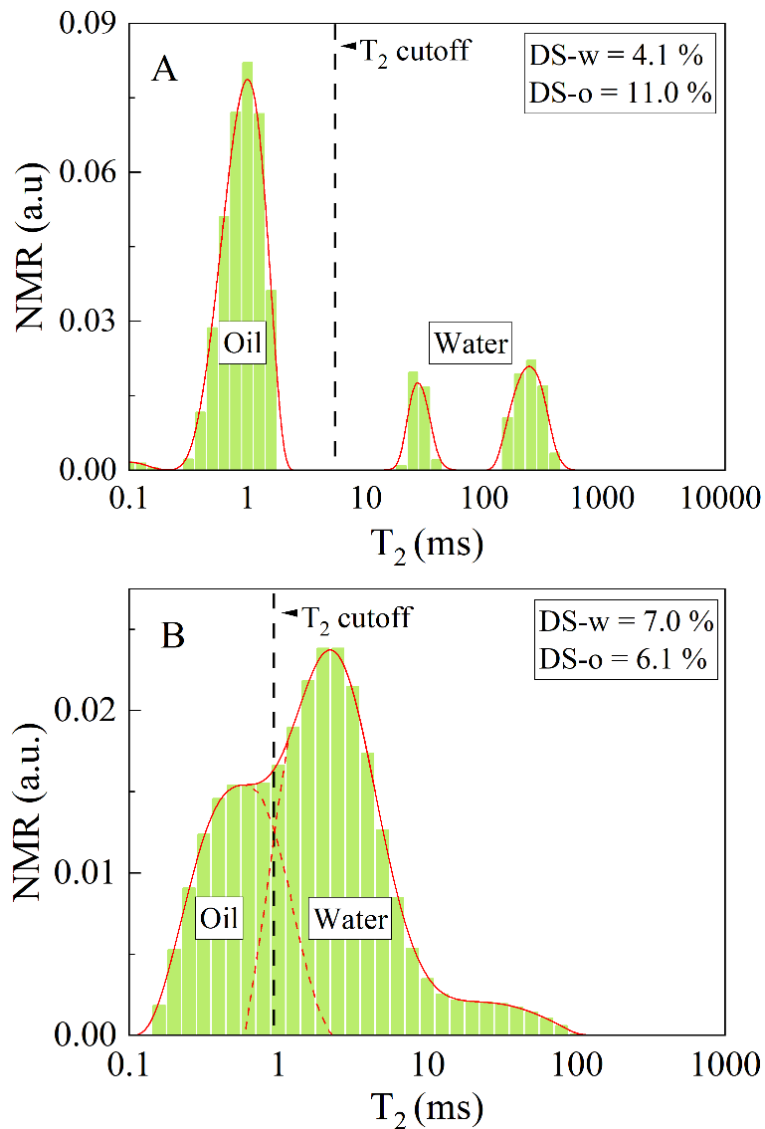


Figure 5: Representative NMR T_2 distributions of two oil-sand samples. (A) An example of distinct oil and water signals where a simple cutoff method can be used for oil-water separation. (B) An example of NMR T_2 distribution with overlapped oil and water signals where deconvolution with T_2 cutoff cannot provide a satisfactory solution. Black vertical dashed lines present potential cutoff times. DS-w and DS-o are percentages of water and oil by Dean-Stark, respectively, relative to solids.

However, in fines and clays, where pores are tiny, the water protons relax faster due to the surface relaxation at the water-rock interface, thus generating the signal in the fast-relaxing part of distribution where it can overlap with the signal originating from heavy oil and bitumen (Figure 5B). In addition to that, the

diffusion coupling effect may further decrease the interpretability of the oil and water signals. This effect occurs in saturated and connected micro- and macropores when water is in diffusional exchange, causing the change in the relationship between T_2 relaxation and pore size distribution⁵². In strong diffusive-coupling conditions, macro- and micropore water signals will merge into a single peak, rendering the single T_2 cutoff and deconvolution approach inaccurate⁵³. Another limitation of this approach is that it requires the separate determination of water and oil NMR amplitudes and the independent measurement of their volume or mass.

Alternative methods involve 2D LF-NMR measurements, where instead of using one NMR parameter (i.e., T_2 relaxation), additional parameters are employed (i.e., T_1 relaxation or diffusion) to obtain so-called 2D NMR maps⁵⁴⁻⁵⁶, which can theoretically help to separate these overlapping bitumen and water signals. Application of 2D maps showed considerable success in fluid saturation evaluation, compared to 1D T_2 relaxation distribution analysis, since T_1 relaxation or diffusion of reservoir fluids can be sufficiently different, thus enabling relatively simple separation of their signals. Unfortunately, 2D NMR is slower and more expensive to run, and there can still be instances where these signals are not distinct, in which case estimation of fluid types and fluid volumes can be challenging and require advanced analysis involving blind-source signal separation (BSS), clustering algorithms, and a certain degree of knowledge in 2D NMR maps interpretation⁵⁷.

Chapter 2 Heavy oil viscosity prediction at high temperatures by low-field NMR relaxometry and nonlinear least squares

2.1. MOTIVATION

Evaluation of crude oil viscosity from LF-NMR data has been proven to be a viable alternative to laborious and time-consuming conventional measurements, which require sample recovery. However, this work shows that for most heavy oil correlations^{10,12-14}, accuracy degrades dramatically with the temperature increase, making them unreliable for continuous viscosity monitoring in oil wells. Another problem commonly seen in practice is vertical and horizontal anisotropy of the oil viscosity within the same heavy oil reservoir and sometimes the same well^{3,59}, meaning that the NMR model must be robust enough to provide satisfactory predictions within the group of the chemically different heavy oil samples.

In this work, we derived a new enhanced NMR viscosity model and three key improvements:

- Enhanced prediction of viscosity for the suite of 23 heavy oils with varied dynamical viscosities (70–21,600 cP) in comparison to the models published in literature;
- Enhanced prediction of viscosity for the JC bitumen sample at elevated and high temperatures (26-200 °C), for viscosity range from 10 to 170,000 cP, in comparison to the models published in literature;
- Nonlinear least squares regression procedure for obtaining the optimal fitting parameters of NMR viscosity models (tuning);

Two separate datasets of dynamic viscosities (η) were created from rheological experiments: viscosities of 23 various heavy oil samples at 30 °C and 50 °C, and viscosities of a single bitumen oil sample at high temperatures (26-200 °C). For NMR viscosity prediction, spin-spin relaxation time (T_2 -relaxation) and relative

hydrogen index (RHI_v) were used as model inputs, while model tuning was achieved by NLS regression. To quantify the effect of tuning on the reduction of prediction errors for our dataset, we evaluated the performance of models both in their default form (reported fitting parameters) and after tuning by NLS regression.

Model performance was evaluated using root mean square error (RMSE), maximum absolute error (MaAE), and adjusted coefficient of determination (adj. R^2).

2.2. THEORY AND EXPERIMENTS

2.2.1. SPIN-SPIN RELAXATION (T_2)

As mentioned before, the oil reservoirs contain various fluids rich in hydrogen. Modern LF-NMR logging devices measure the response of H^+ protons in fluids and provide information about petrophysical properties of rocks and physiochemical properties of fluids *in-situ*. In the case of hydrocarbons, the rate of T_2 relaxation shows a strong correlation with viscosity, which is why T_2 relaxation was used as a theoretical foundation for nearly all NMR viscosity models. Since the relaxation times of oil vary significantly with its chemical composition and temperature, a T_2 logarithmic mean relaxation time is calculated to characterize the whole NMR spectra:

$$T_{2lm} = \text{Exp} \left[\sum \frac{A_i}{A} \cdot \ln(T_{2i}) \right] \quad (13)$$

where A_i is the amplitude from i -th corresponding T_{2i} response.

2.2.2. MOLECULAR SIZE AND INTRAMOLECULAR DISTANCE

Dynamic viscosity correlation with T_2 -relaxation time can be inferred from Bloembergen-Purcell-Pound's (BPP) model, describing T_1 and T_2 -relaxation rate dependency with dipole-dipolar interaction¹⁰:

$$\frac{1}{T_1} = C \left(\frac{\tau_c}{1+\omega_0^2 \tau_c^2} + \frac{4\tau_c}{1+4\omega_0^2 \tau_c^2} \right) \quad (14)$$

$$\frac{1}{T_2} = C \left(\frac{3}{2} \tau_c + \frac{5}{2} \frac{\tau_c}{1+\omega_0^2 \tau_c^2} + \frac{4\tau_c}{1+4\omega_0^2 \tau_c^2} \right) \quad (15)$$

$$C = \frac{3}{10} \left(\frac{\mu_0}{4\pi} \right) \frac{\hbar^2 \gamma^4}{b^6} \quad (16)$$

where τ_c is molecule correlation time, and ω_0 is a Larmor frequency. The parameter C is defined for the $\frac{1}{2}$ spin by gyromagnetic ratio γ , the magnetic permittivity of space μ_0 , reduced Planck constant \hbar , and interproton distance b . Molecular collisions lead to a time-dependent change in molecule orientation and interproton distances. From Equation 14 and Equation 15, it is evident that proton relaxation rates are primarily influenced by correlation time for the liquid substance. Random change of the molecule orientation can be described by a rotational diffusivity D_r , a function of viscosity, temperature, and molecular size. By employing a Debye-Stokes-Einstein (DSE) model for spherical molecules, we can express τ_c as

$$\tau_c = \frac{1}{6D_r} \quad (17)$$

$$D_r = \frac{kT}{8\pi\eta a^3} \quad (18)$$

where k is Boltzmann constant, a is the radius of the spherical particle and η is the dynamic viscosity of the medium. The BPP model and DSE equations were developed for pure homogeneous substances, while crude and heavy oils have a complex chemical composition and molecular structures that contain multiple bonds, chains, solid asphaltene agglomerates, and clusters. Consequently, we can anticipate variability in molecule sizes and interproton distances, which causes fluctuation of parameters a and b (Equation 17 and Equation 18). In that sense, for

any universal NMR viscosity model, discrepancies in predictive ability will grow with the complexity of the chemical composition^{14,24,61}.

2.2.3. T₂-RELAXATION MECHANISMS IN HEAVY OILS

The total T₂ relaxation represents the sum of three relaxation components⁶²:

$$\frac{1}{T_2} = \frac{1}{T_{2\text{bulk}}} + \frac{1}{T_{2\text{surface}}} + \frac{1}{T_{2\text{diffusion}}} \quad (19)$$

The total spin-spin relaxation is governed by a sum of bulk relaxation T_{2bulk}, relaxation influenced by the pore surface T_{2surface}, and relaxation caused by the gradients of magnetic field T_{2diffusion} (Equation 19). In the case of water-wet porous media, surface relaxation is dominant for water and bulk relaxation for oil.

According to the nuclear spin relaxation theory⁶³, there are two extreme cases:

1. Fast motion or extreme narrowing case ($\omega\tau_c \ll 1$), characteristic for small molecules, low viscosities, or high temperatures. In such cases, $T_1 \approx T_2$ (Figure 6).
2. Slow-motion case ($\omega\tau_c \gg 1$) characteristic for relaxation of large molecules in high viscous substances or at low temperatures.

In both cases, the $1/T_2$ relaxation rate is proportional to correlation time τ_c and η/T ratio, respectively.

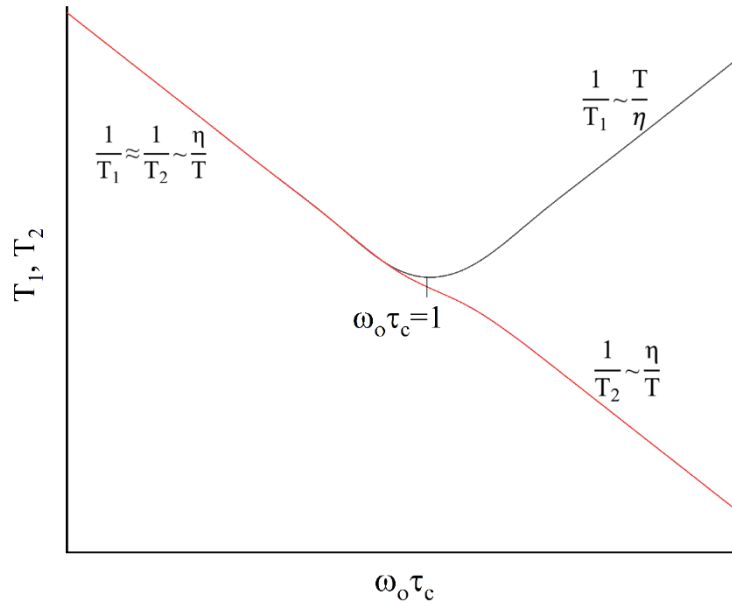


Figure 6: T_1 (black) and T_2 (red) dependence on correlation time (τ_c), according to the BPP relaxation model.

Equations 14-18 describe the molecular relaxation processes governed by a single exponential function which explains why the viscosity models for short correlation times (Figure 6) provide sufficiently accurate predictions for light oils^{16,18,20,21}. Solid-like components in heavy oils induce various relaxation rates as opposed to light oils, and the cumulative spin-echo decay can exhibit non-exponential behavior. This behavior can be approximated by the stretched-exponential function, also known as the Kohlrausch-Williams-Watts function²⁵:

$$G(\tau) = \langle F(0)^2 \rangle e^{-(\tau/\tau_c)^\beta} \quad (20)$$

Function $F(t)$ is a time-dependent function of molecule location and orientation. $G(\tau)$ describes a relationship between function $F(t)$ in different time steps, and β is a stretch parameter ($0 \leq \beta \leq 1$). Equation 20 does not have an analytical Fourier transform, but a modified Cole-Davidson function approach^{23,64} can be used instead.

$$T_{2\text{bulk}} \sim \left(\frac{\eta}{T}\right)^{-\beta} \quad (21)$$

This approach was confirmed to be effective by several authors^{3,15,23,65}. It should be noted that T_1 dependence on correlation time also does not follow the classical

BPP model for heavy oils and bitumen in the slow-motion case, which was experimentally proved by several authors^{15,66}, most recently by Singer et al.²⁷. In Figure 6, instead of the anticipated increase, T_1 plateaus to 3 ms value on a frequency-normalized scale for various viscosity samples. The authors explained the observed plateau effect by combining the dipole-dipole interaction model for intramolecular interactions and the modified Lipari-Szabo model for internal motions of the non-rigid structure.

2.2.4. ECHO SPACING (TE) AND RELATIVE HYDROGEN INDEX (RHI_v)

Due to the presence of solid-like components in heavy oils (e.g., paraffin and asphaltene), the T_2 -relaxation is often so fast that many LF-NMR logging tools cannot measure the whole relaxation spectrum the sample^{15,23,67}. The parameter that expresses the NMR tool's signal sampling rate is known as echo spacing (TE), where $TE \geq 0.1$ ms. Consequently, for heavy oils with a very short mean T_2 relaxation time (T_{2lm}), the logging devices cannot capture the fast-relaxing part of the NMR T_2 distribution. The result is that the tools fail to accurately reflect the actual number of hydrogen atoms (HI) and output HI that is too small since part of the fast-relaxing signal is not measured. Many authors tried to address this issue by adjusting TE using correction coefficient or integrating some form of hydrogen index into the model^{9,22,23,58,67,68}.

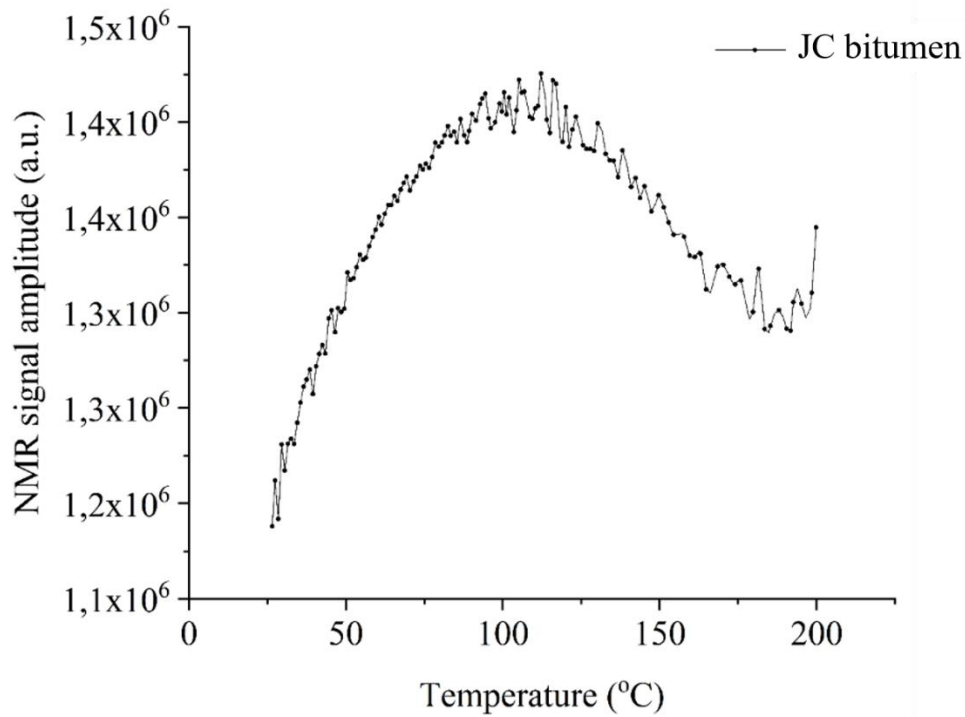


Figure 7: NMR signal amplitude of a single bitumen sample (JC bitumen) in the function of the temperature. The slope of the NMR signal decreases with temperature rise, and approximately at >100 °C, the slope becomes negative due to the Curie effect.

From Figure 8, it can be observed that at temperatures above 100 °C, the NMR amplitude decreases. This is known as a Curie effect^{66,69}, where magnetization loss (NMR signal loss) occurs due to the high temperature of the sample. One of the means to account for this loss is the implementation of the relative hydrogen index (RHI). The RHI represents the relative amount of measured (detectable) hydrogen protons by the NMR device in the oil sample. It is expressed as the ratio of oil and water NMR signal amplitudes per unit mass⁷⁰. In the case of using NMR tools at elevated temperatures, it is compulsory to implement temperature correction for RHI to account for the magnetization loss:

$$RHI = \left(\frac{A_o m_w}{A_w m_o} \right) \left(\frac{T(^{\circ}K)}{T_{amb}(^{\circ}K)} \right) \quad (22)$$

where A_o and A_w are the amplitudes of the oil and water signal respectively, m_o and m_w are masses of oil and water respectively, T and T_{amb} are the temperatures of the oil sample in kelvins. If the RHI is normalized to the sample volume, a relative

hydrogen index for a defined sample volume can be obtained (RHI_v). This normalization is consistent with Curie's expression for magnetic susceptibility, where magnetization is expressed per unit volume⁶⁸. Also, RHI_v is more suitable for application to well logs because the NMR tool detects a defined volume of the formation⁵⁸. Burcaw et al. proposed a simple approach for conversion of RHI to HI⁵⁸:

$$RHI_v = \frac{\rho_o}{\rho_w} \cdot RHI \quad (23)$$

where ρ_o and ρ_w are densities of oil and water, respectively. Note that Equation 23 should be valid under the assumption that sample volume change due to the temperature change is negligible. It should also be noted that Equation 23 represents the hydrogen proton response detected by the NMR device, and it is not to be associated with a true HI of the sample.

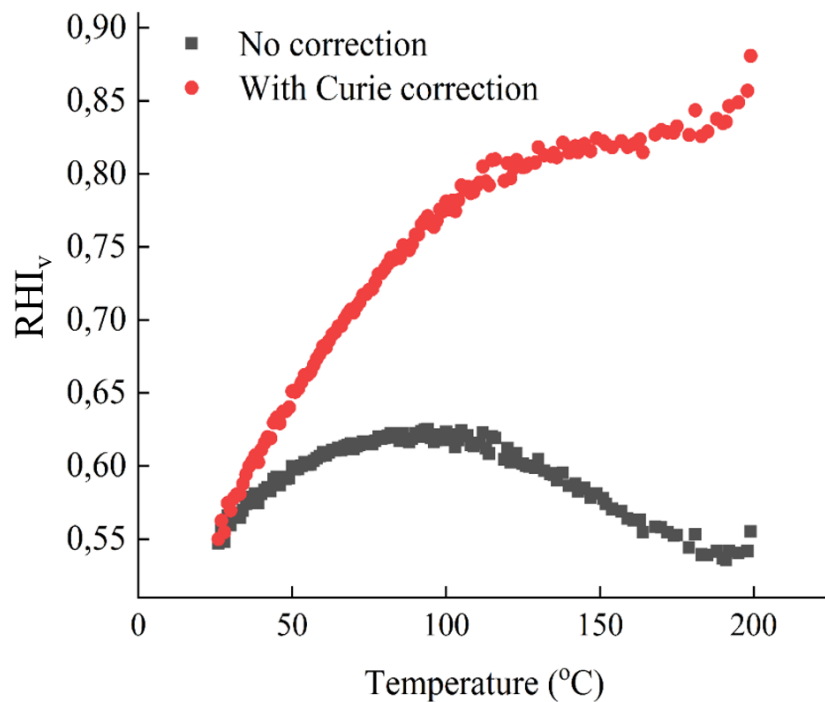


Figure 8: Relative hydrogen index for a defined volume of JC bitumen sample in the function of temperature with implemented correction (red) and without correction for the Curie effect (black).

HI reflects the number of H protons in a liquid, which is a finite value. However, in Figure 8, it is evident that even with Curie correction, RHI_v for JC bitumen changes with the temperature. This is due to the hardware limitation of the LF-NMR tools and in the presence of solid-like components (e.g., asphaltenes), which relax faster than an echo spacing (TE), which essentially means that for heavy oils and bitumens, a significant part of oil signal (i.e., H protons) remains invisible in the T_2 distribution^{19,69,71}.

2.2.5. ENHANCED NMR VISCOSITY MODEL

The RHI_v works as a correction factor by compensating the magnetization loss (Curie effect) at high temperatures. The RHI_v also accounts to a degree for the long TE, meaning that the correction coefficient for the TE term is redundant. For longer correlation times characteristic for very viscous oils ($\omega_0\tau_c \gg 1$), the right addend in Equation 24 contains the stretching parameter d (or β in Equation 21), which accounts for non-exponential relaxation, i.e., the power-law relationship with T_{2lm} . Lastly, the enhanced model contains a T_{2lm} term, inversely proportional to viscosity, which properly works for light oils. This model is expected to mitigate the following known pitfalls in NMR viscosity prediction:

- Magnetization loss due to the high temperature (Curie effect).
- Long echo times (TE) which hinder the detection of solid-like components in heavy oils.
- Non-exponential relaxation (i.e., glass transition) in viscous oils where $\omega_0\tau_c \gg 1$.

Taking into account Equations 15-23, an analytical form of the enhanced NMR viscosity prediction model can be derived as:

$$\eta = \frac{a}{HI^b T_{2lm}} + c T_{2lm}^{-d} \quad (24)$$

where a , b , c , and d are obtained from the NLS regression. The left-hand addend of Equation 24 is adapted from the Bryan et al. model⁹, which correlates measured hydrogen content and T_2 logarithmic mean with oil viscosity.

2.2.6. PREPARATION OF OIL SAMPLES

Twenty-three heavy oils were analyzed in this study. All the samples were recovered from different oil formations and wells from the heavy oil reservoirs in Alberta, Canada. All the samples except one were previously used in another NMR study by Bryan et al.,⁷², and the same methodology was used for sample preparation as described previously. Oil samples were extracted from the core samples by spinning in the Ultracentrifuge Beckman 18-M at 15,000 rpm at 40 °C for approximately 60 minutes. After extraction, the water content levels were reduced below 1.0 wt% using decantation with gravitation for one hour.

The JC bitumen selected for the high-temperature tests had a residual emulsified water content of the oil determined using the Dean-Stark distillation method. The emulsified water and solid impurities were removed from the oil through a proprietary oil-cleaning system developed by the *In-Situ* Combustion Research group at the University of Calgary. Following the cleaning procedure, the emulsified water content was 0.78 wt%.

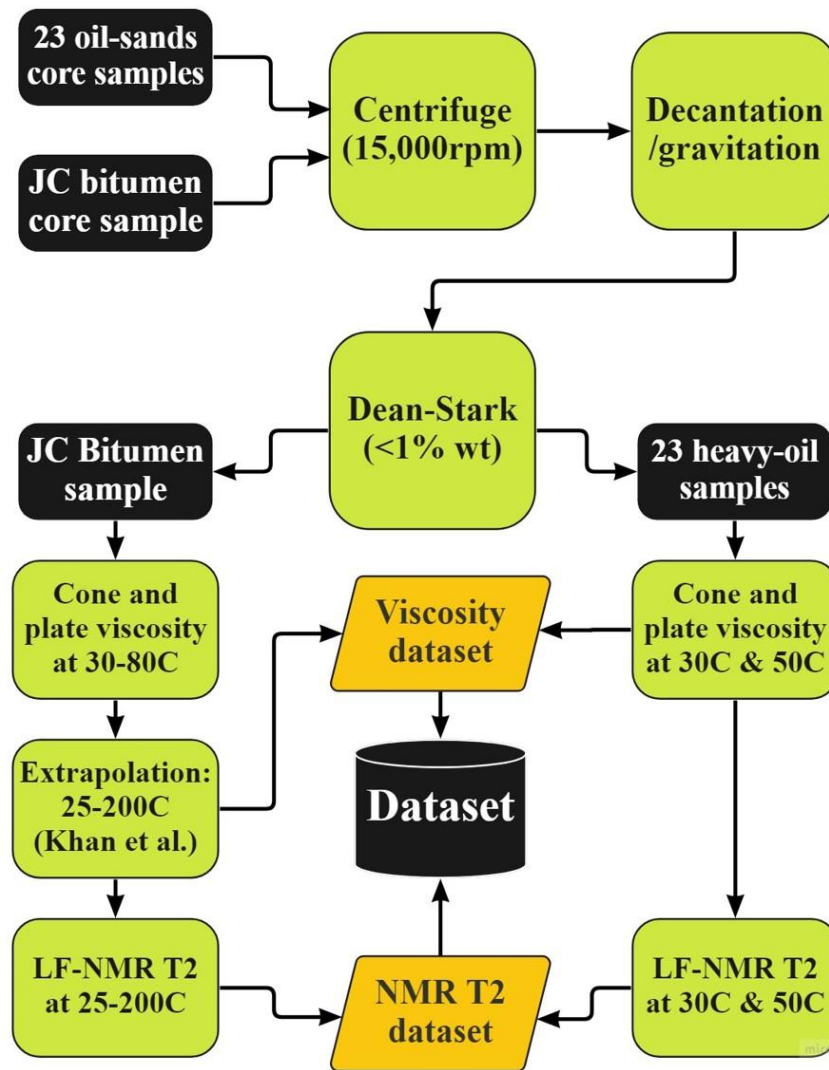


Figure 9: Flowchart representation of experimental program for 23 heavy oil samples and JC bitumen sample

2.2.7. RHEOLOGICAL MEASUREMENTS – 23 HEAVY OIL SAMPLES

Rheological measurements were executed on 23 heavy oils to obtain a reference dataset compared with predictions from NMR viscosity models. Previous studies have shown that cone and plate rotational viscometers provide higher accuracy in measuring viscous fluids than glass-capillary and oscillating-piston viscometers⁷². The Brookfield DV-II-Pro cone and plate viscometer was used, which meets ASTM D4287 industry standard for oil viscosity measurement⁷³. As previously described by Bryan et al.²⁴, the cone diameter was 12 mm with an angle of 1.5°. Shear rate

accelerated from 0.8 s^{-1} to 100 s^{-1} while shear stress was continuously logged. Three milliliters (3 ml) of oil were used for each experiment at two fixed temperatures – $30 \text{ }^{\circ}\text{C}$ and $50 \text{ }^{\circ}\text{C}$. By measuring at two temperatures rather than one, we increased the number of data points. However, four heavy oils had a limited supply, and their measurements were taken only at $50 \text{ }^{\circ}\text{C}$, making 42 data points in total. The viscosity was expressed as a ratio between shear stress and shear rate.

2.2.8. RHEOLOGICAL MEASUREMENTS AT HIGH TEMPERATURE– JC BITUMEN

The most viscous heavy oil sample (JC bitumen) was selected for assembling the high-temperature viscosity dataset. Since Brookfield DV-II-Pro viscometer is equipped with a thermal bath, measurements were made from $30 \text{ }^{\circ}\text{C}$ to $80 \text{ }^{\circ}\text{C}$ on every $5 \text{ }^{\circ}\text{C}$ making 11 data points. For the reliability of the measurements, the experiment was repeated three times at each temperature. Extrapolation of viscosities to from $26 \text{ }^{\circ}\text{C}$ to $200 \text{ }^{\circ}\text{C}$ was carried out using the dynamic viscosity model for gas-free Athabasca bitumens defined by Khan et al.¹¹:

$$\ln \ln (\eta) = A \cdot \ln(T_{\text{abs}}) + B \quad (25)$$

where A and B are empirical constants calculated as a slope and intercept of absolute temperature and measured dynamic viscosity, respectively. The relationship between JC bitumen viscosity and temperature is depicted in Figure 10.

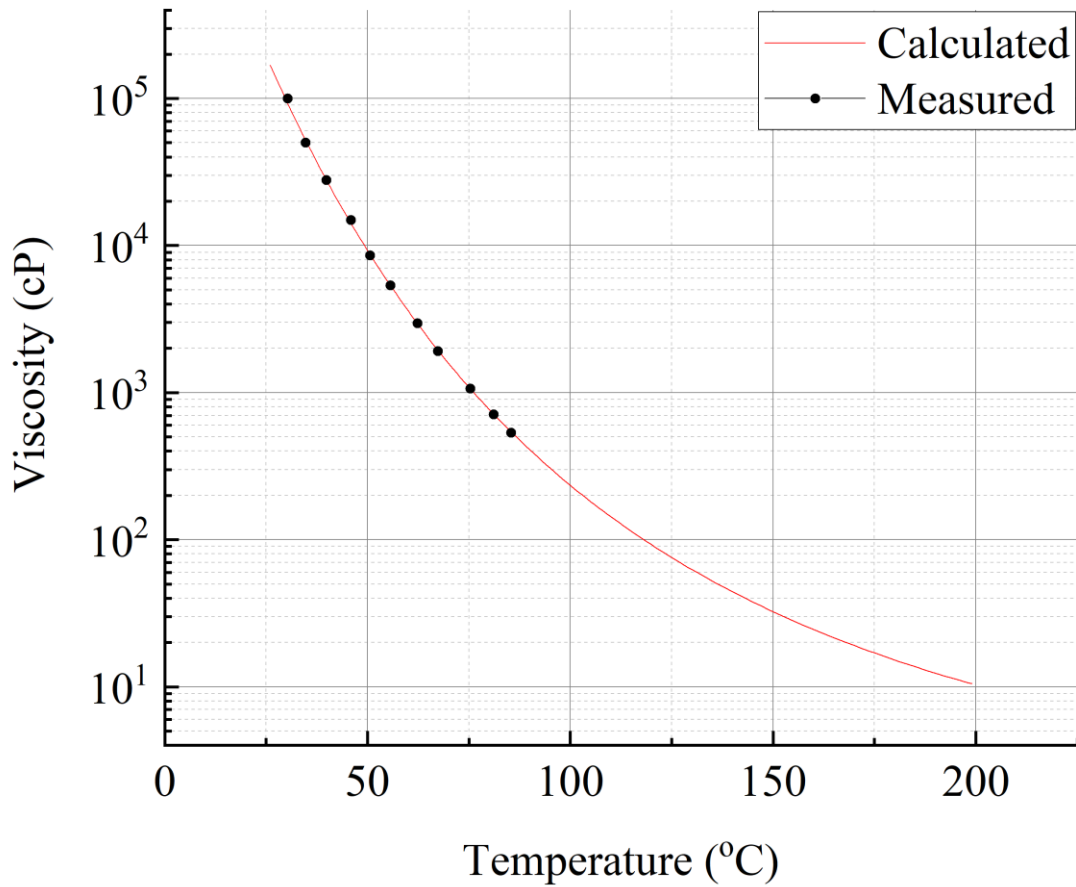


Figure 10: Dynamic viscosity of JC bitumen in 26 °C – 200 °C temperature range. Extrapolation was used for temperatures above 80 °C and interpolation between 26 °C and 80 °C.

2.2.9. NMR EXPERIMENTS – 23 HEAVY OIL SAMPLES

The suite of 23 samples was tested as a part of the previous two studies by Bryan et al.,^{24,70}. The NMR experiments were carried out at 30 °C and 50 °C using a 1.1-MHz LF-NMR Corespec 1000™ relaxometer. The Carr-Meiboom-Purcell-Gill (CPMG) pulse sequence parameters were tuned to reduce the effect of the temperature decrease within a single experiment – TE was 0.3 ms, with 2,600 pulses and a wait time 2,400 ms. Measured data were transformed into T₂ relaxation distribution in the time domain using NNLS inversion software ExpFit⁷⁰. To be consistent with the rheological viscosity dataset, the two samples

left out from viscosity measurements at 30 °C were also left out from NMR measurements at 30 °C. In total, 42 data points were obtained.

2.2.10. NMR EXPERIMENTS AT HIGH TEMPERATURE – JC BITUMEN

NMR experiments on the JC bitumen were carried out at the temperature range from 26 °C to 200 °C using a 2.66-MHz LF-NMR PERM Labmeter. The sample was stored in a polyether-ether-ketone (PEEK) thermoplastic polymer vessel with an integrated non-magnetic thermocouple for continuous temperature logging. The vessel with the oil was heated in the oven up to 200 °C and then inserted into the NMR device. The highest cooling temperature gradient occurred at 170-200 °C (~0.7 °C/min). CPMG sequence was configured to ensure that one NMR experiment was executed in no more than 1 minute with SNR>100; TE was 0.24 ms, the number of pulses was 5,000, and the wait time was 5,000 ms. Experiments were performed automatically consecutively until the sample reached ambient temperature. A total of 136 data points were used for the regression analysis.

It should be noted that PEEK plastic can produce an NMR signal in some instances, and its contribution depends on the shape of the vessel, whether it was extruded or molded, and whether PEEK contains impurities. In this study, the PEEK signal can be observed on NMR spectra, between $T_2 \sim 10\text{-}20$ ms, and it represents <1% of the total signal produced by the bitumen sample, which is why it was evaluated that it is negligible. Similar reports can be found in the literature⁷⁴.

2.2.11. NONLINEAR LEAST SQUARES (NLS) REGRESSION – MODEL TUNING

The NLS regression was performed in Origin Pro software version 2018b to tune the NMR models, that is, to obtain optimal values for their empirical constants (parameters). Where it was possible, the data population were split into the calibration set and the prediction set in the proportion of 70-30%, respectively, minimizing the overfitting. A calibration set was used to tune the model

parameters and subsequently applied to the prediction set. The model calibration was evaluated by comparing predicted NMR viscosity with rheological viscosity.

The NLS regression was performed using Levenberg – Marquardt (M-L) iteration algorithm⁷⁵.

$$\hat{\beta} \operatorname{argmin}_{\beta} S(\beta) \equiv \operatorname{argmin}_{\beta} \sum_{i=1}^m [y_i - f(x_i, \beta)]^2 \quad (26)$$

where $\beta = (\beta_1, \beta_2, \dots, \beta_i)$ are fitting parameters to be obtained from the minimization of the sum of the squared residuals $S(\beta)$ from fitted model $f(x_i, \beta)$ for the given set of independent variables (x_i) and target output (y_i). This is a step-wise (iterative) approach, where initial parameter values are set manually. To avoid convergence to a local minimum, different initial parameters were used and constrained to a specific range in some instances. For each new iteration, the parameter vector β is updated by the new estimate $\beta + \delta$, where δ can be linearly approximated from function $f(x_i, \beta + \delta)$ as:

$$f(x_i, \beta + \delta) \approx f(x_i, \beta) + J_i \delta \quad (27)$$

where J_i is a gradient of function f with respect to parameter vector β :

$$J_i = \frac{\partial f(x_i, \beta)}{\partial \beta} \quad (28)$$

Like in Tikhonov regularization, a damping factor λ is added to regulate the reduction rate of S and for more efficient discovery of a gradient direction. For the initialization of L-M, the λ was set to 0.001, and after each successive iteration was automatically increased or decreased by a factor of 10 relative to the gradient direction, depending on whether the squared residuals were reduced or increased. The L-M algorithm converges when the sum of squared residuals remains unchanged relative to the set tolerance or is equal to zero. The convergence criterion was set to $\chi^2 < 1 \cdot 10^{-3}$.

The results of NLS tuning were assessed using root mean square error (RMSE). (Equation 29). RMSE is a useful statistic for evaluating model prediction accuracy based on the new data.

$$\text{RMSE} = \sqrt{\frac{\sum_{i=1}^n (P_i - O_i)^2}{n}} \quad (29)$$

where P_i is the predicted value, O_i is the observed value, and n is the number of samples.

In the existing literature, the NMR model accuracy is usually assessed visually on cross-plots relative to the prediction bands and by comparing coefficients of determination - R^2 ^{3,22,25,65,67,70}. In this study, an adjusted coefficient of determination and standard coefficient of determination is used for the evaluation of prediction variation captured by the model

$$\bar{R}^2 = 1 - (1 - R^2) \cdot \frac{n - 1}{n - p - 1} \quad (30)$$

$$R^2 = 1 - \frac{SS_{\text{res}}}{SS_{\text{tot}}} \quad (31)$$

where \bar{R}^2 is the adjusted coefficient of determination (COD), n is the number of observations, and p is the number of independent variables (inputs). The standard COD (R^2) is calculated as a difference between the unity and ratio of the sum of squared prediction residuals (SS_{res}) and the total sum of squared residuals (SS_{tot}). Since viscosity may vary up to six orders of magnitude in thermal EOR projects, besides using RMSE and R^2 , the maximum absolute error (MaAE) of the predictions was calculated as an additional statistical metric. The MaAE represents the maximum absolute difference between predicted and observed viscosity values. RMSE and MaAE are negatively-oriented statistics expressed in source units (i.e., centipoises.)

2.3. RESULTS AND DISCUSSION

2.3.1. NMR VISCOSITY PREDICTION – 23 VARIOUS HEAVY OIL SAMPLES

The models tested in this study are listed in Table 1, describing input parameters and the number of fitting (free) parameters.

Table 1: Tested literature NMR viscosity correlations

Model	Input data	Fitting parameters
Straley, 1997	T_{2lm}	2
LaTorraca, 1999	T_{2lm} , TE, T	2
Bryan, 2003	T_{2lm} , RHI	2
Nicot, 2007	T_{2lm} , proton radius (a), inter proton distance (b)	1
Burcaw, 2008	T_{2lm} , HI	3
Cheng, 2009	T_{2lm}	3
Ahmed, 2014	T_{2lm} , TE, T	2
Musin, 2016	T_{2lm}	3
Sandor, 2016	T_{2lm} , TE, T	2
Markovic, 2019	T_{2lm} , RHI_v	4

Figure 11 shows NMR viscosity predictions and observed viscosity for a suite of 23 heavy oil samples at 30 °C and 50 °C with viscosities ranging from 70 – 21,600 cP. The NMR viscosity predictions generated by nine well-known literature models by Ahmed et al.²², LaTorraca et al.⁶⁷, Sandor et al.²³, Bryan et al.⁹, Burcaw et al.⁵⁸, Cheng et al.²⁵, Nicot et al.¹⁵, Straley et al.¹⁶, and Musin et al.³. Predictions made by the enhanced model are shown in Figure 11j. To emphasize the effect of tuning, predictions were produced with NLS tuned parameters (red) and with default model form, where used parameter values were reported by the authors (black). The analytical form of models with fitting parameters obtained by NLS is depicted in the lower right corners. The NLS regression was applied without

splitting the data into the calibration and prediction sets due to the small number of data points (42 in total). Since the oils were sampled from different locations, independent variables (T_{2lm} and RHI_v) show high variability compared to JC bitumen dataset. This is also reflected in varied adj. R^2 scores (Figure 12a), indicating that models captured different amounts of variability related to the response variable (i.e., viscosity). Compared statistical scores are shown in Figure 12.

Figure 11 shows that models by Burcaw et al., and Musin et al., do not contain predictions from the default configuration (black points) because their authors did not propose parameter values explicitly. Moreover, Figure 11 shows that generally, model predictions improve after NLS regression compared to the predictions generated by general model forms to various extent. However, in Figure 12, the effect of tuning and variation in prediction accuracy between models is clearly illustrated. For example, Bryan et al. show RMSE and MaAE scores to be 4.5 and 8 times lower after tuning, respectively, while adj. R^2 score is marginally increased.

Heavy oil models by LaTorraca et al., Ahmed et al., and Sandor et al. show a similar performance since they are based on TE correction and temperature. Sandor et al., and Ahmed et al., proposed multiple models in their work, but for this work were selected the ones with the highest reported score. After tuning, the model by Sandor et al. (Figure 11c) shows that most predictions fall within a factor of one and two at the viscosity range between 30 and 3,000 cP. However, at viscosities $>3,000$ cP, predictions scatter into a factor of two and three, causing the inflation of RMSE and MaAE scores. (Figure 12). The models by LaTorraca et al. and Ahmed et al. have less variance in $>3,000$ cP domain after tuning but tend to overpredict viscosity in the $<6,000$ cP domain, with most predictions falling within a factor of two and three. This is due to the cost function minimization, where the iteration algorithm minimizes larger squared errors in the higher viscosity domain at the expense of accuracy in the $<6,000$ cP domain.

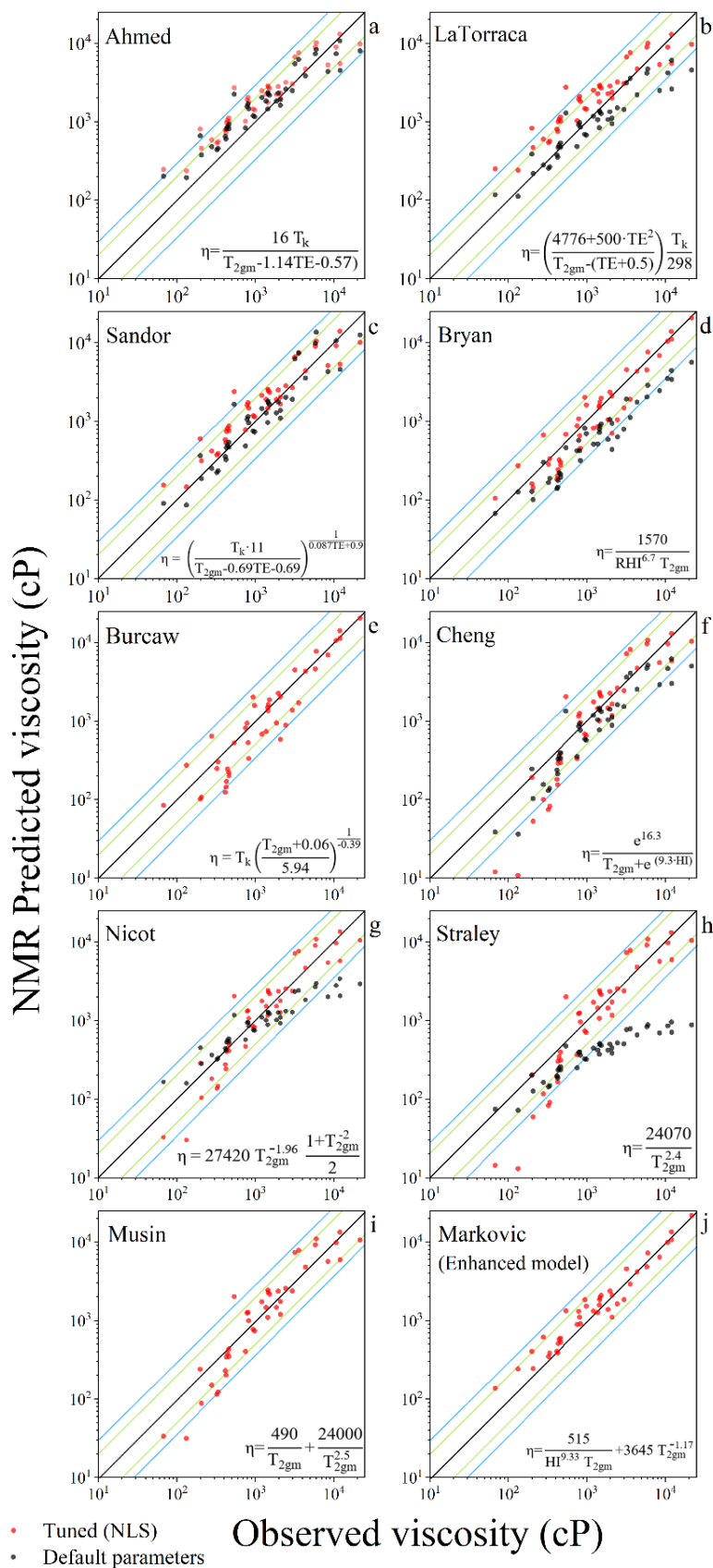


Figure 11: Rheological viscosities compared to NMR viscosities of 23 heavy oils. The Markovic et al. (j) model demonstrates the highest accuracy. A solid black line ($x=y$) presents a perfect prediction.

The models by Nicot et al., and Cheng et al. are heavy oil models, while Straley et al. is a light oil model. All three are based on the power-law T_{2lm} , which accounts for long TE (echo spacing) and have similar performance. Figure 11 and Figure 12 show that the models are affected by NLS to a different extent but generally achieve considerably improved scores after NLS. Most of the predictions in the >600 cP domain fall within a factor of one and two. Recall that Nicot et al., Cheng et al., and Straley et al. models produce large prediction residuals in the domain <600 cP after tuning. Again, this is caused by minimizing squared errors in the higher viscosity domain at the expense of accuracy in the lower viscosity domain. This issue can be solved either by constraining fitting parameters to a specific range, at the expense of the accuracy at higher viscosities, or by using these models for oilfields or wells where the viscosity **oscillates** no more than three orders of magnitude.

Figure 12 shows the segregation of the three models (Markovic et al., Bryan et al., and Burcaw et al.), which perform substantially better than the remaining seven based on all three statistics. Predictions by Markovic et al. fall within a factor of one and two along with the whole range, with only four exceptions in the <600 cP domain. The most accurate predictions are distributed in the $>1,000$ cP domain. Also, Bryan et al. and Burcaw et al. perform almost on par with the enhanced model. The new model has a marginally better score than the latter two. This marginal improvement is due to the power-law parameter, which was not considered in the models by Bryan et al., and Burcaw et al.

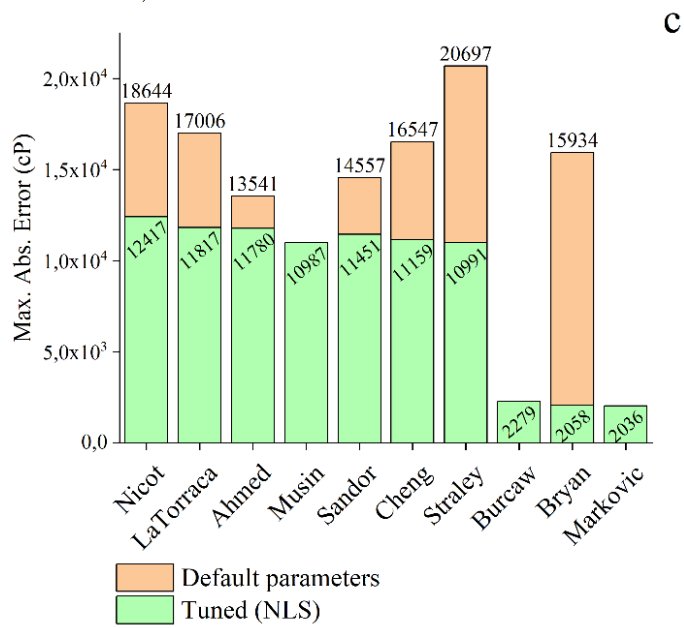
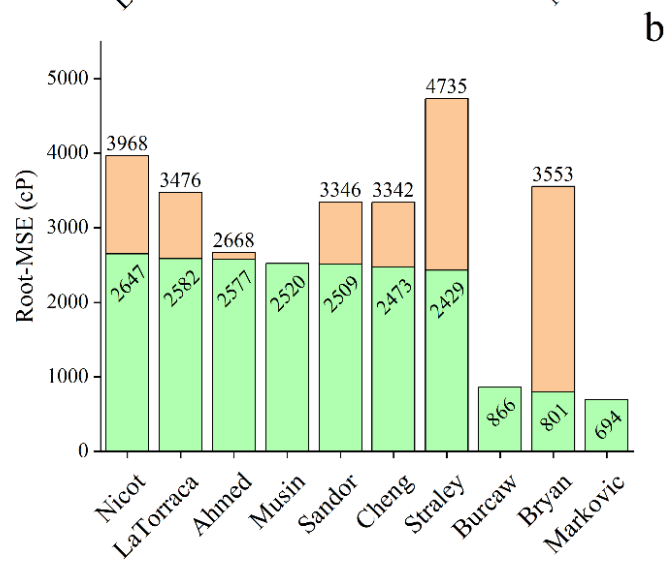
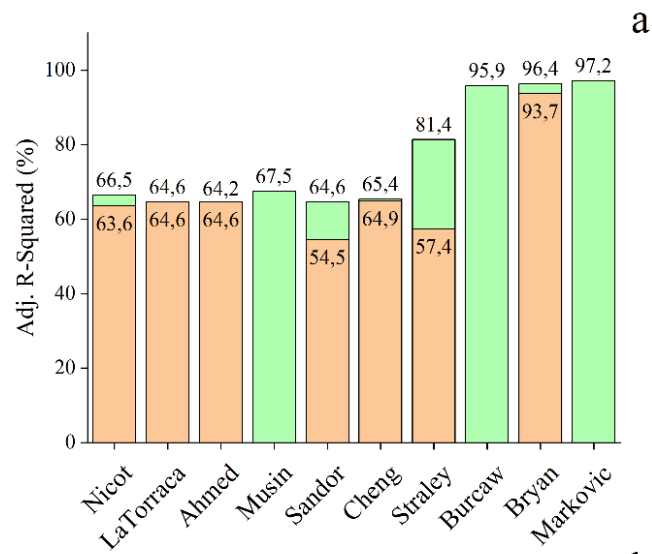


Figure 12: Compared bar chart of adjusted R^2 (a), Root-MSE (b), and MaAE (c) for tuned and default NMR model predictions of 23 heavy oils. Markovic et al. model demonstrate the highest accuracy.

These results demonstrate that the integration of RHI_v , into the correlation, substantially improves the prediction accuracy of heavy oil models in the 70–21,600 cP viscosity range at 30 °C and 50 °C, especially in the >3,000 cP domain, as demonstrated by correlations from Bryan et al., Burcaw et al., and the newly proposed model from Markovic et al. (Figure 11d, Figure 11e, Figure 11j). This finding complements the adj. R^2 scores in Figure 12a show that these three models have the highest explained variability (>96 %). In addition to the RHI_v , the enhanced model contains the power-law term in T_{2lm} , which marginally improves prediction capacity by rectifying the non-exponential relaxation effect of heavy components, for which T_{2lm} and measured RHI_v cannot account.

In conclusion, the new model (Markovic et al.) achieved the most favorable statistical scores, while the models by Sandor et al., Bryan et al., Burcaw et al., and Musin et al. have satisfactory performance only after the NLS regression. The prediction capacity of the models in high temperatures is discussed in the following section.

2.3.2. NMR VISCOSITY PREDICTION AT HIGH TEMPERATURES – JC BITUMEN

To validate the enhanced NMR viscosity model for use in steam EOR projects, it was necessary to examine how its prediction capacity is affected by the temperature increase, by testing it on a JC bitumen sample, with a viscosity range of 10–170,000 cP, for the temperature range 26–200 °C. Apart from the enhanced model, five literature correlations were selected to compare based on their performance in the previous section. These are Straley et al., Cheng et al., Bryan et al., Burcaw et al., and Sandor et al. To avoid overfitting, the JC bitumen dataset was divided into the training set the test set in proportions of 70–30%, respectively.

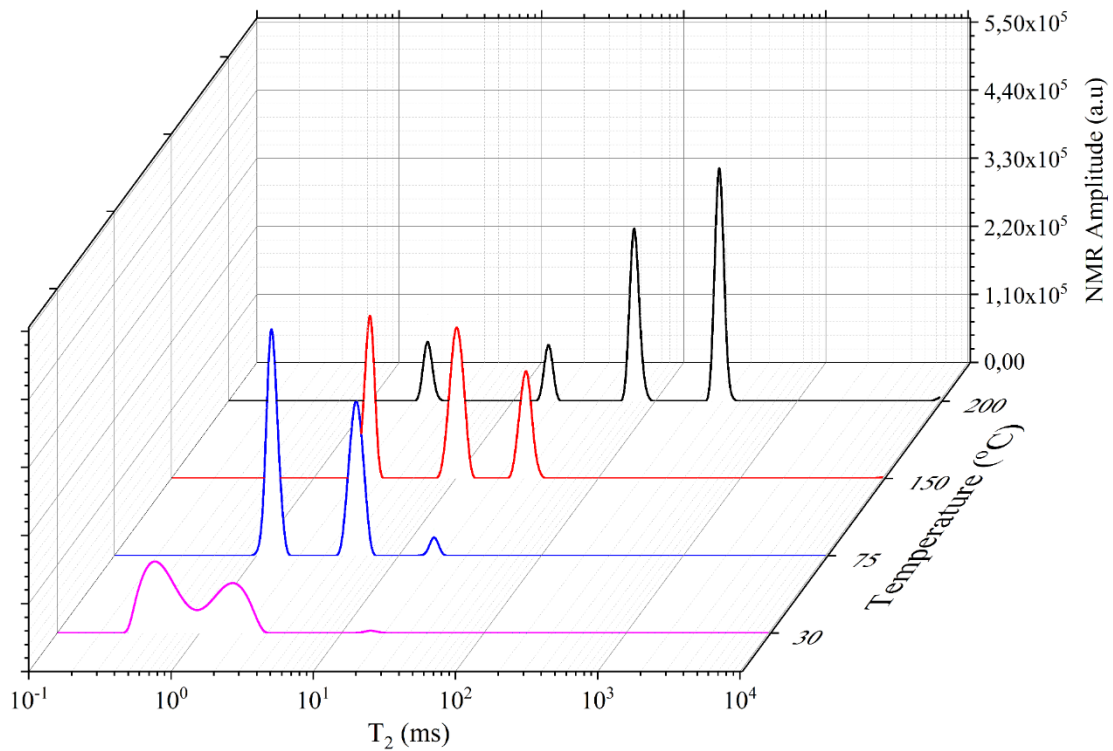


Figure 13: T_2 distribution curves of a single bitumen sample (JC bitumen) in the function of temperature.

Figure 13 presents the T_2 spectrum of JC bitumen, in the time domain, obtained using Tikhonov regularization⁹. The NMR relaxation spectra shift to the right-hand side (slow-relaxing part) with increasing temperature, and the NMR signal amplitude varies with temperature. The distribution curve at 200 °C shows four distinct peaks, which might indicate oil separation to several relaxometry components, possibly heavy and light fractions of the bitumen⁷⁶.

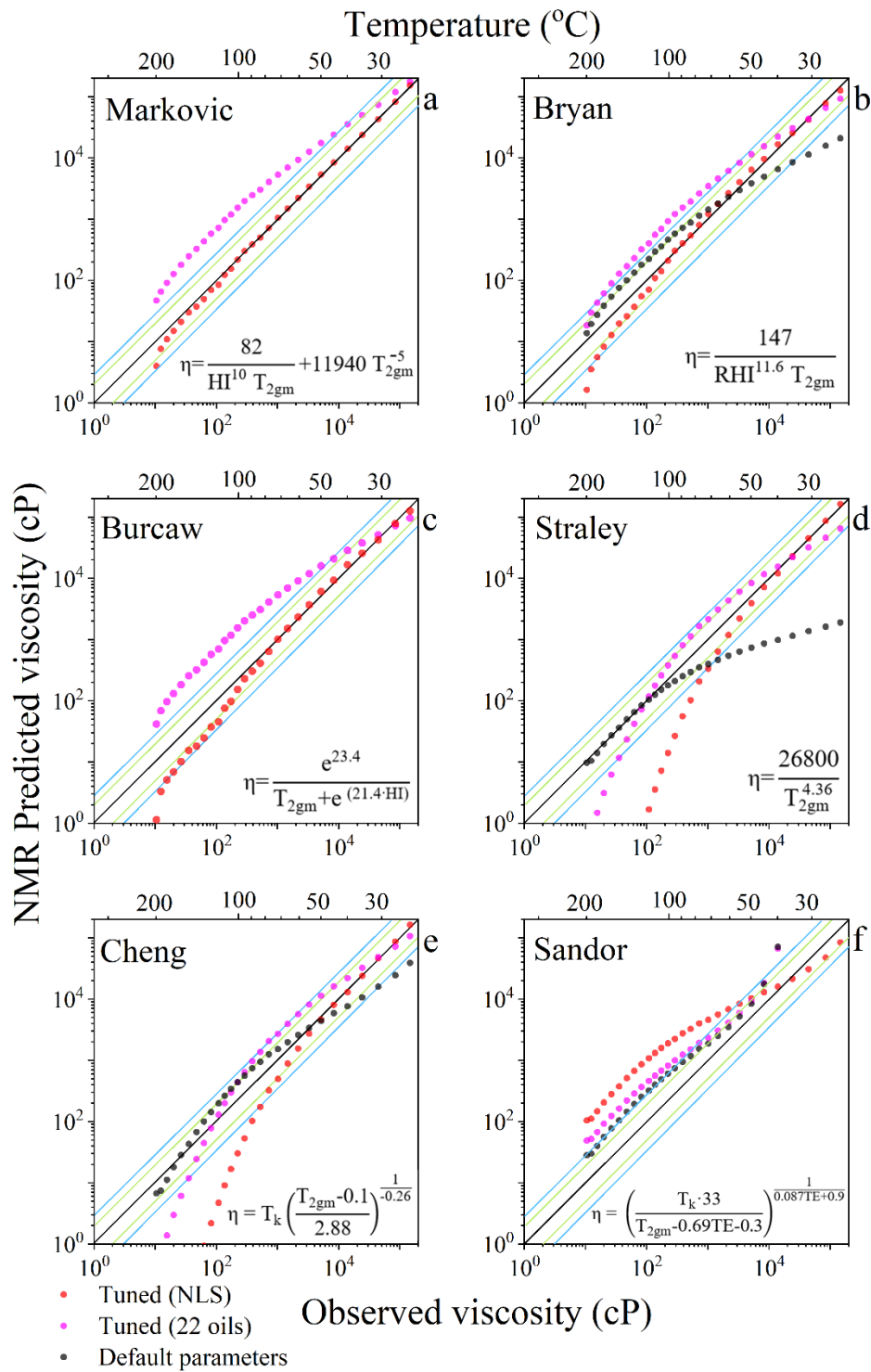


Figure 14: Rheological viscosities compared to NMR viscosities for the JC bitumen dataset. The temperature scale (top axis) is shown for clarity. A solid black line ($x=y$) presents a perfect prediction.

Figure 14 compares predicted and observed viscosity over the 26–200 °C temperature range. The predictions were generated from three model configurations: using default parameter values (black), using parameters from the previous section (magenta), and applying the NLS regression to the JC bitumen training set to calculate the new parameter values (red). The models in the analytical form after NLS regression are depicted in the lower right-hand corners of the plots.

Figure 15 shows three statistical scores used to compare the model forecasting performance. Note that the y-axis in Figure 15a is truncated for convenience. The first observation comes from Figure 15a, where high adj. R^2 scores indicate a low variability of response data (i.e., viscosity predictions), which was expected since only one sample was analyzed. However, the RMSE scores in Figure 15b and MaAE scores in Figure 15c show that NMR models achieve substantially different scores. In this way, using the adj. R^2 alone for the model performance evaluation is not sufficient.

The majority of the predictions by the Sandor et al. model (Figure 14f) fall outside the prediction bands for all three configurations. After NLS tuning, the model improves accuracy in the $>10,000$ cP domain. It should be noted that Sandor et al., for the given dataset, cannot be used with default fitting parameters because the denominator in this correlation becomes negative for the high values of the T_{2lm} , (i.e., high viscosities). This limitation was addressed by changing the constant in the denominator from -0.69 to -0.3 for NLS tuned predictions. Due to the absence of predictions for the mentioned interval, Figure 15 only contains statistics of Sandor et al. correlation for the predictions generated after NLS tuning (see Figure 14f).

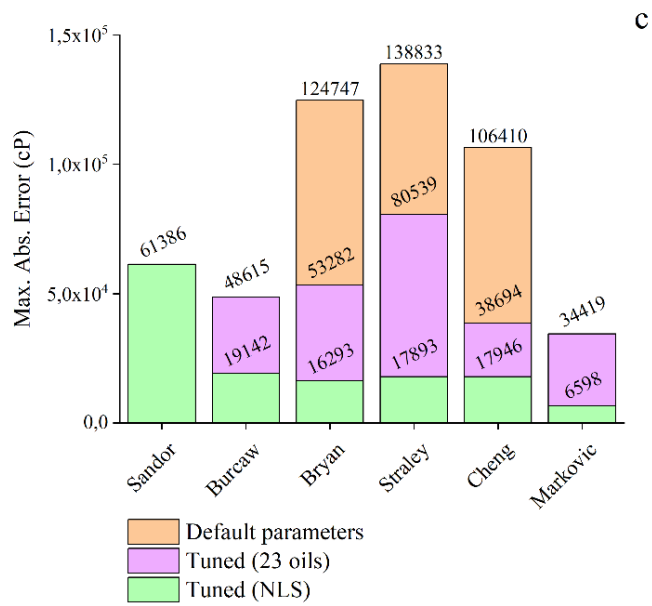
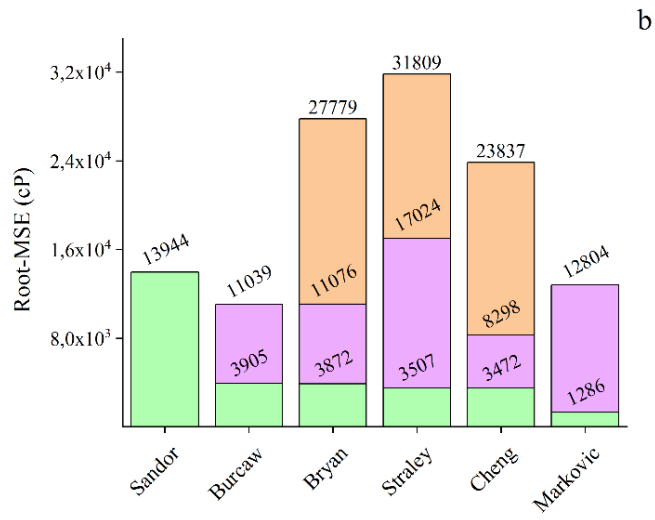
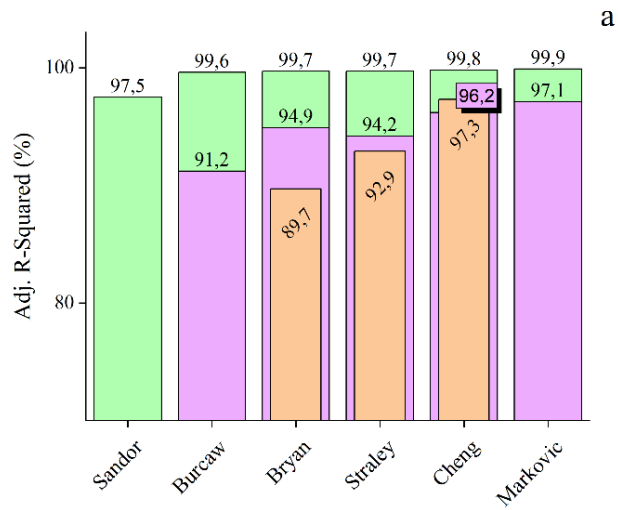


Figure 15: Compared bar chart of adjusted R^2 (a), Root-MSE (b), and MaAE (c) of NMR model predictions for JC bitumen using three model configurations. The model by Markovic et al. demonstrates the highest accuracy after NLS regression.

The Cheng et al. model (Figure 14e) with default fitting parameters reproduces the most accurate predictions compared to the other two correlations with default parameters by Bryan et al. and Straley et al., whose predictions fall within a factor one and two in the 10–10,000 cP interval. Accuracy deteriorates at >10,000 cP, which induces high RMSE and MaAE scores (Figure 15b, 15c, Cheng). In comparison, Cheng et al. underpredicts the viscosity <1,000 cP domain after tuning, especially in the 10–60 cP interval where predictions approach 0 cP, while accuracy is improved in the >10,000 cP domain. As expected, similar prediction behavior is exhibited by the model from Straley et al. This behavior can be attributed to the Curie effect and NLS regression.

The Curie effect manifests through NMR signal loss with temperature increase. Figure 7 shows that the slope of the JC bitumen NMR signal gradually decreases with rising temperature until the inflection point at 100 °C, after which the slope becomes negative. This effect is illustrated in a varying degree for all models in Figure 14 (particularly in Figure 14d, 14e in the <1,000 cP, or >70 °C domains); Sandor et al. overpredict the viscosity in this domain due to the TE coefficient, which overcompensates for Curie effect. However, the tuning process affects this further. During NLS regression, the iteration algorithm minimizes the squared residuals in the domain where the highest errors occur (i.e., >10,000 cP), causing the prediction accuracy to decline in the 10–1,000 cP range. After NLS tuning, this shift in accuracy explains why all models work better in the high viscosity domain.

The combination of heavy oil chemical complexity, signal loss due to long TE (i.e., echo spacing), and the Curie effect represent the main challenge for developing a single NMR model for predicting viscosity in both low and high viscosity systems and for the same oil (JC bitumen) at various temperatures. Each heavy oil and bitumen component relaxes exponentially, resulting in a complex total echo decay

depending on temperature and each component's phase state. The proposed enhanced model addresses these problems by integrating the RHI_v and T_{2lm} power-law term into the correlation. Stretched-exponential derived power-law term considers a smooth distribution of relaxation times for fast-relaxing components. The RHI_v compensates for signal loss since it is in the corrected form while simultaneously accounting for the long echo times (TE). The power-law term in these conditions has two functions: it improves the accuracy in the $>1,000$ cP domain, and it supplements the measured RHI_v at high temperatures (>100 °C) in correcting the predictions in the $<1,000$ cP domain (e.g., Bryan et al., vs. Markovic et al. in >100 °C domain in Figure 14, red points).

Compared to literature models, statistical scores in Figure 14 demonstrate the improved prediction capacity of the new NMR model across the entire viscosity and temperature range. Alternatively, the accuracy of any model presented in this paper can be improved for datasets with smaller viscosity and temperature ranges using the NLS regression or by splitting the calibration set into two or three subsets (e.g., low, medium, and high viscosity subsets), and performing NLS regression individually for each subset.

2.4. SUMMARY

This study demonstrates that LF-NMR relaxometry can be applied not only for viscosity prediction in a broad viscosity range but also at a broad range of temperatures (26-200 °C). The results show that published NMR viscosity models cannot accurately predict heavy oil viscosity at this range of temperatures. The enhanced NMR model was associated with an NLS regression (tuning) and used to predict the viscosity of two distinct datasets: a 23 heavy oils at 30°C and 50°C from several wells and reservoirs in Alberta and a single bitumen sample (JC bitumen dataset) at 26–200 °C. The prediction quality was evidenced by the root mean square error (RMSE), maximum absolute error (MaAE), and adjusted coefficient of determination (adj. R^2). The new model scored an RMSE of 1,286 cP for the JC

bitumen sample compared to the RMSE of 23,837 cP generated by the first runner-up model in default calibration from the literature. For the suite of 23 heavy oils, the enhanced model scored an RMSE of 2,036 cP compared to the RMSE of 15,934 cP generated by the first runner-up literature model. The results also indicate that the new heavy oil NMR viscosity model can be configured for monitoring purposes in high-temperature conditions for order-of-magnitude viscosity monitoring.

Lastly, low-field NMR measurements are fast and non-destructible. The NMR equipment can be used in-situ in observation wells, online, or inline for the heavy oil viscosity monitoring during a thermal EOR project with a 200 °C upper-temperature limit, which corresponds to steam injection temperatures.

Chapter 3 IMPROVED OIL VISCOSITY CHARACTERIZATION BY LOW-FIELD NMR USING FEATURE ENGINEERING AND SUPERVISED LEARNING ALGORITHMS

3.1. MOTIVATION

In the previous section, it was shown that LF-NMR data could be used for viscosity evaluation of various crude oils by using the enhanced NMR viscosity model, which can account for chemical complexity and a wide span of temperatures with the help of NMR derived parameters such as relative hydrogen index (RHI_v) and T_2 logarithmic mean. However, the determination of RHI or RHI_v requires a recovery of the representative oil sample from the given formation, preferably with preserved gas content, for subsequent laboratory measurements, which is often a technically challenging and expensive task^{77,78}. Moreover, oil saturation volumes must be determined independently, which is necessary to normalize the measured oil NMR response by the amplitude of an equal quantity of water²⁴. Unfortunately, in the circumstances like these, the empirical NMR models without RHI do not perform satisfactorily for predicting accurate viscosities in heavy oil and bitumen systems^{79,80,81}. The theoretical and empirical evidence presented in previous sections shows that T_2 -relaxation strongly correlates with oil viscosity^{16,21}. However, in heavier, more viscous oils, the T_2 relaxation behavior deviates from conventional models, changing the T_2 correlation with viscosity. Although studies are being conducted to understand better the underlying physics of H proton relaxation behavior in heavy oils^{26,27}, there is enough scientific evidence to confirm that these variations are associated with the presence of heavy components and their complex molecular structures (e.g., asphaltenes and resins)¹⁴.

This work introduced a supervised learning framework to improve the oil viscosity characterization by LF-NMR relaxometry, using only a single NMR

parameter – T₂ logarithmic mean. Although the emphasis has been made on gradient boosting regression trees (GBRT)⁸² and support vector regression (SVR)⁸³, a several other machine learning algorithms were tested as well, including decision trees (DT)⁸⁴, random forests (RF)⁸⁵, and k-nearest neighbors (KNN)⁸⁶. Multiple linear regression (MLR)⁸⁷ also used. A feature engineering (FE) approach was integrated to maximize the forecasting capacity of the models by deriving new features using the empirical findings from the NMR oil characterization domain⁸⁸. The study results indicate that this strategy can be successfully applied even to small datasets. As most of the underlying mathematical principles of tested algorithms are substantially different, we could observe the study task from different perspectives. The database used for calibration of models in the study was formed from the previously published LF-NMR crude oil data, containing over 130 light and heavy oil samples recovered from various reservoirs in Canada and the USA^{9,66,79}. The study was segmented into two stages. In the first stage, the preprocessing of data was performed together with feature engineering, which enabled the appropriate training of models. The generalization ability of the models was assessed by the K-fold cross-validation, while model performance was recorded using several statistical metrics. In the second stage, the performance of models was compared against another four well-known empirical NMR viscosity models that were trained using the same framework. The code and the data have been uploaded to GitHub repository and are available for use.

3.2. METHODOLOGY

3.2.1. GRADIENT BOOSTED REGRESSION TREES

In supervised learning, gradient boosting represents an ensemble (additive) model that can be used for solving supervised regression and classification problems. The main idea is to derive a model from a set of weak learners, typically decision trees (DTs) or their simplified versions known as decision tree stumps.

The construction of the viscosity model $\hat{\eta} = F(x)$, evolves in sequences or boosting iterations (m). For each iteration, a new decision tree (h) is added to the existing model to minimize the loss function further. This way, an updated and improved version of the model is obtained $F_{m+1}(x)$. This process is repeated until the specified number of boosting iterations is reached ^{84,89,90}.

As the goal is to estimate the vector of viscosities η from the training set (x), which consists of input features from the Table 2, and Table 3, the model can be expressed in the forward stage-wise form as:

$$F_m(x_i) = F_{m-1}(x_i) + h_m(x_i) = \eta_i \quad (32)$$

where $h_m(x_i)$ is the underlying model at m -th iteration for i -th observation. This equation can be rewritten as:

$$h_m(x_i) = \eta_i - F_{m-1}(x_i) \quad (33)$$

From Equation 33, it can be observed that each added h is fitted to prediction residuals. In gradient boosting regression, the residuals are integrated into the concept of negative gradients, enabling the use of other loss functions such as absolute loss and Huber loss⁸⁴. When dealing with datasets with a large number of outliers, the commonly used squared error loss function $L = \Sigma(y_i - F(x_i))^2$ will emphasize the larger residuals. Absolute loss function is not squaring the errors $L = \Sigma|y_i - F(x_i)|$, making it therefore more resistant to outliers. The negative gradient with an absolute loss function can be denoted as:

$$-\frac{\partial L(\eta_i, F_{m-1}(x_i))}{\partial F_{m-1}(x_i)} = \text{sign}(\eta_i - F_{m-1}(x_i)) \quad (34)$$

Since the loss function is minimized by adding a new DT and fitting it to F_{m-1} . The number of DTs can become excessively large, which can result in overfitting the training data. To prevent it, a shrinkage coefficient (ν) is introduced in the calculation of $F_m(x)$, which gauges the contribution of each tree $h_m(x_i)$.

$$F_m(x_i) = F_{m-1}(x_i) + \nu h_m(x_i) \quad (35)$$

This coefficient is also known as the "learning rate," and its optimal value can be estimated using some of the parameter search techniques ⁹¹. It should be noted that learning rate ν is in the strong inverse relationship with number of DTs, that is the number of boosting iterations (M). Usually, lower values of ν lead to a

smoother convergence if used with larger values of M ⁸⁴. A more detailed explanation of gradient boosting concepts can be found elsewhere ^{82,89,90,92}.

3.2.2. SUPPORT VECTOR MACHINES FOR REGRESSION (SVR)

The SVR is a sophisticated and straightforward supervised learning (SL) algorithm used in classification and regression tasks. The SVR is based on the structural risk minimization (SRM) principle, which was confirmed to have better performance compared to empirical risk minimization (ERM) used, for instance, in neural networks. In simpler terms, SRM prevents the overfitting of the model by balancing two inversely related hyper-parameters and consequently making a gap between the training set errors and test set errors smaller while reducing model complexity. In contrast, in ERM, a single objective minimizes the training error. What made support vector machines so famous was the introduction of kernels – the arbitrary functions whose purpose is to map the dot product of input features into the higher-dimension feature space. This functionality enables the utilization of hyperplanes, which are particularly useful in non-linear classification problems. Fortunately, the same concept was generalized for regression tasks ⁹³. In addition, SVR has been proven to be an effective method even in application to small datasets, which is a necessary implication for the task at hand.

In terms of viscosity prediction by NMR parameters, SVR has to be associated with our input features (T_{2lm} , TE, and T) and output vector η (Tables 2 and 3). Suppose we arrange all the preprocessed input features in a matrix form as $x = [x_1, x_2, x_3 \dots x_n]$, where x_n are column vectors of inputs. The measured viscosity instances can be rewritten into a response vector $\eta = [\eta_1, \eta_2, \eta_3, \dots, \eta_n]$. Thus the dataset can be defined then as $\{(x_i, \eta_i)\}_{i=1}^n$. Where n is the number of oil samples. The support vector machine regression between input and response vector can be written as:

$$\eta: f(x) = W \cdot \phi(x) + b \quad (36)$$

Here, $\phi(x)$ is the interpretation of an input matrix x in the higher-dimension space, while W and b are weight vector and bias terms. The latter two are obtained by minimizing the risk function:

$$\text{Min: } \frac{\|W\|^2}{2} + C \frac{1}{n} \sum_{i=1}^n L_\varepsilon(\eta_i, f(x_i)) \quad (37)$$

$$L_\varepsilon(\eta_i, f(x_i)) \begin{cases} 0 & \text{if } |\eta_i - f(x_i)| \leq \varepsilon \\ |\eta_i - f(x_i)| - \varepsilon & \text{otherwise} \end{cases} \quad (38)$$

where, the $\|W\|$ term is a magnitude of a vector of feature weights, which reduces the function's sensitivity to the perturbations in input x (i.e., flatness), thus gauging the robustness of a model. The right-hand side term quantifies the prediction error, measured by the L_ε loss function (Equation 38). The magnitude of residuals $|\eta_i - f(x_i)|$ is compared with the predefined value of ε , so that the residuals smaller than ε are ignored, but residuals larger than ε are penalized. Since any ε can be defined, the C parameter is introduced to regulate the tradeoff between the flatness of the $f(x_i)$ and penalty size for residuals larger than ε ⁹³. The optimization of Equations 37 and 38 can be simplified by introducing slack variables (ξ_i, ξ_i^*) instead of prediction residuals⁸³:

$$\text{Min: } \frac{\|W\|^2}{2} + C \frac{1}{n} \sum_{i=1}^n (\xi + \xi^*) \quad (39)$$

$$\text{Subject to: } \begin{cases} \eta_i - W \cdot \phi(x_i) - b \leq \varepsilon + \xi_i \\ W \cdot \phi(x_i) + b - Y_i \leq \varepsilon + \xi_i^*, & i = 1, \dots, n \\ \xi_i \geq 0 & \xi_i^* \geq 0 \end{cases} \quad (40)$$

To find the local minimum with respect to the given constraints, one can introduce Lagrange multipliers, in which case Equation 36 is transformed into:

$$\eta: f(x) = \sum_{i=1}^n (\alpha - \alpha_i^*) \cdot K(x_i, x_j) + b \quad (41)$$

where α and α_i^* are Lagrange multipliers and $K(x_i, x_j)$ is the kernel function, which maps the input features into the higher-dimension space. Further details about support vector machine regression can be found elsewhere^{83,93}.

3.2.3. DATABASE OF RHEOLOGICAL AND NMR MEASUREMENTS

The oil data were collected from our previous research and other published works^{9,66,79}. In all studies, the experimental procedure was similar: the dynamic viscosity of oils was determined using conventional laboratory instruments (i.e., cone and plate rheometers), whereas the T₂-relaxation spectra of the samples were obtained after raw materials data mathematical inversion from the measurements made by LF-NMR relaxometers. For this study, 282 data points were used for model development.

3.2.4. PREPROCESSING AND ANALYSIS OF THE DATASET

The preprocessing and analysis of all rheological and NMR data were performed using Python environment version 3.7.2 with the scikit-learn package and OriginPro 2019b⁹¹. The feature dataset consists of T_{2lm}, TE, and T, while viscosity observations were stored as output vectors. The data was divided into the training set and a test set in the 3:1 proportion, respectively. This way, we obtained a training set of 211 data points and a test set (unseen data) of 71 data points, which was used to estimate model accuracy only.

3.2.5. FEATURE ENGINEERING AND TRANSFORMATION

Feature engineering (FE) is a process in which domain knowledge is applied to perform appropriate transformations of the inputs and to extract new information from their known empirical relationships. This strategy proved to be effective in reducing the complexity of SL models, which in turn led to an increase in prediction performance⁸⁸. In our case, this entailed: (1) the transformation of inputs T_{2lm}, TE, T, and target output η , and (2) deriving new inputs from empirical relationships of T_{2lm}, TE, and T with target output η .

Table 2 shows that the ranges of inputs and outputs are out of scale, which implies that a particular transformation should be applied to normalize the data. Also, the

observed viscosity data has a long-tailed distribution as it is skewed to the right-hand side of Figure 16a, with over 95 % of samples distributed between 0.8 – 100,000 cP range. In the field of statistics, the observations outside three standard deviations (outliers) typically degrade the forecasting performance of the models and can be, therefore, omitted ⁹⁴. In our case, however, the outliers correspond to extra-heavy oils and bitumens (e.g., > 180,000 cP). In practice, the natural reservoirs in which these oils reside are often thermally treated in order to facilitate their recovery ⁹⁵. Therefore, if these samples were omitted from the training data, the valuable information about their T₂-relaxation behavior at high temperatures would be lost. This information was preserved by applying a simple logarithmic transformation to all features, which normalized the distribution of the data. The effect of log-transformation is illustrated in Figure 16b, on the example of target output η . Also, the log-transformation reduced nonlinearity of the dataset, which, in theory, should improve the performance of the SL regression models, which are efficient in solving linear problems (i.e., multiple linear regression and support vector regression).

Table 2: Descriptive statistics of input variables (features) and observations of oil viscosity η .

Input features	Units	Range/values	Mean	Standard deviation
Dynamic viscosity (η)	cP	0.87- 867,634	12,978	61,373
T ₂ logarithmic mean (T _{2lm})	ms	0.23-1239.9	59.4	165.6
Echo-spacing (TE)	ms	[0.1, 0.24, 0.3]	-	-
Temperature (T)	K	299.15-468.15	337.4	45.1

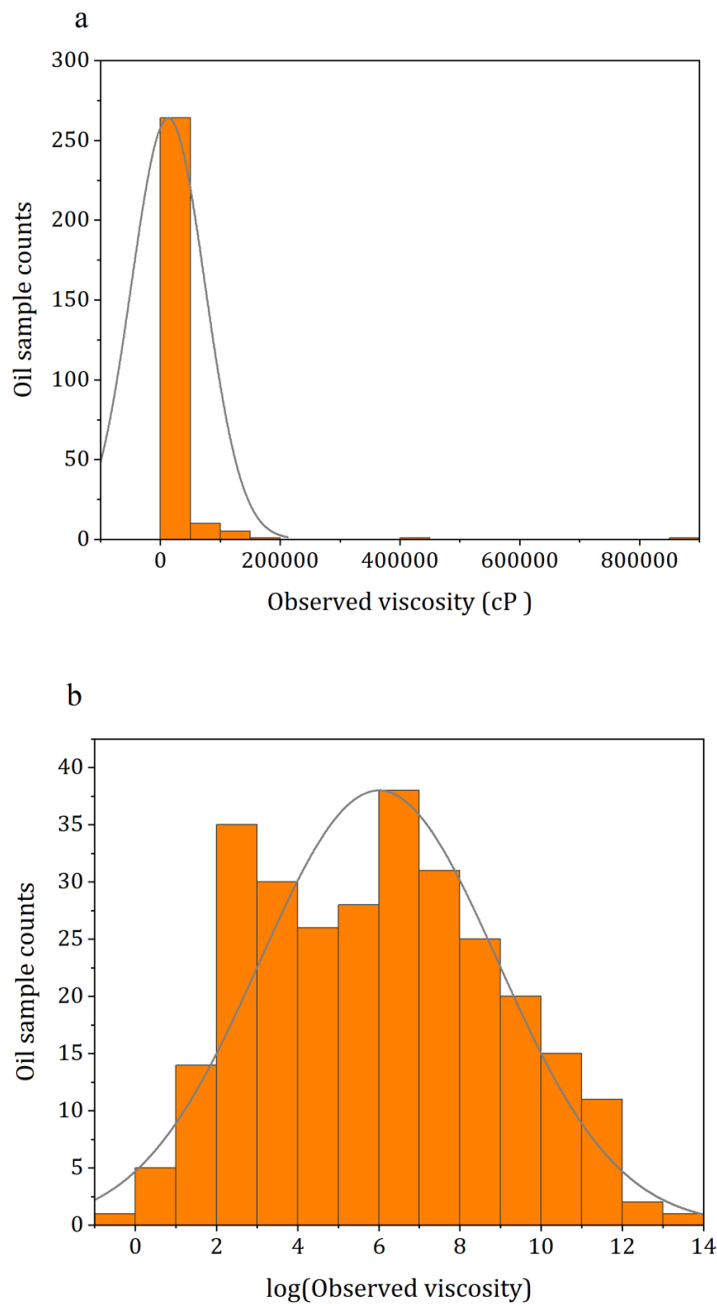


Figure 16: Distribution of oil viscosity η before (a), and after the log transformation (b).

In the second stage, we derived the new features by employing the findings from previous studies^{23,25,96}. To evaluate the importance of newly derived features, we employed the GBRT algorithm. One of the benefits of ensemble models such as GBRT is their capability of feature ranking by their relative contribution to the prediction accuracy, thus making the interpretation and selection of new features

more convenient. It should be noted that there are some downsides to feature ranking. For instance, two or more features may have a comparable correlation with the output. During feature ranking, one feature will be assigned a higher rank, which will lead others to get a lower rank, thus potentially leaving out a strong predictor⁹⁷. Figure 17a shows the ranking of the seven new features, alongside with T_{2lm} , TE, and T, with the bottom ones being the most relevant. The ranking is achieved by assessing the reduction of the training error generated from splitting the nodes of the DTs. Therefore, the features which reduce the training error more frequently during splitting will be ranked higher. Note in Figure 17a that T_{2lm} -related features ($\log(T_{2lm})/TE$, $\log(T_{2lm})$, and T_{2lm}) capture most of the variability (~72 %), while T-derived features ($\log(T)/TE$, $\log(T)$, and T) capture about 25% of the variability. This variability distribution was expected, considering that the T_{2lm} strongly correlates with η , whereas T_2 -related features become more important at high temperatures when severe NMR signal loss occurs.

In contrast, the TE-related features ($1/TE$, $\log(TE)/TE$, TE, and $\log(TE)$) affect prediction accuracy negligibly, with each being less than 1%. This is because the NMR measurements used as inputs for this study were all acquired to optimize the signal of fast relaxing fluids, i.e., through the use of small TE values. If this dataset were to be expanded to systems with larger TE values (0.6 – 1.2 ms), TE's impact would be higher. Within this dataset, the impact of TE was removed from further consideration. However, even with the perceived insignificance of TE, it should be noted that features that include the TE in the denominator demonstrate higher relevance (e.g., $\log(T)/TE$ and $\log(T_{2lm})/TE$). Figure 17b illustrates the relative importance of the remaining six features used for the training of the SL models. Finally, Table 3 summarizes the statistical description of log-transformed viscosity (i.e., target output) and engineered inputs used for SL viscosity forecasting alongside original features (T_{2lm} , TE, and T).

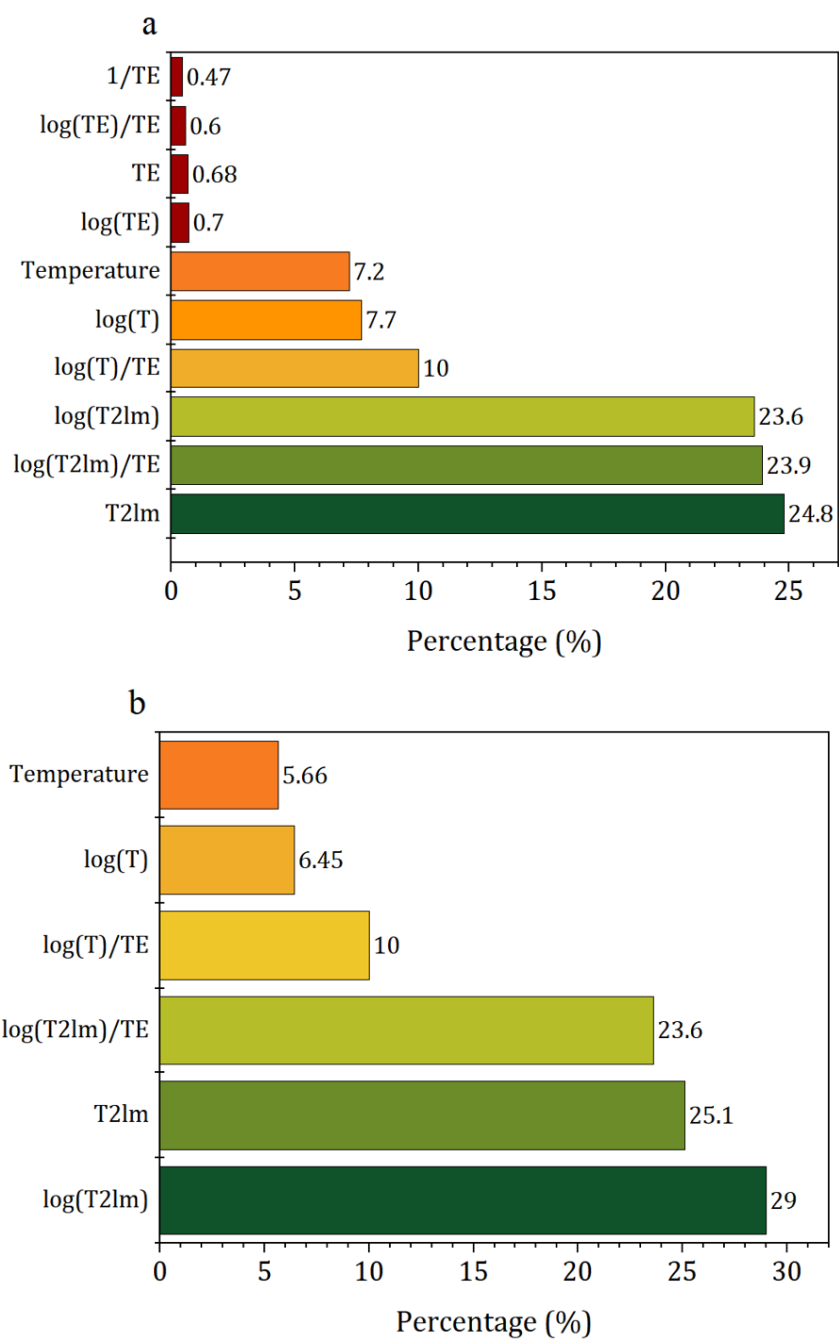


Figure 17: Relative feature importance (ranking) by GBRT model of all input features, (a) before, and (b) after removal of redundant TE-derived features with less than <1% relative contribution.

Table 3: Descriptive statistics of engineered features used for training of SL models

Engineered features	Range	Mean	Standard deviation
Log(η)*	-0.1372-13.6735	6.0015	2.9336
Log(T_{2gm})	-1.4696 - 7.1227	2.0813	1.9129
Log(T)	5.7009 - 6.1487	5.8132	0.1248
Log(T)/TE	19.04 - 57.14	34.55	16.25
Log(T_{2gm})/TE	-14.69 - 71.22	14.74	19.23

*- target output

3.2.6. EVALUATION METRICS

Five statistical metrics were chosen for the evaluation of the prediction performance of the models, including root mean square error (RMSE), mean absolute error (MAE), mean square logarithmic error (MSLE), mean absolute percentage error (MAPE), and adjusted coefficient of determination (\bar{R}^2). All metrics are negatively oriented statistical measures (i.e., smaller values are favorable), except \bar{R}^2 which is positively oriented.

The RMSE is regularly employed in scientific studies to evaluate model performance ^{36,98}. In this study, the RMSE is the square root of the average of squared differences between predicted viscosity and observed viscosity and is expressed in the centipoises:

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (\eta_i - \hat{\eta}_i)^2} \quad (42)$$

where n is a number of samples, η_i is a predicted and $\hat{\eta}_i$ is the observed viscosity. However, this metric can be sensitive to outliers, which can inflate the value of RMSE ⁹⁹. To address this issue, MAE is introduced for the calculation of averaged prediction errors of the models, in centipoises:

$$MAE = \frac{1}{n} \sum_{i=1}^n |\eta_i - \hat{\eta}_i| \quad (43)$$

In contrast to RMSE, MAE does not square the differences between predicted and observed viscosity, making MAE less sensitive to outliers¹⁰⁰. In this manner, the MAE score gives less weight to the large prediction residuals and, therefore, can be used as a control measure in RMSE interpretation. The shared disadvantage of RMSE and MAE is that both metrics do not provide any information about percentual differences between predictions and observations. MSLE accounts for this by associating squared differences between *log-scaled* predictions and observations:

$$\text{MSLE} = \frac{1}{n} \sum_{i=1}^n (\log(\eta_i + 1) - \log(\hat{\eta}_i + 1))^2 \quad (44)$$

In this manner, the MSLE avoids the heavy penalization of prediction errors in the high viscosity domain, as is the case with RMSE and MAE. Instead, it considers the relative percentual difference between observation and prediction rather than the size of their residual¹⁰¹. In addition to MSLE, MAPE illustrated the relative percentual difference between sums of errors. MAPE represents the mean of the sums of absolute percentage errors of viscosity predictions. This metric enabled a more intuitive interpretation of the model forecasts since the errors are expressed in percentages¹⁰²:

$$\text{MAPE} = \frac{1}{n} \sum_{i=1}^n \left| \frac{\eta_i - \hat{\eta}_i}{\hat{\eta}_i} \right| \cdot 100\% \quad (45)$$

Lastly, the proportion of model variance is typically expressed by the coefficient of determination (R^2) which is a standard measure of goodness-of-fit for the regression models:

$$R^2 = 1 - \frac{\sum_{i=1}^n (\eta_i - \hat{\eta}_i)^2}{\sum_{i=1}^n (\eta_i - \bar{\eta})^2} = 1 - \frac{\text{SS}_{\text{res}}}{\text{SS}_{\text{tot}}} \quad (46)$$

Although this metric provides a fast and straightforward evaluation, it might inflate due to the addition of new variables obtained from feature engineering. This inflation is a well-known problem that can be addressed by adding a term that penalizes the score with each additional predictor¹⁰³:

$$\bar{R}^2 = 1 - (1 - R^2) \cdot \frac{n - 1}{n - p - 1} \quad (47)$$

where p is a number of features. Note that $\bar{R}^2 < R^2$.

3.2.7. GBRT OPTIMIZATION

As mentioned in section 3.2.1, a choice of an arbitrary differentiable loss function ('loss' parameter), can be made according to the statistical properties of the dataset. The NMR viscosity dataset contains a substantial number of outliers, which implied using an outlier-resistant loss function, such as the least absolute deviation (LAD)⁹⁴. To test this premise, 5-fold cross-validation was executed for four commonly used loss functions: Huber loss, least squares, least absolute deviation, and quantile loss. The rest of the parameters and hyper-parameters were fixed to default values. Based on the lowest mean validation error, it was found that the GBRT configuration with LAD loss function generated the most stable predictions in terms of all error metrics (Table 4).

Table 4: Results of four loss functions used for optimizing the model performance after 5-fold cross-validation. The LAD function exhibits the best performance based on MAE_{cv} , $RMSE_{cv}$, $MSLE_{cv}$, \bar{R}^2_{cv} cross-validation (CV) scores.

Test scores	'Loss' parameter			
	LAD	Huber	LS	Quantile
MAE_{cv}	6332	7319	9072	6969
$RMSE_{cv}$	17,714	22,289	34,305	29,480
$MSLE_{cv}$	0.198	0.251	0.26	0.464
\bar{R}^2_{cv}	0.58	0.348	-0.54	-0.14

The 'criterion' parameter can be determined in the same manner. This parameter allows a user to select the function that will estimate the DT node split quality. Usually, in regression tasks with DTs, the difference between the observed and predicted value is quantified by mean squared error (MSE). Subsequently, the node splitting for a particular DT will be achieved so that the lowest MSE value is

obtained. Since MSE heavily penalizes outliers, MAE was expected to perform the splitting task more efficiently (Table 5).

Table 5: Results of three commonly used loss functions for estimating the node splitting quality after 5-fold cross-validation. The MAE function performs optimal splitting based on MAE_{cv} , $RMSE_{cv}$, $MSLE_{cv}$, \bar{R}_{cv}^2 cross-validation (CV) scores.

Test scores	'Criterion' parameter		
	MAE	MSE	Friedman-MSE
MAE_{cv}	3666	5756	5757
$RMSE_{cv}$	9925	16286	16287
$MSLE_{cv}$	0.149	0.183	0.183
\bar{R}_{cv}^2	0.86	0.66	0.64

The next step was to find the optimal hyper-parameter values for the GBRT model. According to ^{84,92}, five hyper-parameters have a considerable impact on GBRT model performance:

- Number of trees (M): maximum number of estimators or boosting iterations (n_estimators).
- Learning rate (ν): shrinkage coefficient, which regulates the individual tree prediction contribution, where each tree is being scaled by $0 < \nu < 1$.
- Subsample (λ): the proportion of the data for fitting to the individual trees.
- Max depth (J): maximum depth (size) of a tree. This value constrains the number of nodes in the tree.
- Max features (ψ): number of features used in the search for the optimal split of a tree node.

Mathematically speaking, these hyper-parameters are mutually dependent (also observable from Equation 35), which is why it was required to use a more sophisticated optimization technique other than pure trial-and-error. Therefore, these were evaluated simultaneously with the help of GridSearchCV (GS-CV), an exhaustive search cross-validation algorithm available in a scikit-learn package ⁹¹.

From the computer science standpoint, this metaheuristic approach iteratively optimizes an algorithm by searching for an appropriate combination of hyper-parameters in multidimensional real-valued parameter space (grid), relative to some measure of accuracy (e.g., R^2). This approach captures the interaction between the hyper-parameters, therefore significantly reducing the optimization time. However, due to discrete data (i.e., TE) and other distributions in the input data, the grid-search optimization may fail to discover the best hyper-parameter configuration, even with the appropriate transformations applied to the dataset. Also, with a growing number of hyper-parameters, its' utilization becomes computationally intensive. Hence, optimization was assessed further using error curves. Table 6 shows the hyper-parameters and their value range, which were optimized using the GS-CV approach. Recall that 'loss' and 'criterion' parameters were fixed according to results in Tables 4 and 5.

Table 6: GBRT hyper-parameter optimization by grid-search based on 5-fold cross-validation

GBRT hyper-parameters	Value range/method	Optimal values	Score
$n_estimators$ (M)	[1-500]	[220]	RMSE: 8704
$learning_rate$ (ν)	[0.01-1.0]	[0.03]	MAE: 3377
$subsample$ (λ)	[0.1-1.0]	[1.0]	MSLE: 0.136
max_depth (J)	[1-8]	[4]	MAPE: 29
$max_features$ (ψ)	[auto, sqrt, log2]	[log2]	\bar{R}^2 : 0.91

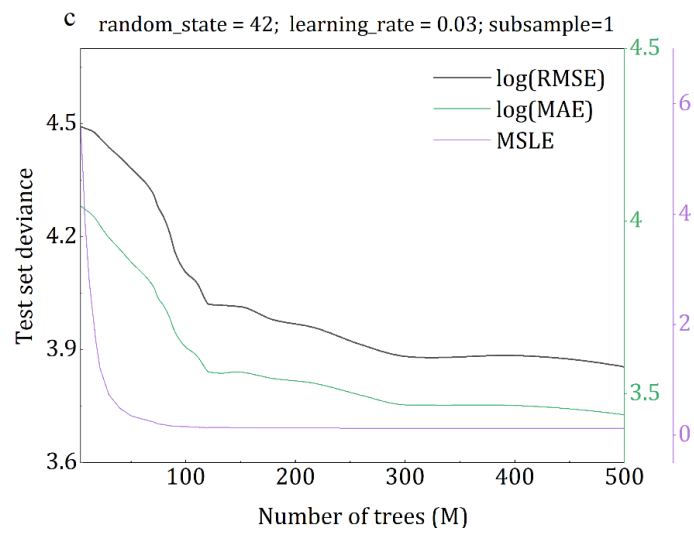
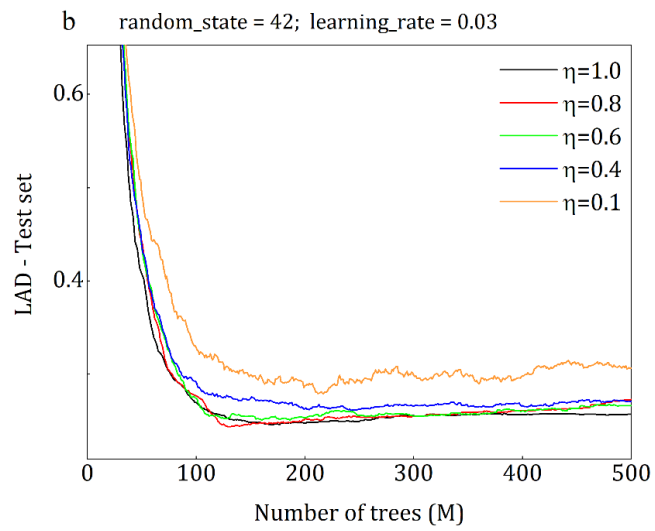
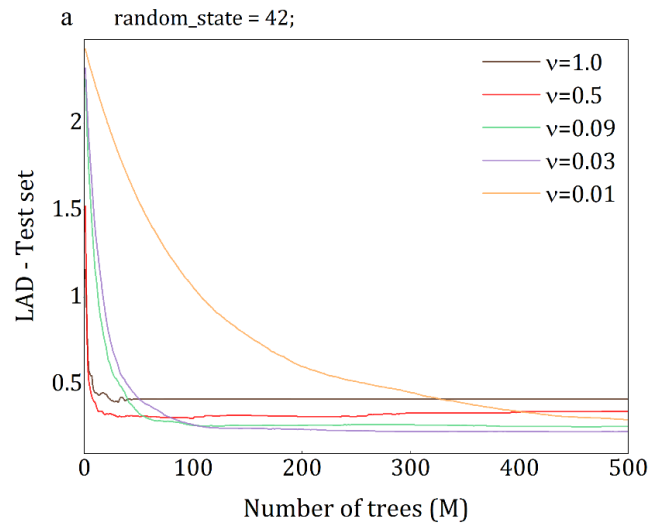


Figure 18: The test set GBRT model performance in terms of least absolute deviations (LAD) for various learning rates (a), and subsample sizes (b) relative to the number of trees M . Bottom plot (c) illustrates the model accuracy evaluation as a function of M , in terms of MAE, RMSE, and MSLE.

Figure 18a shows how the GBRT model deviance evolves with different learning rates (ν) as a function of a number of trees M . The GBRT loss is expressed in the least absolute deviations. Larger values of ν (e.g., $\nu=1$) lead to faster convergence, that is, smaller values of M are needed for the deviance to converge. However, when a value is decreased ($\nu=0.01$), the contribution of every additional estimator is reduced further, leading M to increase to ensure smooth convergence, which also means an increase in computational cost. Since the apparent tradeoff exists between these two parameters, the parameter grid-search cross-validation was used as a strategy for obtaining their appropriate values.

Additionally, subsampling λ is a parameter that enforces the variance reduction of the sample population. In GBRT applications for large datasets, this technique proved helpful for improving computing performance and accuracy⁸⁴. Figure 18b, however, shows that no subsampling ($\lambda =1$) leads to the smoothest and lowest deviance for the given input, possibly due to the small number of data points. Also, alternating the λ parameter has only a minor effect on deviance magnitude. In fact, the variations are so minor that one must zoom in on the y-axis to observe this behavior (note y-axis scales of Figure 18a and 18b).

Finally, the maximum depth of all trees was restricted to the same size ($J=4$), as determined by the GS-CV, which agrees with recommendations in literature⁸⁴. The optimal number of features for the best split of the tree node was found to be $\psi=2$ (i.e., $max_features = "log2"$). It should be noted that the value of the latter has the least impact on the prediction performance of the GBRT model, and therefore, using the default value (i.e., "auto") is also acceptable.

3.2.8. SVR OPTIMIZATION

In Equation 41, the term $K(x_i, x_j)$ represents the kernel function. The standard kernel functions used in SVR are linear, polynomial, sigmoid, and Gaussian. The NMR input parameters are in the nonlinear relationship with the oil viscosity; therefore, the kernel must capture this relationship once the input features are mapped into a higher dimension space. In these circumstances, the Gaussian, or radial basis function (RBF) kernel has proven to be effective¹⁰⁴. From Equation 40, the kernel function can be expressed as:

$$K(x_i, x_j) = \exp\left(\gamma \cdot \|x_i - x_j\|^2\right) \quad (48)$$

where γ is the width hyper-parameter of the RBF kernel. Hence, there are three main hyper-parameters which need to be optimized:

- Gamma (γ): RBF kernel specific parameter which defines the support vector's radius of impact.
- Epsilon (ϵ): the insensitivity radius- ϵ within which the prediction residuals are ignored ($loss=0$). This value controls the number of support vectors (SVs) and the smoothness of the function.
- Regularization parameter (C): hyper-parameter, which affects the size of the penalty applied to model predictions. If too large, the model may store an excessively large number of SVs and cause overfitting.

Literature findings show that the behavior of these hyper-parameters is interrelated, which should be considered during their optimization⁹³. Thus, the GS-CV approach was used to simultaneously approximate C , ϵ , and RBF kernel parameter γ (Table 7). Also, their in-depth assessment was performed from the analysis of the error curves (Figure 19).

Table 7: SVR hyper-parameter optimization by grid-search based on 5-fold cross-validation

SVR hyper-parameters	Range/method	Optimal values	Score
Gamma (γ)	['scale', 0.1-1·10 ⁻⁵]	[5·10 ⁻⁴]	RMSE: 8704 MAE: 3377
Epsilon (ϵ)	[1-1·10 ⁻⁵]	[1·10 ⁻⁴]	MSLE: 0.136 MAPE: 29
Regularization (C)	[1-500]	[25]	\bar{R}^2 : 0.91

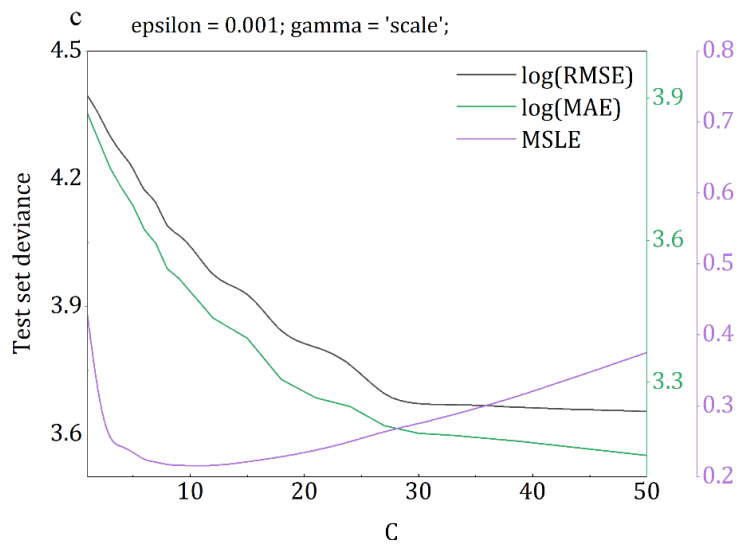
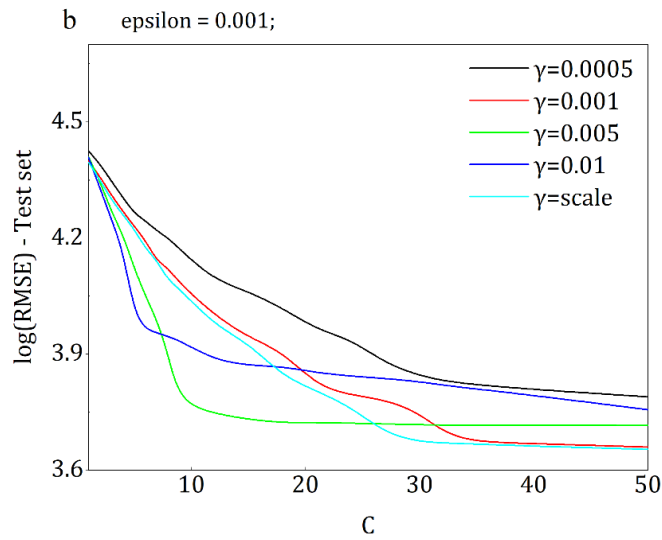
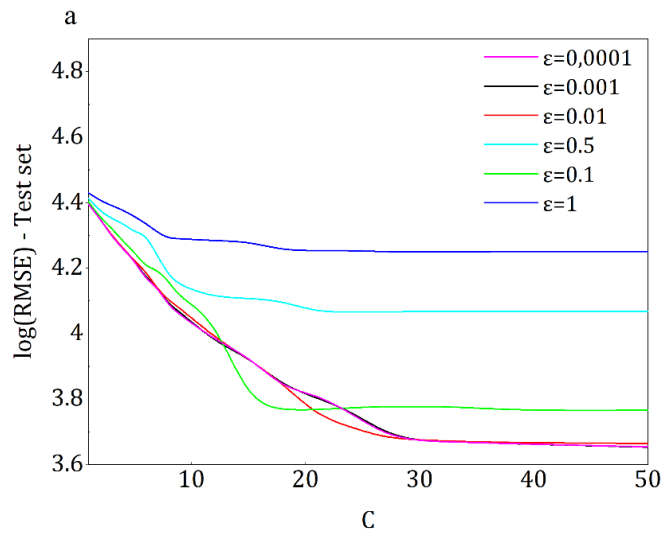


Figure 19: Test set SVR model performance in terms of log-normalized RMSE for various values of ϵ (a), and γ (b) with respect to regularization C . Bottom plot (c) illustrates the accuracy of optimized SVR model as a function of C , in terms of three error metrics; $\log(\text{RMSE})$, $\log(\text{MAE})$ and MSLE.

Figure 19a presents how SVR model prediction accuracy behaves for various values of radius- ϵ in the function of C . For the $\epsilon=10^{-4}$ obtained by GS-CV, it was found that the SVR model utilized over 70 % of the data samples as support vectors, which indicates overfitting ¹⁰⁵. For values of $\epsilon=10^{-3}$ and $\epsilon=10^{-2}$, the deviance converged smoothly at $C=30$. The number of SVs was reduced by increasing ϵ to 10^{-3} (Figure 19a, black) while preserving nearly identical accuracy.

Figure 19b shows deviance for the fixed ϵ and various radii of individual SV impact γ . The smoothest convergence and lowest deviation are achieved when $\gamma='scale'$, which is the value when an inverse of the number of features is scaled by their standard deviation. Interestingly, the GS-CV obtained $\gamma=5 \cdot 10^{-4}$, but according to its' plot (black curve), the deviance converges when $C>50$, at which point the SVR model attempts to perfectly predict each entry from the training set (hard-margin SVM behavior). Since this might lead to overfitting and increased model complexity, the γ was set to *'0.001.'*

As a final step, the model with fixed ϵ and γ hyper-parameters was evaluated in Figure 19c, where three metrics were utilized to evaluate the tuned SVR model. While both RMSE and MAE follow the same decreasing trend, the MSLE error decreases until the $C=12$ inflection point, after which it starts increasing. This behavior is due to the inflation of residuals in the low viscosity domain. To restrict the further growth of residuals and to preserve the overall model performance, the regularization was set to $C=25$, in line with the grid-search results (see Table 7).

3.3. RESULTS AND DISCUSSION

This section is divided into two parts. In the first part, we compare SVR and GBRT model performance against four other popular regression models, whereas in the second part, a performance of four well-known empirical NMR viscosity models was considered. The models are compared using the five error metrics introduced in chapter 2.7. Also, the cross-plots with predicted and observed viscosities are provided for the in-depth analysis.

3.3.1. SUPERVISED LEARNING MODELS

The performance of the GBRT and SVR was evaluated against an additional four SL algorithms, including multiple linear regression (MLR)⁸⁷, K-nearest neighbors (K-NN)⁸⁶, decision trees (DTs)⁸⁴, and random forests (RF)⁸⁵. Their optimization was performed with GS-CV, similarly as for GBRT and SVR.

Table 8: GS-CV hyper-parameter optimization results for all supervised learning algorithms which were tested in this work.

Model	Hyper-parameters	Range/method	Optimal values
Decision trees (DTs) ⁸⁴	criterion	['mse', 'mae']	['mse']
	max_features	[1, 2, 3, 'sqrt', 'log2', 'auto']	[3]
	max_depth	[1-4]	[4]
	min_samp_leaf	[1-4]	[3]
	min_samp_split	[1-4]	[2]
	splitter	['best', 'random']	['best']
K-Nearest Neighbors (KNN) ⁸⁶	n_neighbors	[1-50]	[3]
	weights	['uniform', 'distance']	['uniform']
	algorithm	['ball_tree', 'kd_tree', 'brute']	['ball_tree']
	p	[1, 2]	[1]
Random forests (RF) ⁸⁵	n_estimators	[1-80]	[7]
	criterion	['mse', 'mae']	['mae']
Support vector machines for regression (SVR) ⁹³	gamma	['scale', 0.0005-0.1]	[0.01]
	epsilon	[1-0.0001]	[0.0001]
	C	[1-500]	[20]
Gradient boosted regression trees (GBRT) ⁸⁴	loss	['ls', 'lad', 'huber', 'quantile']	['lad']
	n_estimators	[1-500]	[220]
	criterion	['mse', 'mae', 'friedman_mse']	['mae']
	learning_rate	[0.01-0.1]	[0.03]
	max_features	[auto, sqrt, log2]	[log2]
	max_depth	[1-8]	[4]
	subsample	[0.1-1.0]	[1.0]

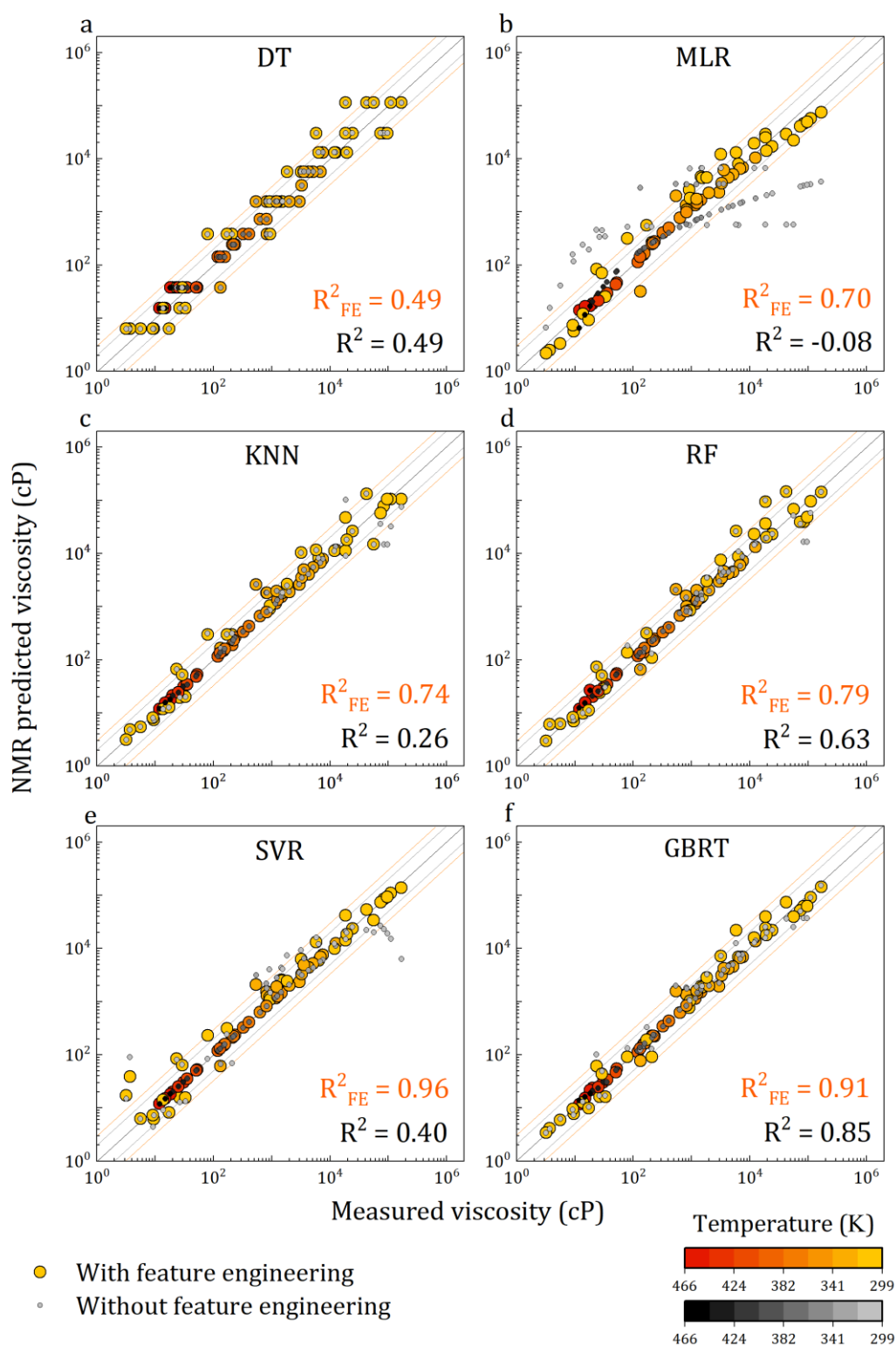


Figure 20: Comparison of NMR SL viscosity model predictions and observations. Note that the grayscale points are predictions of models generated without FE, while warm color points are predictions with FE. Lighter colors indicate lower temperatures (from

25 °C/299 K), and more intense, darker colors indicate higher temperatures (up to 200 °C/466 K). GBRT and SVR models with integrated FE demonstrate the best performance.

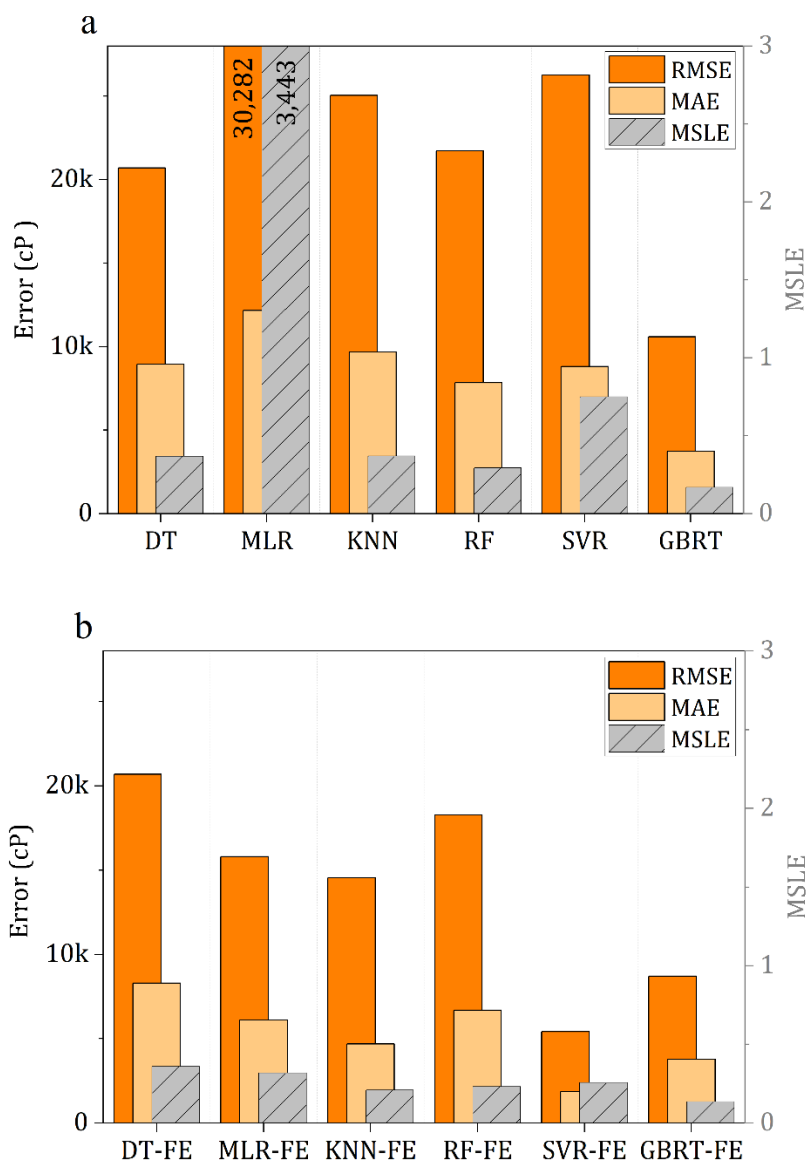


Figure 21: Compared statistical scores of SL models without FE (a) and SL models with integrated FE (b). SVR-FE and GBRT-FE demonstrate the best statistical performance.

When Figures 19 and 20 are examined together, one can note that the overall performance of each model improves after the integration of FE. This effect is, however, not proportionally pronounced for all models. For instance, the MLR-FE model's prediction variance has reduced dramatically after employing FE (Figure 21b). Interestingly, for GBRT and RF models, the variance-reducing effect from FE

is much smaller than in the latter's case (Figures 19e and 19c). The same observation applies to the DT model, the base estimator of GBRT, and RF models (Figure 20a).

This difference in performance comes from the difference in the underlying mathematical principles of these models. As the MLR model is *linear*, the nonlinearity reduction from the log transformation naturally improved the model's generalization and stability. Additionally, the integration of new features reduced the variance of the predictions, which resulted in a further shrinking of residuals. A similar is valid for the SVR model, though to a smaller extent (Figure 20f) ¹⁰⁶. However, in the case of RF, GBRT, and DT models, the log transformation did not impact the performance because the background tree-branching process does not rely on numerical values of the features but instead uses the rank of the features, which remained the same after transformations ⁸⁴. Thus, the variance reduction came solely from capturing more variability from the new features derived in the second step of FE. Furthermore, when we compare the DT scores in Figure 21, with those by GBRT, we observe a massive gap in performance, which perfectly illustrates the advantage of ensembles of DTs over a single DT. One of the reasons for the poor performance of single DT models is their 'habit' of overfitting the training data, making them unstable with unseen data. Therefore, ensembles of DTs generate a variance that minimizes the overfitting ⁸⁴.

Lastly, the KNN is a simple algorithm where the output values are forecasted based on the similarity between the input features. This similarity is calculated as a distance (e.g., Euclidian, Manhattan, etc.) from k-instances, defined by the user ⁸⁶. Feature engineering improves KNN scores almost proportionally to RF and GBRT models, however not enough to minimize the large residuals in the high viscosity domain, which causes the RMSE and MAE scores to rise (Figures 20a and 20b).

Another remark is that the significant temperature variations seem to have a negligible effect on the performance of all supervised learning algorithms. The

predictions in the highest temperature domain overlap with the $x=y$ line in all six cases, demonstrating that each algorithm has appropriately captured the relationship between observed oil viscosity and NMR signal loss that occurs at high temperatures. In comparison, empirical models tested in this work^{16,23,25,96}, exhibit poor performance in these conditions⁷⁹, as seen in the following chapter.

3.3.2. EMPIRICAL NMR MODELS

The performances of GBRT-FE and SVR-FE models are compared with four well-known empirical NMR viscosity models based on T_{2lm} , TE, and T. These models were developed by Straley et al.¹⁶, Nicot et al.⁹⁶, Cheng et al.²⁵, and Sandor et al.²³. Previous research showed that tuning by non-linear least squares (NLS) improves the performance of empirical models⁷⁹. However, the viscosity dataset in the present study has long-tailed distribution with many outliers at higher viscosities, which dominate the sum of squares minimization, thus ultimately leading to erroneous model fit and misleading statistical scores^{107,108}. Hence, the model fitting was performed using the orthogonal distance regression (ODR), proved to be a successful technique for dealing with outliers¹⁰⁹. Figure 22 and Figure 23 demonstrate the superior performance of both SVR-FE and GBRT-FE models in terms of all statistical metrics. The curvature of the viscosity forecasts by empirical models in Figure 22 reflects the combined influence of NMR signal loss due to the Curie effect, which occurs at high temperatures, and fast relaxation by solid-like components in heavy oil, which led to poor model generalization.

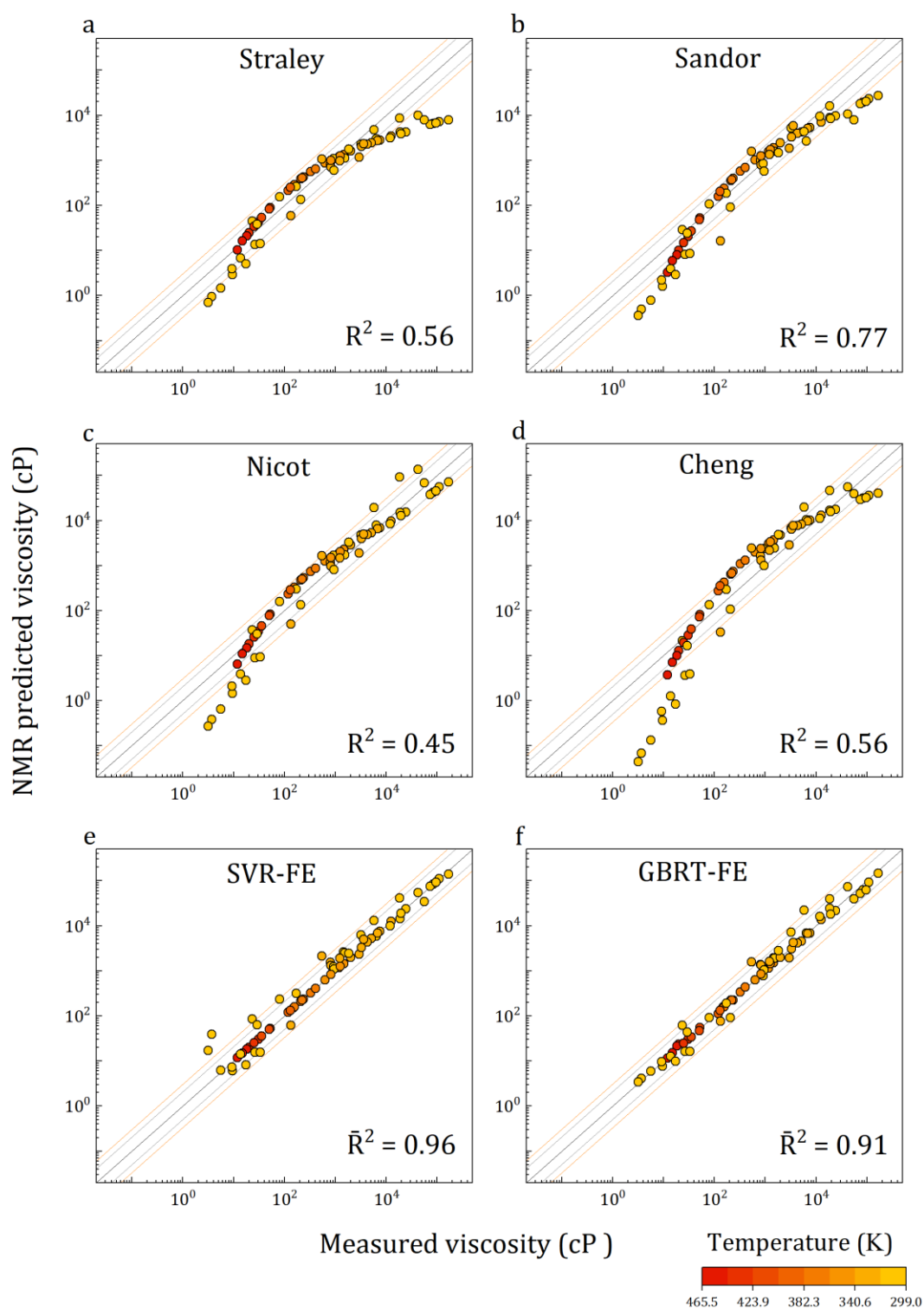


Figure 22: Performance comparison of empirical NMR viscosity models (a, b, c, and d) with SVR-FE (e) and GBRT-FE (f) models. GBRT-FE and SVR-FE demonstrate significantly better performance.

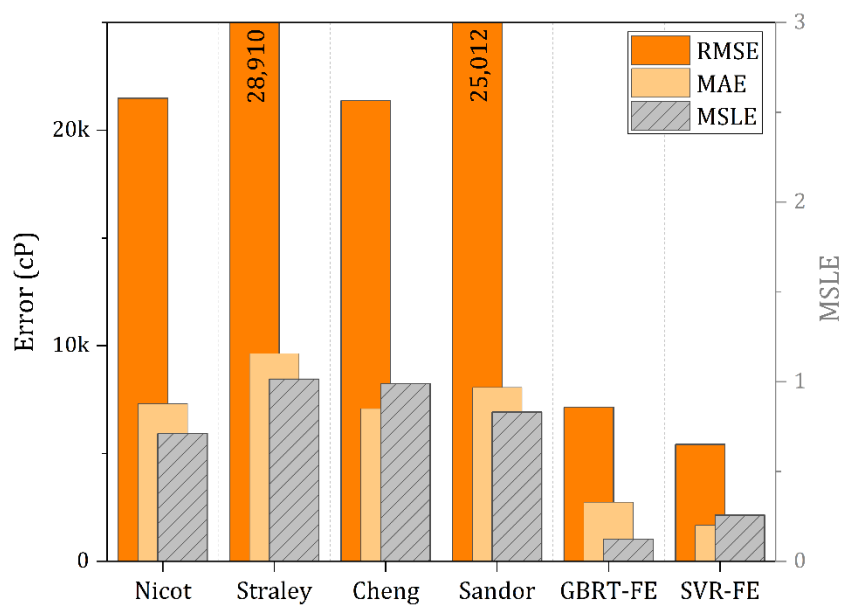


Figure 23: Compared statistical scores of four empirical NMR viscosity models and SVR-FE and GBRT-FE supervised learning models in terms of RMSE, MAE, and MSLE. SVR-FE and GBRT-FE demonstrate significantly better statistical performance.

3.3.3. SVR-FE vs. GBRT-FE

When cross-plots from Figure 20 and Figure 21 are analyzed along with scores in Figure 22, Figure 23, one can conclude that SVR-FE and GBRT-FE models have superior test performance than any other SL model. For instance, in the case of MLR-FE, K-NN-FE, and RF-FE, the SVR-FE model, on average, scores 2.5 times lower RMSE and MAE, while GBRT-FE achieves nearly two times lower scores. When their performance is compared to empirical models, the difference in performance is even more substantial; the SVR-FE model has about 4.5 times lower RMSE and MAE scores, whereas GBRT-FE achieves nearly 3.5 times lower scores.

The principal difference in the performance of these two models is related to their precision (i.e., variance), which is evidenced by their different MSLE and MAPE scores. For instance, the SVR-FE model has a better MSLE score than empirical models (~4.5 times lower) but compared to SL models, KNN-FE and RF-FE marginally outperform SVR-FE. The same is true for MAPE scores and percentage error box plots when further examined in Figure 24. The GBRT-FE model, on the other hand, scored the best MSLE and MAPE scores in this work. These results imply that SVR-FE has the highest accuracy but somewhat lower precision (i.e., variance), relative to GBRT-FE, KNN-FE, and RF-FE models. For more convenience, all evaluation scores are summarized in Table 9.

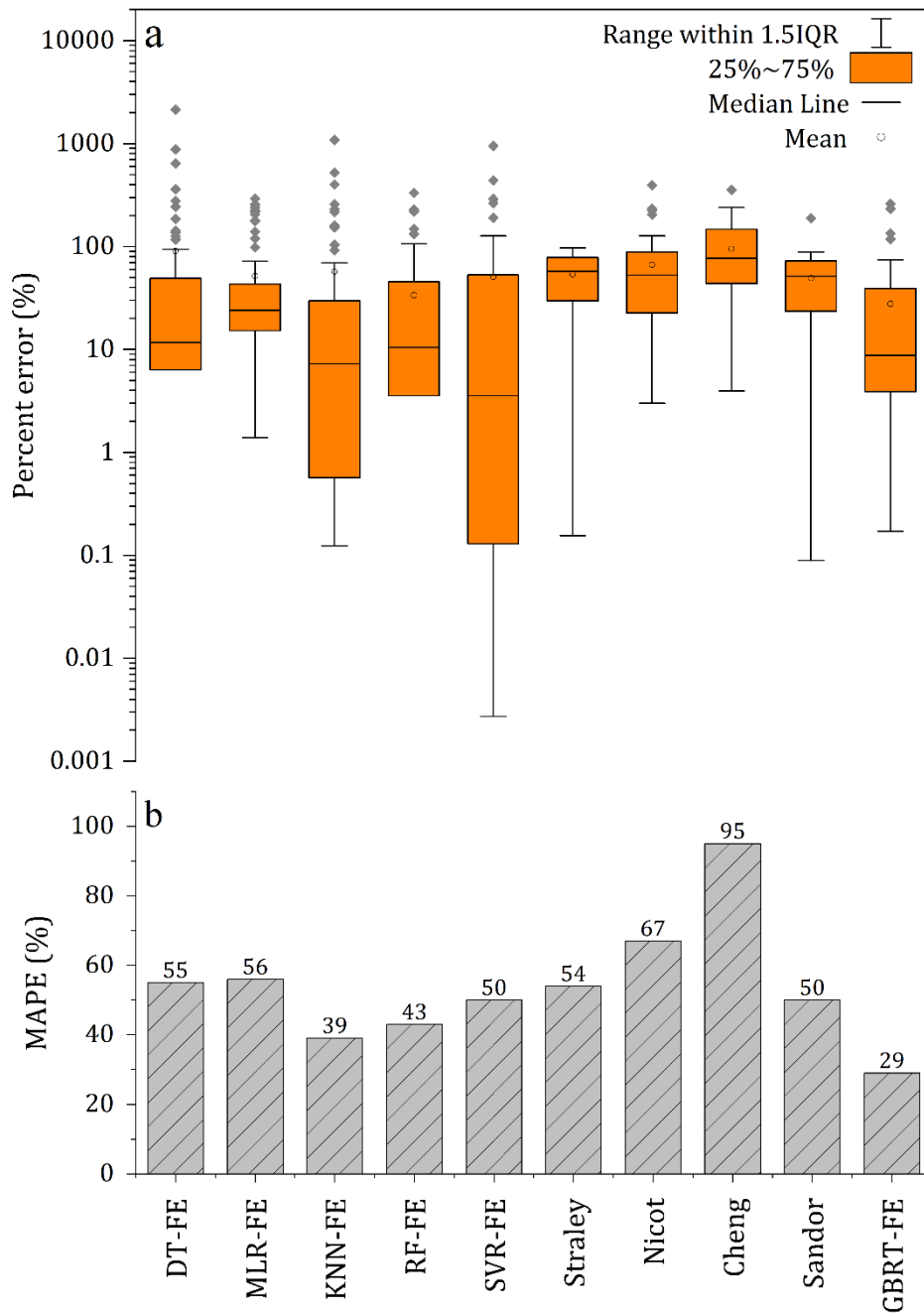


Figure 24: Percent error box plots (a) and MAPE scores (b) for six supervised learning models with feature engineering, and four empirical models. Note that in the plot (a) the y-axis is in log-scale. GBRT-FE model demonstrates the best performance in terms of MAPE.

Table 9: Compared view of statistical scores for all SL and empirical models. Bolded values correspond to the best score.

Model	Test scores				
	RMSE (cP)	MAE (cP)	MSLE	MAPE (%)	R ²
DT	20,712	7968	0.368	55	0.49
MLR	30,282	10,858	3.443	282	-0.08
KNN	25,044	8642	0.369	47	0.26
RF	21,725	6989	0.293	44	0.63
SVR	26,266	7858	0.749	93	0.40
GBRT	10,587	3331	0.168	32	0.85
DT-FE	20688	7409	0.359	55	0.49
MLR-FE	15,808	5447	0.319	56	0.70
KNN-FE	14,559	4182	0.210	39	0.74
RF-FE	18,285	5979	0.232	43	0.79
SVR-FE	5418	1671	0.257	50	0.96
GBRT-FE	8704	3377	0.136	29	0.91
Straley	28,910	9638	1.014	54	0.58
Sandor	25,012	8066	0.831	50	0.83
Nicot	21,489	7306	0.712	67	0.46
Cheng	21,371	7085	0.990	95	0.59

3.3.4. PHYSICAL IMPLICATIONS OF SVR-FE AND GBRT-FE PERFORMANCE

From the physical point of view, meaningful insight can be extracted from the models' fundamental understanding. Although SVR-FE is moderately stable and achieves good accuracy, it struggles with capturing additional variability from the oil samples' diverse chemical compositions. This behavior can be observed in Figure 20f, where SVR-FE predictions favor the high-temperature heavy oil sample over the other samples. This occurs due to the structural risk minimization principle, which balances model complexity to avoid overfitting the training data and ensures the best possible generalization of new data. In other words, the model can be adapted to perform with more precision, but that would likely

deteriorate its generalization ability. However, two possible strategies could rectify this; firstly, SVR heavily relies on feature engineering, which implies that the SVR training on a set of lighter or more chemically alike oils would improve the forecasts' precision by preserving good generalization. The second strategy would be to expand the database by adding more NMR data from new samples.

On the other hand, GBRT-FE effectively handles the discrepancies from chemically diverse set oil samples and a wide span of temperature and viscosity. This is due to its stage-wise estimator addition principle, where overfitting is controlled by tuning the learning rate and restriction of tree sizes. In this way, the GBRT hyper-parameters limit individual trees' contribution, but by adding many estimators, the model manages to "learn" nuanced relationships that stem from mixed oil chemistry, thus outperforming the SVR approach. As a result, GBRT-FE achieves the best tradeoff between variance and bias for the task at hand at a negligible increase in computational costs.

On another note, models presented in this study have certain limitations originating from (a) NMR hardware configuration and (b) data availability.

- a) One of the limitations of the presented SL models is that they were trained on NMR oil data acquired with echo-spacing (TE) ranging from 0.1-0.3 ms. Thus, the NMR data acquired using older NMR tools where echo-spacing (TE) values are hardware-limited to longer TE (0.3-1.2 ms) might have less reliable predictions. Reliability might be particularly problematic for heavy oils and bitumens, where due to the fast relaxation of solid-like constituents, the NMR device fails to measure the whole T_2 -relaxation spectra, which would cause the models to underpredict the real viscosity^{23,67,96}. However, by adding new NMR data to the dataset acquired using longer TE, preferably from heavy oil samples, the SL algorithm could capture the relationship between long TE and viscosity, compensating for the undetected part of the T_2 spectra.

- b) Small datasets are very common in petrophysics, especially NMR data, due to the confidentiality regulations of oil companies and high well-logging costs, making the application of artificial intelligence challenging. Additional data acquired from heavier oils and at various temperatures would make these models more robust to the chemical diversity of oils and various temperature conditions.

3.4. SUMMARY

In this study, we used SVR and GBRT algorithms to develop NMR models for oil viscosity prediction using NMR T_2 -relaxation time, echo-spacing and temperature as an input, and dynamic oil viscosity as the target output. Also, a strategy to reduce the variance of the forecasts was introduced, where domain knowledge was used to implement feature engineering. Model performance was assessed against four other popular SL algorithms and another four analytical models from the literature. The SVR-FE and GBRT-FE have achieved statistically most favorable scores in the study in terms of five error metrics: RMSE, MAE, MSLE, MAPE, and \bar{R}^2 .

In summary, GBRT-FE demonstrated the best overall generalization ability, thus producing predictions with a well-balanced variance-bias tradeoff. Consequently, the use of GBRT-FE might prove as a viable solution in circumstances where a wide span of oil types (light oils, heavy oils, and bitumens) is being tested at various temperatures. Environments like these correspond to heavy oil reservoirs undergoing or being screened for thermal treatment and other EOR approaches such as solvent injection or a miscible gas injection¹¹⁰. On the other hand, the SVR-FE model exhibited a high accuracy but could not account for the variability originating from the diverse chemical composition of the oils at the level that the GBRT-FE model did. These findings indicate that SVR-FE would be a better choice when sets of chemically more similar oils are being studied (e.g., only light or only heavy oils) at various temperatures. In such conditions, the variance of SVR-FE

predictions would reduce to the degree where high accuracy and precision come into play, such as in laboratory NMR characterization of petroleum fractions or contactless non-invasive oil viscosity monitoring in mechanical systems.

Finally, the proposed strategy for supervised learning application proved to be effective even for a small dataset, suggesting that this approach can be extended to characterize other physicochemical properties of oils, fuels, and petroleum distillates researchers work with relatively smaller datasets.

Chapter 4 APPLICATION OF XGBOOST MODEL FOR *IN-SITU* WATER SATURATION DETERMINATION IN CANADIAN OIL-SANDS BY LF-NMR AND DENSITY DATA

4.1. MOTIVATION

As discussed in section 1.5.3, the main issues related to the determination of water content in oil-sands by LF-NMR are:

- Insufficient understanding of dominating T_2 relaxation mechanism in fine pore-space saturated by fluid (T_2 bulk + T_2 surface);
- The diffusive-coupling phenomenon associated with the water relaxation between macro- and micro-pores, and;
- Resultant overlapping of water and oil signals in T_2 distribution.

As these issues are intertwined, the determination of T_2 cutoffs for splitting the T_2 distribution to producible and bound fluids and interpretation of fluid types becomes a laborious task in which seemingly minor errors can lead to erroneous predictions of water saturation and, therefore of OOIP.

In this work, we postulated that the combination of LF-NMT T_2 data and bulk density data could be used to effectively separate the contributions of oil and water signals to a degree at which an accurate determination of relative water fraction is possible. For model derivation, two machine learning approaches based on Extreme Gradient Boosting (XGB) were employed. The first modeling approach is based on a feature engineering process that reduces the number of inputs while maximizing model generalization capacity. This was achieved by deriving new features using empirical knowledge from the T_2 distribution analysis domain and a feature extraction technique based on information theory. In contrast, the second approach considers as input the whole NMR T_2 distribution of the sample, aiming to preserve all available information originating from fluids residing in the sample pore space. The dataset comprised 82 oil-sands core samples recovered

from northern Alberta in Canada. Water content percentage relative to of the total mass of the sample was determined by Dean-Stark extraction (%DS-w). The model training and prediction test scores of the models were evaluated using three statistical metrics and a leave-one-out cross-validation (LOOCV). These scores were compared with water content predictions based on the previously published deconvolution approach⁵¹.

4.2. THEORY

4.2.1. LF-NMR MEASUREMENTS FOR WATER SATURATION DETERMINATION

Three main processes comprise the total T_2 relaxation; bulk relaxation, surface relaxation, and diffusion relaxation due to the gradient in a magnetic field. In this work, the benchtop LF-NMR relaxometer was used in which the gradient is absent, thus the diffusion term can be neglected.

$$\frac{1}{T_2} = \frac{1}{T_{2B}} + \frac{1}{T_{2S}} \quad (49)$$

$$\frac{1}{T_{2S}} = \rho_2 \left(\frac{S}{V} \right) \quad (50)$$

T_{2B} represents the relaxation occurring in bulk fluids or fluids in large pores, and T_{2S} quantifies the relaxation of fluids in smaller pores. Also, ρ_2 is T_2 surface relaxivity, and S/V is a ratio of the fluid volume and surface of the pore. Each of these mechanisms will contribute to the total relaxation in varied proportions depending on reservoir rock properties and physicochemical properties of the fluids, such as rock wettability, pore size and pore surface area, fluid viscosity and chemical composition.

4.2.2. XGBOOST PRINCIPLES

XGBoost stands for Extreme Gradient Boosting (XGB), and it presents an implementation of the gradient boosting decision trees¹¹¹. The main principle of gradient boosting is to utilize the individual weak learner, such as a decision tree, and in a stage-wise manner, add iteratively new trees, to minimize further the objective function. This process continues for the specified number of boosting iterations, after which the prediction model is obtained in a final form. The

algorithm uses gradient descent to minimize the objective function by finding the direction of the “steepest” descent. XGBoost, on the other hand, employs several improvements resulting in better overall generalization and computational speed. Some of the key advantages are using second-order gradients, which contribute to a better understanding of the direction of the loss function minimum, and enhanced regularization techniques such as lasso regression (L1) and ridge regression (L2) which reduce the model complexity and overfitting. The XGBoost model can be expressed as:

$$\hat{y}_i = \sum_{k=1}^K f_k(x_i) \quad (51)$$

where \hat{y}_i is predicted Dean-Stark water content (%DS-w), x_i is a vector of input features and f_k is a tree at the k -th instance. A new tree f_t is added iteratively by minimizing the objective function as:

$$f_t = \sum_{i=1}^n L(y_i, y_i^{(t-1)} + f_t(x_i)) + \Omega(f_t) \quad (52)$$

where L presents the specified loss function, y_i is observed %DS-w in a sample, $\hat{y}_i^{(t-1)} + f_t(x_i)$ is the predicted %DS-w at the $t-1$ iteration, and Ω is a regularization term, or a penalty function. Regularization term can be denoted as:

$$\Omega(f_t) = \gamma T + \frac{1}{2} \lambda \sum_{j=1}^T w_j^2 \quad (53)$$

where γ is L1 and λ is L2 regularization parameters, T is the number of leaf nodes in a tree, and w_j are the weights of leaves. The addition of new trees f_t is performed in a stage-wise manner such that the loss between the prediction and observation is minimized, with respect to the regularization term Ω to prevent the overfitting and gauge the model complexity. Smaller values of Ω enable the better generalization of a tree. The detailed mathematical description of the XGBoost algorithm and additional tuning and regularization parameters is available elsewhere¹¹¹.

4.3. METHODOLOGY

4.3.1. EXPERIMENTAL PROCEDURE AND DATA PREPROCESSING

Oil-sand samples were collected in northern Alberta in Canada from a single delineation well. Two sets of 82 whole core samples were recovered. The first set was used for laboratory LF-NMR measurements, and a second set represented sister samples used in Dean-Stark extraction for determining the relative fraction of water, oil, and solids. Samples for NMR experiments were stored in glass vials and measured using a Corespec 1000™ benchtop LF-NMR relaxometer at reservoir temperature (6 °C) and ambient temperature (25 °C). The Carr-Purcell-Meiboom-Gill (CPMG) pulse sequence was used for obtaining T₂-relaxation distribution. The CPMG parameters were predetermined after a series of test NMR experiments on different oil-sand samples. There were two aspects which had to be taken into account. The first was to tune the CPMG parameters to detect the fast relaxing heavy oil and clay-bound water signals. This was achieved by setting the shortest echo time TE that the equipment allowed (0.2 ms). The second aspect was achieving a lower signal-to-noise ratio (SNR) to simulate the well-logging *in-situ* NMR tool output by reducing the number of trains, which in turn resulted in a noisier signal. After trial rounds of measurements, the following parameters produced optimal T₂ distribution and SNR (Table 10).

Table 10: Optimal CPMG pulse sequence parameters for detection of fast relaxing clay-bound water and heavy oil signals.

CPMG pulse parameters	Values
Echo time, TE (ms)	0.2
Number of pulses, N _p	5000
Wait time/post train delay (ms)	6500
Number of trains, NT	10

For the dataset, the range of SNR varied from 5 to 56, with an average of 23. The ExpFit in-house software for multi-exponential analysis of the NMR signal was

used. The representation of the signal after Inverse Laplace Transform (ILT) was obtained using Tikhonov regularization¹¹². The practice has shown that the regularization parameter helps avoid oscillations in solution associated with noise and provides smooth T_2 distributions⁸. The regularization parameter can be determined by direct and indirect methods such as Butler-Reed-Dawson, L-curve, or generalized cross-validation⁸. In the case of oil-sands, after initial analysis, the regularization parameter was determined directly and $\alpha=0.05$ was found to provide the most stable solution for most samples. The density values of these samples were measured beforehand by X-ray Computed Tomography (X-ray CT) using GE 9800 CT scanner as a substitute for the density logging data.

The experimental program for Dean-Stark extraction, LF-NMR measurements, and X-ray CT density measurements is illustrated in the flowchart (Figure 25).

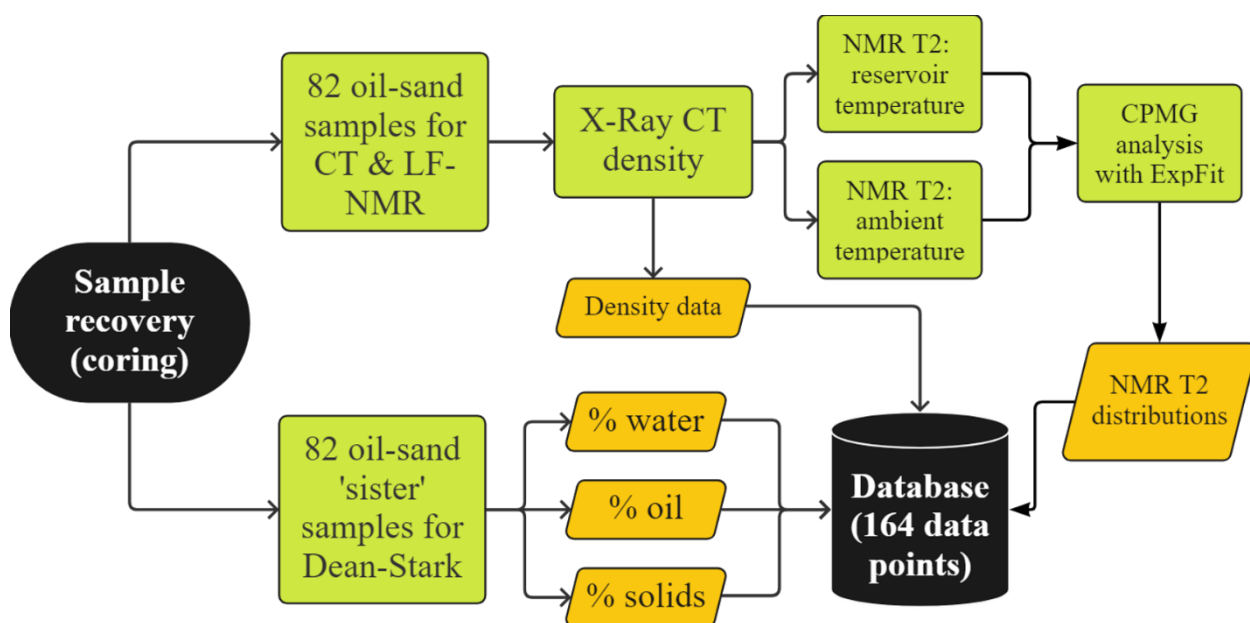


Figure 25: Flowchart representing the experimental program for oil-sands samples by X-Ray CT, LF-NMR T_2 measurements, and Dean-Stark extraction.

The final size of the dataset comprised 164 data points – 82 T_2 distributions at ambient temperature and 82 T_2 distributions at reservoir temperature, with corresponding density data and Dean-Stark sample composition. To compare the

performance of machine learning models with the well-known peak deconvolution approach, the prediction of water content by LF-NMR measurements was also performed using the T_2 cutoff approach developed by Bryan et al.⁵¹.

The data processing and model training was performed in Python 3.9 environment, while figures were produced using OriginPro 2019b software. For XGBoost model development and training, the dataset was randomly split into a training set and a test set in 0.25:0.75 proportion, respectively. To ensure the reproducible split of the data, a random split seed was fixed to *random_state = 2*. The XGBoost models were optimized using Bayesian Optimization (BO), while the training quality was evaluated by leave-one-out cross-validation (LOOCV). The forecasting performance of the models was evaluated using three error metrics and residual distribution analysis. These steps will be discussed in detail in the following sections.

4.3.2. XGBOOST MODEL BASED ON FEATURE ENGINEERING (XGB-FE)

Feature engineering (FE) is a process in a part of a machine learning pipeline where domain knowledge is utilized to extract the most relevant information from the raw data. In this work, we used feature engineering to extract information from the NMR T_2 -relaxation distribution. The complete FE model derivation procedure is illustrated in Figure 26.

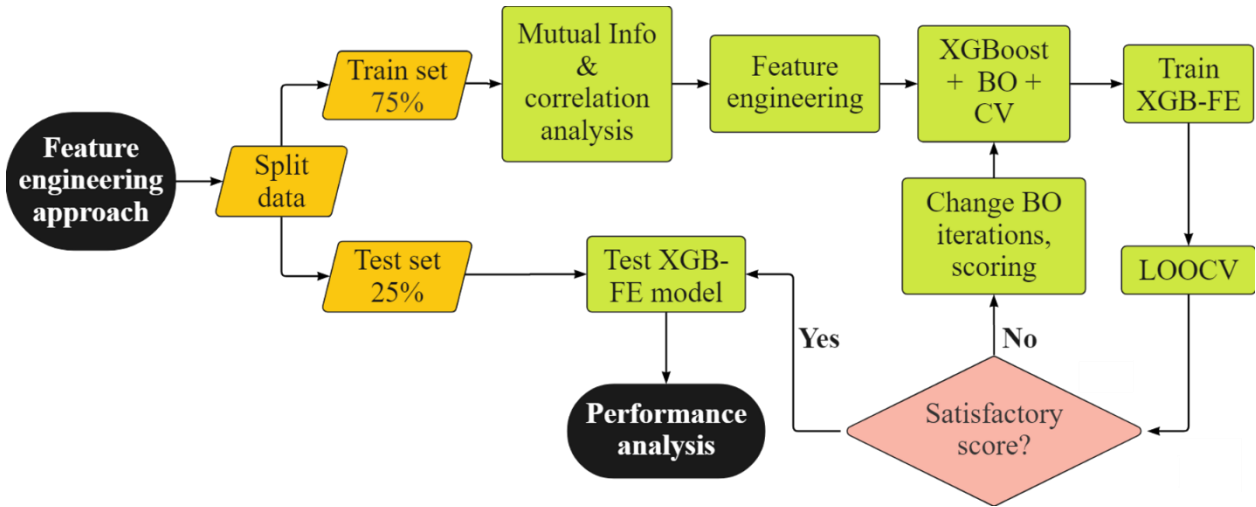


Figure 26: Flowchart for XGB-FE model development.

In petrophysics, the T_2 -relaxation is regularly analyzed by geoscientists to determine fluid saturations in reservoirs, differentiate between different types of fluids, study pore size distribution, and evaluate the physicochemical properties of fluids. However, depending on the task, some parts of the T_2 distribution may have more relevance than others. In the context of studying the water content in oil-sands by NMR, we use feature engineering to reduce the amount of unnecessary information while preserving the data carrying the most information about the water in samples. A time-domain distribution of the T_2 -relaxation was obtained by processing the spin-echo signal decay using a mathematical inversion. As the T_2 distribution has a form of a continuous function, the discretization was performed for data binning which simplifies the input of data into the machine learning model. After the discretization, the T_2 data was presented as a frequency distribution by 52 bins, with each bin corresponding to a particular T_{2i} relaxation time in milliseconds. We defined five new NMR T_2 features to limit the number of inputs.

As the T_2 distribution of relaxation times is represented on the semi-logarithmic scale, the standard parameter for representing the average T_2 relaxation is T_2 logarithmic mean (T_{2lm}):

$$T_{2lm} = \exp \left[\sum \frac{A_i}{A} \cdot \ln(T_{2i}) \right] \quad (54)$$

where A_i is an amplitude at the corresponding T_{2i} bin, and A is a total NMR amplitude. Empirical evidence shows the strong relationship between viscosity of fluids and T_{2lm} , implying that in a water-oil system where distribution tends to be multimodal due to their different relaxation properties, the T_{2lm} provides a better measure of central tendency favoring both fast and slow relaxing parts of the distribution.

To account for the variation in T_2 distribution (i.e. narrow vs. wide peaks), the T_2 standard deviation was defined as:

$$T_{2std} = \sqrt{\frac{\sum(A_i - \mu)^2}{N}} \quad (55)$$

where μ is the T_2 distribution mean, and N is the number of the T_2 bins.

The T_{2p} was defined as a location of a maximum value (peak) of the T_2 amplitude on T_{2i} axis. This parameter is used in the petrophysical practice for the separation of bound and producible fluids and fluid typing since T_{2p} gives an indication of whether the largest amplitude portion of the signal corresponds to low or high T_2 values.

$$T_{2p} = \max \left(f(T_{2_1}), \dots, f(T_{2_n}) \right) \quad (56)$$

Intelligent algorithms like XGBoost have gained popularity due to their ability to generalize complex data dependencies in large datasets and achieve state-of-the-art forecasting results. However, a small dataset is used in this study, where the overlapping of water and oil T_2 signals are likely to remain hidden or poorly represented. So, instead of allowing the algorithm to search through the whole NMR T_2 distribution, we can 'show' it where to look for the patterns and changes in the amplitude. One of the essential parts of the T_2 distribution in sandstones is the empirical clay-bound water T_2 cutoff located at 3 ms, which presents the boundary between capillary-bound and clay-bound fluids in a water-saturated core^{6,113}. In order to capture the possible T_2 response of clay-bound water and

monitor its signal variation with different training samples, we defined a T₂ bound fluid (T_{2bf}) interval as:

$$T_{2bf} = \sum_{0.1(ms)}^{3.0(ms)} A_i \quad (57)$$

However, this parameter cannot be used on its own to describe the changes in water content since the oil signal may also be located in the relevant interval. The true T₂ cutoff value in petrophysical practice is usually determined by performing lab tests on the saturated core samples (i.e., centrifuging), and even then, the use of a fixed or averaged T₂ cutoff value leads to the erroneous prediction of producible fluids. Instead, we attempt to obtain insights into the true T₂ cutoffs using a feature extraction technique called Mutual Information (MI) regression, based on the information entropy between variables. In classical regression analysis, statistical tests like F-test are carried out to study the degree of the linear association or continuous analysis of covariance (CANOVA) for the non-linear association between variables. However, mutual information is not ‘concerned’ whether the variables have apparent linear correlation or covariance of zero, and they may still be stochastically dependent. This is the case in studying the changes in the conditional probability of one variable when another is modified¹¹⁴. In other words, by using MI regression, one can measure the level of association of the specific parts of T₂ distribution with the target output (i.e., water content by Dean-Stark), regardless of their correlation or covariance. The score is measured in natural units of information or ‘nats’ based on natural logarithms and powers of *e*. The MI regression was performed on the training set using a Python library `sklearn.feature_selection` class `mutual_info_regression`.

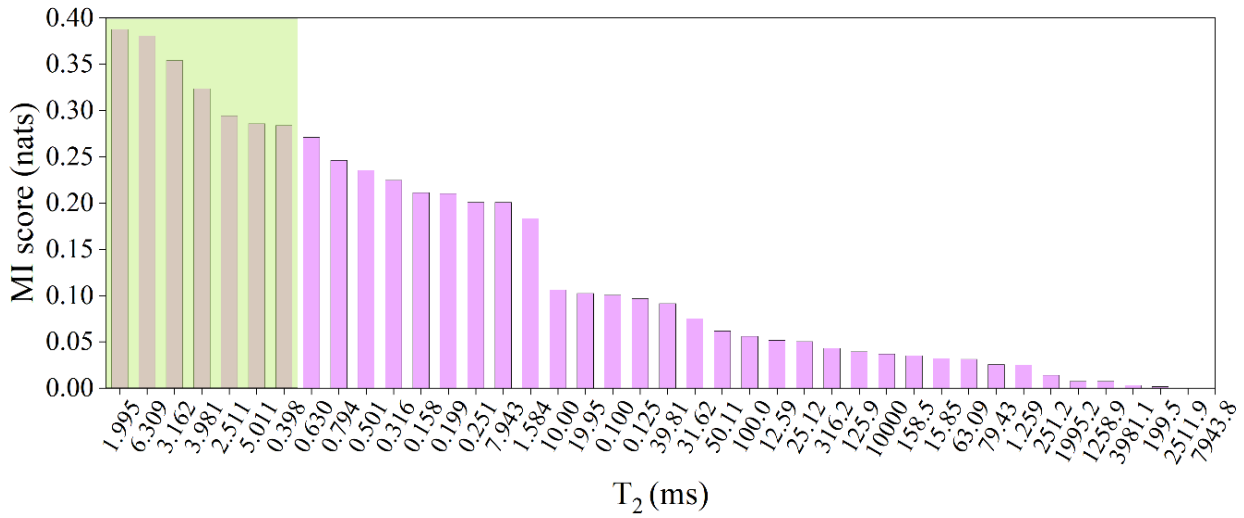


Figure 27: The mutual information regression results applied to the training set T_2 distributions of the oil-sand samples relative to the Dean-Stark water content (DS-w). The shaded area presents the continuous cluster of T_2 responses with a strong mutual association with DS-w, which were used to calculate the T_2 cutoff range parameter – T_{2cr} .

Figure 27 shows the relative mutual information scores of T_2 responses, where higher values indicate a stronger association with water content by Dean-Stark. For this dataset, the responses from 1.99-6.30 ms have the highest association with the water signal and form a continuous cluster between 10^0 and 10^1 decades along the T_2 semi-log scale, suggesting that most theoretical T_2 cutoff values lie in this interval. Therefore, the T_2 cutoff range (T_{2cr}), was defined as:

$$T_{2cr} = \sum_{1.99 (ms)}^{6.30 (ms)} A_i \quad (58)$$

As previously mentioned, the T_2 surface relaxation and diffusive coupling play a vital role in identifying clay-bound water, which causes the overlapping of the water and oil signals. Unfortunately, to determine their contribution, a sample recovery for the subsequent lab experiments is required. However, a common practice in well-logging is to combine NMR and bulk density logs to improve interpretation. Therefore, the bulk density was used as an additional parameter which we postulate is associated with T_2 surface relaxation and diffusive coupling.

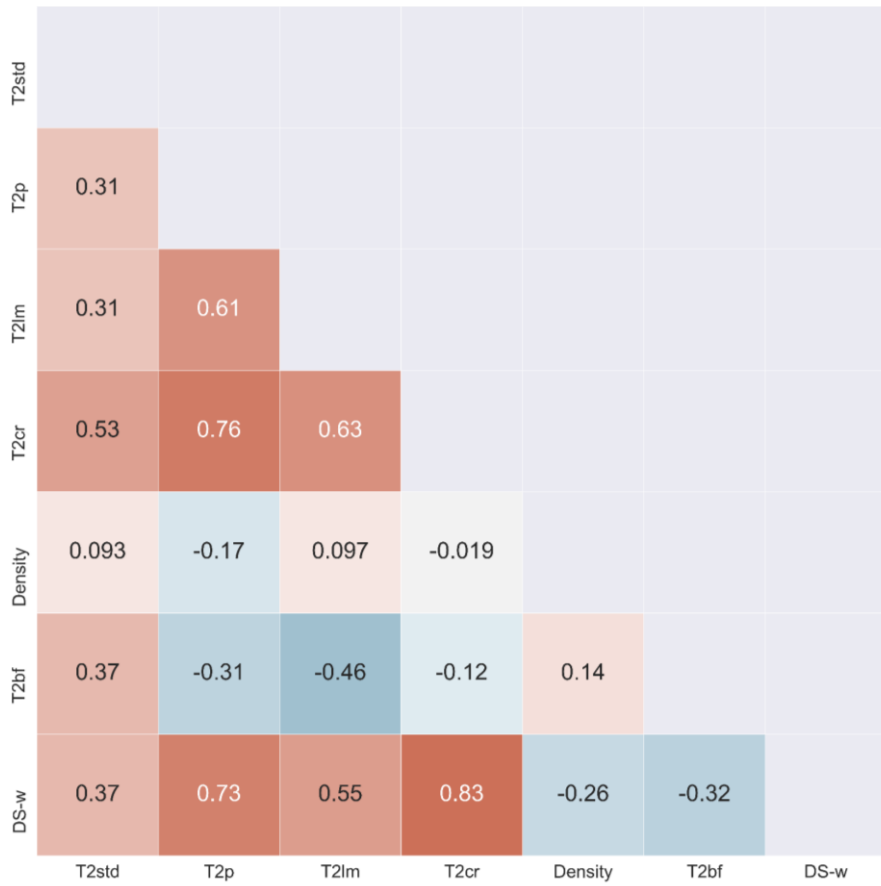


Figure 28: The diagonal correlation matrix shows the linear dependence between six input features with Dean-Stark water content (DS-w). Scores represent Pearson's correlation coefficient and are color-coded (heatmap).

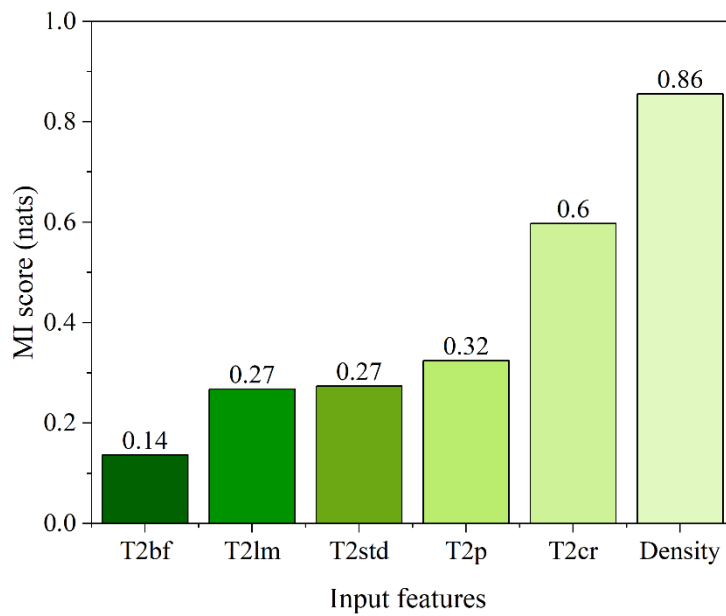


Figure 29: Mutual information regression scores for five NMR parameters and bulk density (input features) relative to the Dean-Stark water content (DS-w).

The correlation matrix (Figure 28) shows the linear dependence between the input features and target output. According to the Pearson score, T_2 cutoff range and T_2 peak, and T_2 logarithmic mean features exhibit the strongest positive correlation with the water content by Dean-Stark (DS-w). The T_2 standard deviation shows a moderate degree of positive correlation, while T_2 bound fluid and density features show moderate to low negative correlation with DS-w. Interestingly, when compared with mutual information scores from Figure 29, it can be observed that all features are ranked by score accordingly to Pearson's scores except for density which has the highest MI score (0.86 nats), indicating its strong stochastic (nonlinear) dependence with DS-w, thus justifying integration of density measurements into the model. Therefore, the XGB-FE model was developed using the six features presented in Table 11.

Table 11: Descriptive statistics of six input features used for XGB-FE model development.

Statistic	T_{2std} (a.u.)	T_{2p} (ms)	T_{2lm} (ms)	T_{2cr} (a.u.)	T_{2bf} (a.u.)	ρ (kg/m³)
Count	164,0	164,00	164,00	164,00	164,00	164,00
Mean	0,016	11,57	1,84	0,18	0,31	1626
Std	0,005	4,54	1,22	0,13	0,10	80
Min	0,006	1,00	0,32	0,00	0,10	1442
25%	0,013	8,00	0,90	0,07	0,23	1581
50%	0,016	13,00	1,61	0,18	0,30	1634
75%	0,019	15,25	2,49	0,29	0,38	1677
Max	0,032	20,00	8,59	0,50	0,54	1842

4.3.3. XGBOOST MODEL BASED ON THE FULL T_2 RELAXATION DISTRIBUTION (XGB-FS)

The second modeling method facilitates the complete sample T_2 distribution. There are two main incentives for this approach. First, the T_2 relaxation distribution contains a large amount of information about the fluids residing in the pore space, indicating that using a single or even a few features to characterize the whole distribution may lead to significant information loss and, therefore to poor model forecasting performance⁶⁵. By using the entire T_2 distribution, variations such as changes in slope or local minima can implicitly be used to help separate oil and water signals. Secondly, predictions generated by the full- T_2 distribution model provide a good baseline for comparison with the feature engineering and conventional deconvolution approaches. Therefore, the input features were arranged as $X = [A_1, A_2, A_3, \dots, A_{52}, \rho_i]$, where A_i is the i -th column vector of the amplitudes at the corresponding T_{2i} bin, and ρ_i is a column vector of density measurements. The water content by Dean-Stark (DS-w) was arranged as $Y = [DS-w_1, DS-w_2, \dots, DS-w_n]$, thus defining the dataset as $\{(X_i, Y_i)\}_{i=1}^n$ where n is the number of oil-sands samples. The complete XGB-FS model derivation the procedure is illustrated in Figure 30.

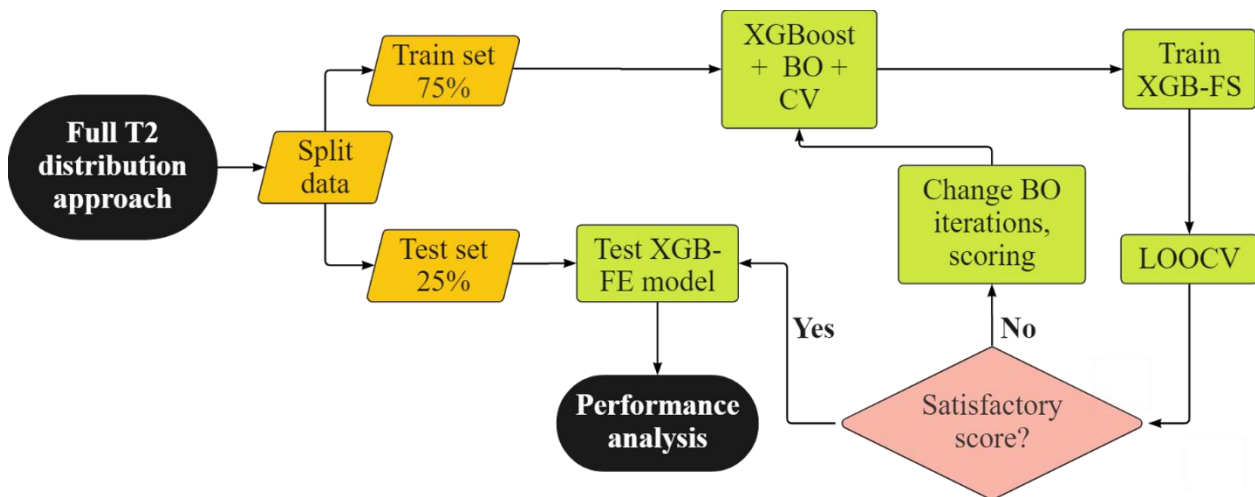


Figure 30: Flowchart for XGB-FS model development.

4.3.4. MODEL OPTIMIZATION

The XGBoost algorithm contains many hyperparameters which enable fine model tuning. From the standpoint of statistical learning, the tuning usually involves the use of iterative algorithms which search for a suitable combination of hyperparameters in real-valued parameter space relative to the specified measure of model forecasting performance (e.g., mean squared error). However, as the number of parameters grows, the optimization becomes computationally expensive due to the combinatorial explosion, making the manual optimization or exhaustive grid searching techniques inefficient. In contrast, Bayesian Optimization (BO) sets a probabilistic approach where each successive combination of hyper-parameters is selected based on the information obtained in the previous optimization step, thus avoiding the redundant calculations for unlikely parameter combinations and reducing the number of required iterations to reach the global minimum of the objective function.

The BO was performed in Python using scikit-optimize package class `skopt.BayesSearchCV`. The hyper-parameters and their optimal values are presented in Table 12.

Table 12: Results of Bayesian Optimization with 5-fold cross-validation, for XGB-FS and XGB-FE models.

XGBoost hyperparameters	Search range	XGB-FS optimal	XGB-FE optimal
n_estimators	[50-1000]	[650]	[300]
learning_rate	[0.004-0.1]	[0.008]	[0.053]
subsample	[0.7-1.0]	[0.7]	[0.6]
max_depth	[6-12]	[8]	[7]
objective	['squared_error', 'pseudo_huber']	['pseudo_huber']	['squared_error']
grow_policy	['depthwise', 'lossguide']	['lossguide']	['lossguide']
booster	['gbtree', 'dart']	['gbtree']	['gbtree']

4.3.5. PERFORMANCE METRICS AND MODEL VALIDATION

The forecasting performance of the models was evaluated using three performance metrics, including coefficient of determination (R^2), root mean squared error (RMSE), and mean absolute error (MAE). The R^2 is the positively oriented metric used in regression for representing the amount of model variance, and how well the model predictions generalize the observations. However, R^2 alone does not provide information on prediction errors. The RMSE is another regularly employed error metric, used alongside R^2 , but under the assumption that residuals follow the normal distribution¹¹⁵. As a result of the heavy penalization of larger residuals, the RMSE is a convenient metric for revealing the differences in performance between multiple models with normally distributed residuals. At the same time, large residuals can cause the inflation of the RMSE score, which is why MAE can be used for additional evaluation. The MAE measures the mean magnitude of model prediction errors, but in contrast to RMSE, the errors are not squared. Therefore RMSE scores are always greater or equal to MAE scores. These two metrics can be used together to estimate the variation in errors, where $RMSE = MAE$ indicates no variation in the magnitude of prediction residuals. Recall that both RMSE and MAE are negatively oriented scores expressed in %DS-w.

$$R^2 = 1 - \frac{\sum_{i=1}^n (y_i - \hat{y}_i)^2}{\sum_{i=1}^n (y_i - \bar{y})^2} \quad (59)$$

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (y_i - \hat{y}_i)^2} \quad (60)$$

$$MAE = \frac{1}{n} \sum_{i=1}^n |y_i - \hat{y}_i| \quad (61)$$

where y_i is predicted %DS-w, \hat{y}_i is observed %DS-w, \bar{y} the sample mean, and n presents the number of samples.

The further model performance evaluation and validation were performed using leave-one-out cross-validation (LOOCV) due to its convenience for use on small datasets (Figure 33). Cross-validation is a resampling method in which the sample subsets are drawn repeatedly from the training set, followed by model refitting for each subset, thus providing information on model fitting variability. In LOOCV, the samples are drawn for one observation at a time, while the rest of the data is used for model training. Therefore, this process has a number of iterations equal to the number of samples, making it computationally expensive for large datasets. In addition to LOOCV, the permutation tests were conducted to assess the significance of 5-fold cross-validated model prediction scores with 150 random permutations. This enabled the evaluation of the statistical significance of model predictions and their inputs by a permutation test P-value.

4.4. RESULTS

In this section, the performance of three models is presented, including the XGB-FE model based on the XGBoost algorithm with feature engineering, the XGB-FS model based on the XGBoost algorithm using the whole sample T₂ distribution, and a peak deconvolution approach (Bryan et al⁷). To assess the model performance in more detail, residual plots (Figures 30-2, 30-5, and 30-8) and quantile-quantile plots (Figures 30-3, 30-6, and 30-9) are used for the analysis of the residual normality, and model variance and bias. All results are summarized in Figures 30-32.

Analysis and comparison of error statistics, cross-plots, and distribution of residuals indicate that the XGB-FE model achieves the highest accuracy and generalization ability in the prediction of water content in oil-sand samples. Figure 31-1 shows that apart from slight overprediction in the 3-5% DS-w range, all XGB-FE predictions spread along the x=y line with low variance, achieving the highest R² score in the study (R²=0.90). Figure 31-2 shows the constant low variance of the residuals, indicating that the model inputs capture variation in the data

correctly. Finally, the Q-Q plot (Figure 31-3) confirms the residual normality and thereby the underlying assumption that XGB-FE model residuals follow the normal distribution (low bias, low variance). Finally, the XGB-FE model achieves 1.5-3 times lower RMSE and MAE scores compared to the XGB-FS and Bryan et al.⁷ models indicating the best generalization ability of the three.

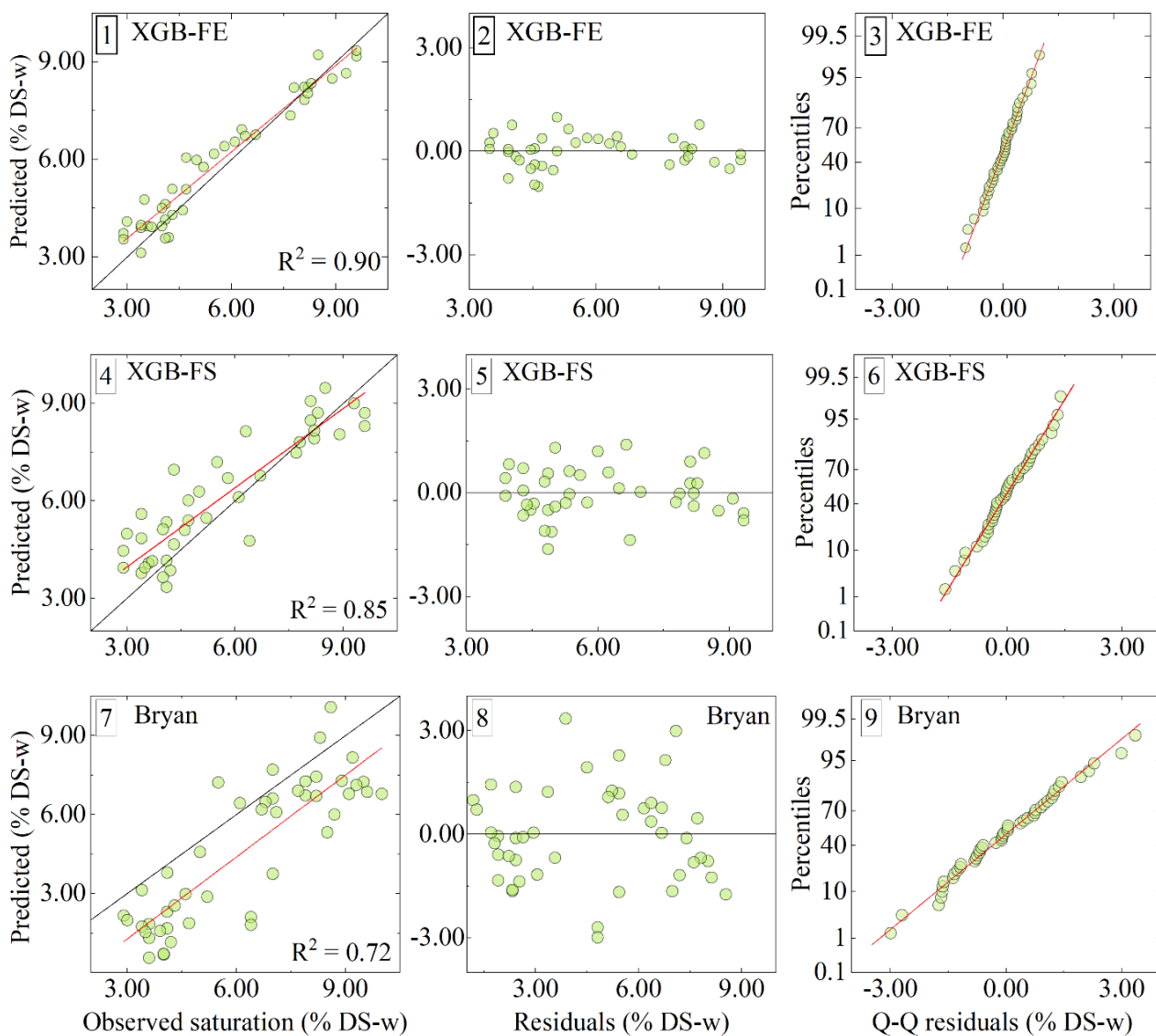


Figure 31: Evaluation of XGB-FE, XGB-FS, and Bryan et al.⁷, performance by cross-plots between the model predictions and observed saturation in %DS-w (1, 4, 7), distribution of regular residuals (2, 5, 8), and quantile-quantile plots for comparing distributions between predictions and observations and evaluating normality of residuals (3, 6, 9).

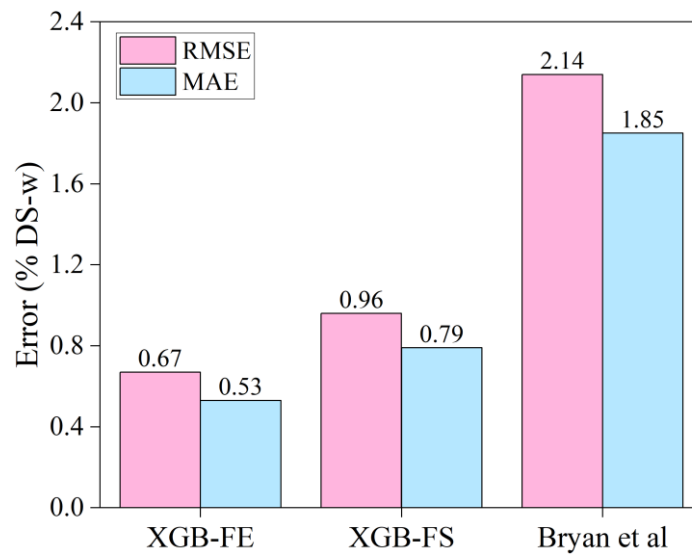


Figure 32: Comparison of RMSE and MAE test prediction scores for the three models (*random_state=2*).

As for the XGB-FS model predictions, Figures 30-4, 30-5, and 30-6 show a similar residual distribution to XGB-FE (normality and bias). However, the residual variance is increased but constant, therefore achieving a somewhat lower R^2 score ($R^2=0.85$) and 1.5 times higher RMSE and MAE than XGB-FE. From Figure 31-7, it can be observed that the Bryan et al.⁷ model generally tends to underpredict the water content in samples. In addition, Figures 30-8 and 30-9 show inflated but the constant variance in the distribution of residuals, while residual normality still holds with some local perturbing. As a result, Bryan et al.⁷ model RMSE and MAE scores are the highest (Figure 32).

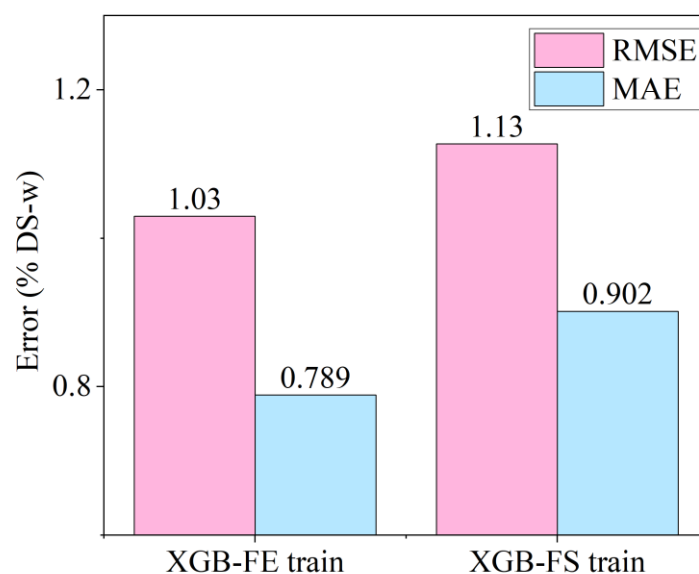


Figure 33: Leave-one-out cross-validation (LOOCV) scores for XGB-FS and XGB-FE machine learning models for the training set with fixed random split seed '*random_state=2*'. Note y-axis was truncated for convenience.

4.5. DISCUSSION

The two machine learning models in this study were designed to test two principal hypotheses. First, to confirm that the integration of density measurements into the machine learning models can help separate the contribution from overlapping oil and water signals. Second, to show that the derivation of new LF-NMR T_2 features can improve the generalization ability of the machine learning model to the degree that can enable the accurate forecasting of water content by LF-NMR in oil wells (*in-situ*).

Table 13: Comparison of XGB-FS model performance with and without *bulk density* parameter.

XGB-FS		
Statistic	wo/ Density	w/ Density
RMSE (%DS-w)	1.08	0.96
MAE (%DS-w)	0.86	0.79
R ²	0.72	0.85

Table 14: Comparison of XGB-FE model performance with and without *bulk density* parameter.

XGB-FE		
Statistic	wo/ Density	w/ Density
RMSE (%DS-w)	0.91	0.67
MAE (%DS-w)	0.73	0.53
R ²	0.81	0.90

Bulk density measurements are used together with LF-NMR measurements in petrophysical practice to improve the interpretation of well logs^{19,116}. LF-NMR measures the response of the fluids in the rock pore space, therefore carrying information about the fluids and pore size distribution of the rock. On the other hand, density logging equipment measures the response of the solids (rock matrix) together with fluids. The two are related in terms of T₂ surface relaxation, which depends on the rock pore to surface ratio with the fluid volume and the diffusive-coupling effect. This dependence can also be observed from the prediction test scores of the XGB-FS and XGB-FE models with and without bulk density as one of the model inputs. Prediction scores from Tables 13 and 14 indicate that models achieve better scores with the integration of bulk density,

therefore confirming the relationship between Dean-Stark water content and density discovered by mutual information regression.

To affirm the second hypothesis: when XGB-FS and XGB-FE are compared (Figure 31), it can be observed that XGB-FE achieves better performance, especially in terms of prediction variance. The XGB-FE variance reduction supports two premises. First, the engineered features properly capture all the relevant information from the T_2 distribution, indicating negligible information loss. Secondly, in the feature engineering case, the XGBoost algorithm generalized the variability in the data with the output more effectively, suggesting that for smaller datasets, the appropriate feature engineering enables the XGBoost algorithm to discover dependencies within the data more effectively than for a large number of raw information (53 features in case of XGB-FS). In other words, the new features contain all the relevant parts of the T_2 distribution compressed into a few values, which ultimately reduces the XGB-FE model complexity and enables better generalization of the relationship between inputs and a target variable (DS-Sw).

According to Figure 29, along with a bulk density (MI=0.86 nats), the T_2 cutoff range feature ranks second by MI score (0.60 nats), indicating the variability of the sum of T_2 responses between 1.99 – 6.30 ms has a strong relationship with water signal. The location of the T_2 peak (T_{2p}), T_2 standard deviation of the spectrum (T_{2std}), and T_2 logarithmic mean (T_{2lm}) achieve similar MI scores (0.27 and 0.30 nats, respectively), signifying that these features alone do not capture enough information about the water content. Finally, the sum of T_2 responses representative of the empirical clay-bound water part of the T_2 distribution (0.1-3.0 ms) shows the least association with the target (DS-w). Although these features alone cannot explain variance in data effectively, their mutual interaction can improve it. Since MI does not consider this mutual interaction between features relative to the target output, the correlation matrix can be used. For instance, Figure 28 shows that the T_{2cr} vs. T_{2p} and T_{2cr} vs. T_{2lm} have a strong positive correlation (0.76 and 0.63, respectively), while T_{2bf} vs. T_{2lm} have a moderate

negative correlation. These interactions are likely to be generalized in the XGB-FE model training process, thus explaining improved performance. Furthermore, the permutation test score of XGB-FE using 150 permutations generated a P-value of 0.001, compared to the XGB-FS P-value of 0.007. In both cases, the P-value is well below 0.05, showing a very low likelihood of obtaining such model performance purely by chance.

As for the deconvolution approach (Bryan et al.⁷), the main challenge lies in separating overlapping fluid contributions in T_2 distribution. Even under the assumption that T_2 cutoff and deconvolution are performed such that a precise distinction between fluid signals is possible, the issue of how to associate the amplitudes with respect to mass persists. This approach leads to underprediction of water content for the given dataset, indicating that the oil and water signals are not sufficiently separated. The machine learning-based approach is more robust because it removes the necessity to manually identify peak separation and the errors associated with visually separating oil and water signals, especially in the case of NMR measurements acquired at low SNR.

It is essential to point out the limitations of these models, which are related to reservoir lithology (a) and SNR of the measurements (b):

(a) The models presented in this study were derived for the oil-sands reservoir, so their application is limited only to similar reservoir types. However, the presented approach can be extended for use in other types of oil reservoirs under the assumption that a sufficiently large amount of observations is available.

(b) The SNR achieved by the benchtop LF-NMR relaxometers can be up to 30 times higher than the SNR values obtained using well-logging tools. In this study, the NMR signal-to-noise ratio was, on average 20, which can still be considered high relative to the logging tools where the SNR of 3-5 is considered satisfactory¹¹⁷. Although the recent research demonstrated that the XGBoost algorithm is sufficiently robust even

with noisy data¹¹⁸, an additional validation using the data obtained by the LF-NMR logging tools would be desirable. It is worth noting that, in lower SNR samples, the deconvolution approach will be even more challenging, and the value of using just the general properties of the T_2 distribution and XGBoost may be even further enhanced.

As a follow-up study, the procedures for NMR measurements with a controlled saturation and desaturation of samples, similar to those reported in recent literature⁵⁶, would enable deeper sensitivity analysis of the features derived in this work and further improvement XGB-FE model. In such a setup, the Dean-Stark measurements could be replaced by the more cost and time-effective mass-volume measurements, ultimately allowing the collection of a larger database, at which point the application of artificial neural networks (ANNs) would be possible.

It is also worth noting that logging equipment configuration can be substantially different from desktop NMR relaxometers, which may cause inconsistencies between NMR T_2 distributions obtained in the lab and the field. This can cause the variable performance of proposed NMR data-driven model, which is why the parameters of the NMR logging device, such as TW, TE and number of trains, should be relatively consistent to the values reported in this study.

4.6. SUMMARY

This study presents the approach which integrates extreme gradient boosting with LF-NMR measurements and bulk density data for the water saturation determination in oil-sands. Two models were developed using full NMR T_2 distribution (XGB-FS), and feature engineering (XGB-FE). It is concluded that;

- Feature engineering can effectively extract vital information from NMR T_2 distribution using domain knowledge and mutual information regression.
- Integrating bulk density data as a model input notably improves the XGB-FS and XGB-FE forecasting performance.

- XGB-FE achieved RMSE = 0.67%, MAE = 0.53% and $R^2 = 0.90$ in predicting relative water content by Dean-Stark, a substantial improvement compared to deconvolution method.

These results suggest that the XGB-FE model can be extended for the improved *in-situ* water saturation determination.

Chapter 5 CONCLUSIONS

This thesis focuses on nonlinear problems in petrophysical logging, such as characterization of bitumen and heavy oil viscosity and water saturation quantification, on the example of Canadian oil-sands by LF-NMR relaxometry data. To model the relationship between the NMR outputs and experimental data and, we employed various statistical and machine learning tools, which reduced uncertainties associated with data interpretation and enabled us to capitalize on new, previously unknown relationships. In conclusion, we found that:

- Heavy oil and bitumen viscosity can be analytically approximated from T_2 relaxation data, even in the high-temperature conditions, by integrating T_2 logarithmic mean, relative hydrogen index per unit volume (RHI_v), and power-law corrected T_{2lm} . The statistical scores of the new analytical model demonstrate a superior generalization ability compared to all approaches in the literature to date.
- Machine learning-based modeling (gradient boosting and support vectors in particular) using only one NMR parameter (T_{2lm}) can provide highly accurate predictions of oil viscosity. These models work well even for a set of chemically diverse light, heavy and extra-heavy oils. It also overcomes issues related to NMR hardware limitation (finite echo spacing), magnetization loss at high temperatures (Curie effect), and additional costs and uncertainties associated with the determination of RHI_v .
- The Extreme Gradient Boosting algorithm can be utilized for improved water and oil saturation evaluation with only T_2 relaxation and bulk density data required as an input. This approach bypasses issues stemming from a poor understanding of dominating T_2 relaxation processes in micro and macro-pores, diffusive coupling, and consequential oil and water signal overlapping. It also does not require determining the T_2 cutoff value and associated laboratory tests.

Even though a large number of experimental data was used for this work (hundreds of observations), the obtained datasets are considered small from the machine learning point of view (tens of thousands of observations). This work demonstrates that even relatively small datasets such as those characteristic for petrophysical laboratory tests can be used for machine learning modeling by performing feature engineering, given that the understanding of the problem and associated processes are sufficiently understood.

This work also lays the foundations for further research. The following recommendations can be made:

1. The integration of bulk density measurements into the machine learning models for viscosity determination to perform additional validation for *in-situ* applications.
2. Additional LF-NMR and bulk density measurements (dataset expansion) would further improve the understanding of T₂ surface relaxation and diffusive coupling in connected micro- and macropores. This would also enable us to use artificial neural networks with more success.
3. The combination of other conventional logging data with NMR data for machine learning modeling shows excellent potential for other *in-situ* applications such as rock wettability characterization or quantification of saturates, aromatics, resins, and asphaltenes in hydrocarbons.

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