R&D for a new concept EIC nucleon polarimeter based on chemical hyperpolarisation

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Abstract

We propose to establish the capabilities for a new technology, Chemical hyperpolarisation, to address a clear technological challenge of the EIC - accurate and rapid measurement of the degree of polarisation in the stored proton beam. Chemical hyperpolarisation is a new technology developed by Chemists to enhance the nuclear polarisation of liquid (and gaseous) media for use in medical research. We aim to develop a prototype for the world's first spin polarised liquid drop system. Such a system used with the EIC would offer rapid polarimetry information and eliminate issues with destruction of scattering media by the beam. Our R&D program will provide accurate (and first) measurement of the key parameters for hyperpolarised (frozen) liquid droplets in vaccuum - including relaxation times, degree of polarisation and drop-to-drop variations. Such data is crucial to provide a baseline for next stage development. Simulations of different ChHP polarimeter designs with realistic droplet properties and EIC beam parameters will be obtained using an adapted Geant4 simulation in which spin-spin dependencies for the key reaction processes are implemented.

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1 Motivation for the research

Chemical hyperpolarisation (ChHP) is a new technology initially developed for medical Magnetic Resonance (MR) research. It enables room temperature chemical spin-polarisation of nuclei in a liquid (or gaseous) substrate, utilising non-equilibrium chemical reactions with a catalyst and a supply of para hydrogen to replenish the polarisation. The application of ChHP in particle and nuclear physics research would offer exciting new opportunities to achieve nuclear spin polarisation in dense media - with particular and immediate impact for targets and polarimetry at the intensity frontier. Compared to traditional methodologies for polarising dense media (e.g. Dynamic Nuclear Polarisation - DNP) it can be achieved with modest power consumption, fewer staff and resources and without the need for ultra-low temperature cryogenics. Our nuclear and hadron physics research group at York is well placed to advance this field, being located next to one of the world's leading ChHP centres, the Centre for Hyperpolarisation in MR (ChyM), with whom we have existing strong links and research networks through our initial R&D studies.

ChHP polarised media is cheap to produce and can thus be used on a one shot basis, without the need to recycle through the beam. This will address known problems associated with the destruction of dense scattering media due to the intensity of the proton beams, which has been an issue for the carbon foils at RHIC, and will be exacerbated at the EIC. The ChHP process is also known to be stable to large temperature variations. Unlike traditional methodologies (such as DNP), ChHP methods do not require strong magnetic fields, either during the polarisation procedure or to subsequently hold the polarisation, a property which is extremely appealing for applications. The direction of the polarisation can be adjusted to an arbitrary direction using modest and rather simply achievable fields.

Polarised ChHP liquid media have potential in EIC physics to form the basis of a new-concept proton polarimeter capable of measuring the beam polarisation regularly and with timescales and position resolutions much smaller than the complementary polarised jet targets. The main aims of this 1 year R&D project are:

- Develop a dropper system which creates pellets of hyperpolarised fluid which enter a pipe under vaccuum - and then freeze. Measure the drop-to-drop consistency of the degree of polarisation for these frozen pellets using an existing MRI device - a key parameter in its suitability for the basis of a polrimeter. Evaluate for the first time the expected increase in relaxation time of frozen (over liquid) hyperpolarised fluid.

- Carry out simulations of prototype polarimeter apparatus using realistic EIC beam and ChHP pellet properties. Include the crucial spin-spin dependencies of elastic proton-proton scattering in the particle reaction routines in the Geant4 simulation. Develop a workable baseline design for a new concept polarimeter based on ChHP pellets and extract its key properties - measurement accuracy, measurement duration, systematics.

1.1 Outline of the Chemical hyperpolarisation process

The polarisation methodology to be used, in which we have significant experience, is Signal Amplification By Reversible Exchange (SABRE). This is a form of chemical hyperpolarisation which leaves the target molecule unchanged, allowing for a continuous build-up of polarisation. In SABRE (see Figure 1), the chemical hyperpolarisation process happens through a transient organometallic complex binding the parahydrogen and substrate to the catalyst, with the spin-order transfer occurring through J-couplings between the hydrides and the target substrates. The partially-polarised substrate molecules then dissociate from the complex and accumulate in solution, freeing up the catalyst for further spin-order transfer. Proton polarisation levels of up to 50% have been achieved using SABRE [1].

The most common SABRE precatalysts are of the form [Ir(NHC)(COD)Cl] (where NHC=N-heterocyclic carbene ligand, COD=1,5-cyclooctadiene), as in Figure 2, which undergo a hydrogenation reaction to form the active catalyst species, typically of a form similar to $[Ir(NHC)(H)_2(Sub)_3]$. Typical SABRE substrates are N-containing heterocycles. The catalyst represents a very small fraction of the molar volume of the ChHP. The dominant compponent of the ChHP fluid is the substrate. We have worked with Pyridine substrates (and a number of variants), which have comparable dilutions (fractions of polarised protons to the non-polarised protons in nuclei) as the butanol used in DNP polarised targets.

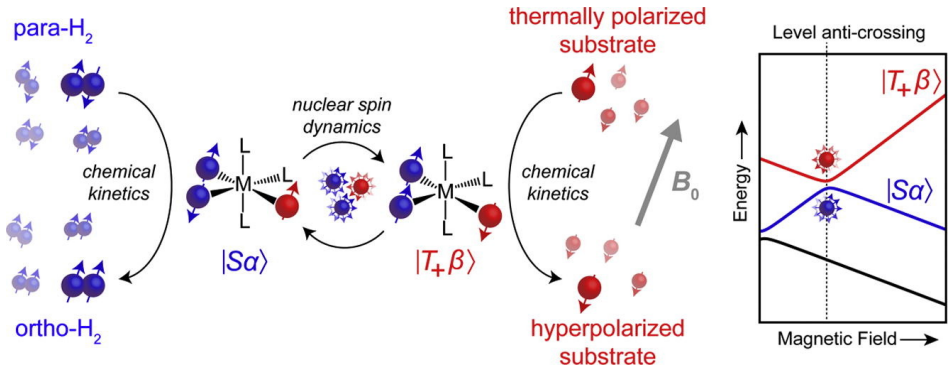

Figure 1: Reaction diagram of SABRE polarisation process [2].

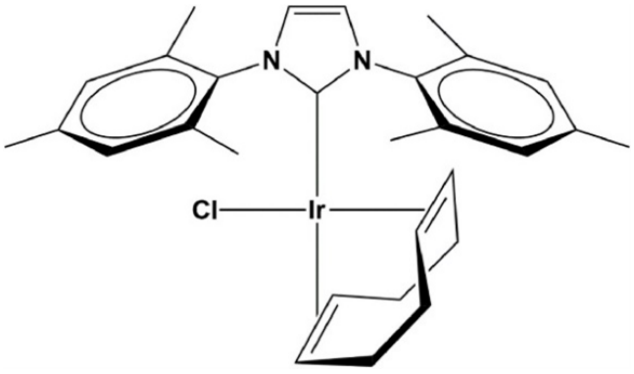

Figure 2: 'IMes' ([Ir(IMes)(COD)Cl]) SABRE precatalyst. IMes = 1,3-Bis(2,4,6-trimethylphenyl)-1,3-dihydro-2H-imidazol-2-ylidene [3].

1.2 R&D to date

In the past few years the York hadron physics group and colleagues at CHyM have made real progress in establishing the scalability of the ChHP process to larger volumes and optimising the process for its use in particle and nuclear physics. We have shown that the relaxation time of the chemically hyperpolarised protons can be extended by factors of 5 (up to almost 3 minutes), that very low catalyst concentrations are feasible, that the relaxation time is largely independent of the strength of the holding magnetic field and that the substrates used in the process can provide fast Cerenkov signals and/or scintillation light (creating the possibility of active polarised detectors). For more details on recent results relevant to this project see Appendix A.

1.3 Proton beam polarimetry at RHIC - Current techniques

Proton polarisation methods currently in use at RHIC include an absolute polarisation measurement, which utilises a polarised hydrogen gas jet [4], and a relative polarisation measurement that utilises ultra-thin carbon foils [5]. The absolute polarisation is obtained by a polarised hydrogen gas jet with low energy scattered protons from reactions of the beam with the gas jet characterised by an array of Si strip detectors. This gives precise information but requires long measurement periods to achieve adequate statistics (e.g. 0.4% stat accuracy in an 8 hour measurement). This means only the average absolute polarisation over the measurement period can be obtained. This limitation comes from the achievable count rate from the low density gas jet. Currently the relative polarisation, obtained more frequently than the absolute determination, involves scattering of the proton beam from a thin carbon foil and determining the very small spin-orbit asymmetries (of order 0.1%) in the azimuthal distributions of proton-carbon scattering. These asymmetries are measured in arrays of Si strip detectors. It is known that even with current RHIC intensities, the foils become destroyed and have to be replaced during beam periods. There is currently no proven technology for new foils to achieve these types of rapid polarisation measurements with the intensities of the EIC.

1.4 Proton beam polarimetry at the EIC - Challenges

The requirements for beam polarisation determination at the EIC are challenging, with systematic uncertainties on the order of $\frac{dp}{p} \sim 1\%$ or lower required, as specified in Section 11.9 of the Yellow Report [6]. The absolute and relative polarisation measurement methods employed at RHIC can, in principle, be utilised again at the EIC. However, the increased luminosity of the EIC presents significant challenges for both measurement methods if they are to be repurposed. In particular, the increased proton beam currents at the EIC will cause the thin carbon foils, utilised in the relative polarisation measurement, to disintegrate rapidly. The beam heating at RHIC is already an issue for the carbon foils, where they often need to be replaced during a run [5]. There is an open request from EIC management for new complementary polarimetry technologies for the EIC. This is driven by the current gap in technology to replace the carbon foil with material that is more resistant to damage. It is established that the current carbon foil technologies will fail at the EIC and current R&D

to establish materials that will not be damaged by the beam is underway.

This project has the potential to provide a complementary and entirely new methodology for relatively rapid polarisation measurements during EIC operations. It also offers new methodologies to measure the polarisation profile of the beam.

1.5 Chemical hyperpolarisation for proton beam polarimetry at the EIC

Chemical hyperpolarisation, as detailed in Section 1.1, can produce highly polarised protons (bound within the neutral substrate molecule), and we aim to create a dropper system which will produce frozen pellets of ChHP media to intersect the proton beam. Measurement of reactions dependent on the spin orientation of the protons in the beam and the pellet (which can be oriented in arbitrary direction in space on a pellet-by-pellet basis) offer a new opportunity for polarisation determination. The simplest reaction process will be elastic proton-proton scattering as exploited in the hydrogen jet polarimeter. We anticipate the ChHP pellets only need to pass through the beam once and single use pellets would have minimal cost (the substrates are typically $200 per litre) and only minute quantities of catalyst are required. However, the potential for recycling of the pellet material is also possible.

Key to the proposal is the viability of creating the ChHP pellet source. At previous beam facilities (e.g. CELSIUS, COSY), cryogenic hydrogen pellet target systems, which intersected the proton beam, have already been developed and successfully used [7, 8]. The key aims of this project are to establish if proton-spin polarised ChHP pellets can be achieved and whether these pellets have the necessary properties to form the basis of a new-concept polarimeter system. We will develop a pellet dripper system where the polarised fluid droplet enters an evacuated pipe with high, approaching beam-grade, vaccuum. The droplets will freeze [9] which will result in further extension of the relaxation time (beyond what we have already obtained for liquid media in the York labs). This would be a world-first measurement and publishable in its own right. The measurement will utilise our existing MRI imaging system to measure the polarisation and relaxation times of the droplets (see Appendix A).

A pellet target system for proton beam polarisation measurements has many potential benefits. The pellets will be produced and used rapidly, unlike with the thin carbon strip measurements currently utilised at RHIC, there are no concerns with the pellets disintegrating in the beam. The pellets themselves should have a negligible effect on the beam quality. With pellet sizes comparable to those already achieved at WASA ($\sim$ 40-60 μm diameter [8]), then a future system could offer the possiblity of measurements of the polarisation profile of the beam through variation of the interaction point. Such measurements are not possible with the polarised hydrogen jet target due to the width of the jet (FWHM $\sim$ 6.5 mm [4]). Further comparisons between the properties of the WASA pellet target and the two targets utilised at RHIC are given in Table 1. It is also worth pointing out that this system does not necessarily need to replace any existing beam polarisation measurements systems, it could easily be utilised in conjunction with other methods for cross-checking and systematic error reduction. Additionally, the pellet generation and drop rate could be tuned to enable beam-target interactions selectively with individual beam bunches. To improve the position resolution, a pellet tracking system, similar to Ref. [10] could eventually be implemented.

Target	Size	Density	References	Notes
WASA Pellet	40-60 μm diameter	$\sim 1\times10^{16}$ atoms/cm^2	[7, 8]	Hyperpolarised drops will have comparable density of protons.
RHIC H-Jet	6.5 mm FWHM	1.3×10^{12} atoms/cm^2	[4]	Long measurement times ($\sim$ 8 hrs)
RHIC C-Foil	$\sim$5 μm width	1.5×10^{14} C atoms/cm	[5, 11, 12]	Will disintegrate rapidly in EIC proton beam

Table 1: Comparison between various target properties for the WASA pellet target, the RHIC polarised hydrogen jet target and the RHIC thin carbon foil target.

In addition to the pellet system, a prototype (simulated) detector system to measure the spin-spin asymmetries from proton-proton scattering would also need to be developed. It is likely that such a system would be developed along the lines of the RHIC proton jet beam polarimetry systems, where silicon strip detectors are utilised.

It is informative to estimate the potential benefits of ChHP pellets over the polarised gas jet. The accuracy of a polarimeter measurement is given by:

$$\Delta P = \sqrt{\frac{2}{NF^2}}$$

where F is the figure of merit and N is the number of useful events recorded (a product of the yield and the acceptance of the polarimeter). In comparison with the jet target (where the proton polarisation is around 100%), there will be roughly a factor of two reduction (ChHP gives around 50%). For ChHP there will be an increase of around 4 orders of magnitude in the density of polarised protons (Table 1) in the target pellet (note that the pellet volume can be varied). For the same measurement period and assuming the same polarimeter acceptance, ChHP would therefore potentially give a reduction in ΔP by a factor of 50. Equivalently a measurement accuracy that takes 8 hours to achieve with the jet polarimeter could, in this idealised situation, take around 10 minutes. However, this very rough estimate ignores a number of key aspects that will be addressed in this R&D project and listed below.

- There will be effects from background of (non-polarised) protons and those bound in nuclei in the pellet which will affect the achievable performance. Methodologies of kinematic suppression and subtraction of these background events from the polarised scattering yield will be explored through the simulation program and will affect the achievable performance. We note that unpolarised ChHP pellets could be produced readily from a ChHP system, which could be incorporated into EIC measurement protocols if found to be beneficial in addressing the contribution of this background.

- The denser pellet targets will produce higher event rates. Simulation studies with a prototype detector system are necessary to ascertain any issues with characterising the events of interest in the detector systems. The initial studies are crucial to set the detector requirements to operate effectively with different ranges of pellet size.

- One of the benefits of ChHP is the possiblity to orient the direction of the pellet polarisation arbritarily. There are likely to be advantages in using such a measurement - not only through construction of asymmetries between orthogonal polarisation directions but also using pellet streams with polarisations (sequentially) rotating in steps. Also it is well known that longitudinal spin-spin correlations (at larger momentum transfers than typical for the CNI region used with the jet polarimeter) give sizeable asymmetries in cross sections in opposite spin configurations, up to a factor of 4[13] (although at energies lower than at the EIC). It may be possible to exploit such correlations with a ChHP polarimeter located at a suitable position of longitudinal polarisation in the beamline. However, such methodologies clearly need to be explored through simulation to establish feasibility.

Key to the prototyping and performance evaluation of a polarimeter detector apparatus is accurate simulation. Achieving this relies on the implementation of spin-spin dependence of elastic scattering processes in Geant4, as these are the basis of the polarimeter operation. The Geant4 development and its application in detailed EIC simulation is a skilled but crucial task for the project - crucially relying on the requested level of PDRA support (Kay) if it is to be achieved in the proposed timescale.

1.6 Possible impacts beyond the current project

We have already made significant advances in optimising the catalysts, fields and substrates to achieve macroscopically large volumes (beyond $\sim$cm^3) of polarised material. Such large volumes will not be needed for the tasks in Section 1.5, but if achieved would open up the possibility to utilise the spin-spin correlation to measure the polarisation of final state nucleons at the EIC. This is a missing observable which would impact science cases from DVCS to DVMP. We will continue to optimise this as part of the project as it will also be relevant to the main task in Section 1.5.

If our project aims are achieved, other opportunities with polarised ChHP droplet targets at the EIC are possible. As the technology can polarise other non-zero spin nuclei (Deuterium, ^{15}N,..) this may offer currently unanticipated EIC physics opportunities. Scoping the possibilities for polarised (active) room temperature liquid target systems using ChHP in campaigns at various halls in Jefferson Lab will also be progressed by this R&D.

1.7 The project team

The project team contains leading scientists in the field of Chemical hyperpolarisation, including the head of the ChyM facility in York (Ducket). There are two experts in EIC simulation (Zachariou, Kay) who have delivered benchmark simulations for the succesful ECCE proposal [14, 15]. Watts has decades of experience in polarimetry - having led the construction and design of the large acceptance nucleon spin polarimeter with the Crystal Ball at MAMI [16]. Bashkanov has decades of experience working with WASA [17, 18], including the pellet target systems and has expertise in nucleon polarimetry.

1.8 Yearly objectives and deliverables

In summary, the principle objectives of this project would be:

1. To develop and test a ChHP based polarised hydrogen pellet dripper system
2. Obtain a first measurement of the polarisation and relaxation time of frozen polarised hydrogen pellets in vacuum. Assessment of pellet-to-pellet variations
3. Further optimise the ChHP process to enhance the polarised hydrogen yield and optimise the substrate/catalyst combinations for its use in nuclear and particle physics.
4. Deliver simulations of a prototype ChHP polarimeter system for proton beam polarisation measurements at the EIC. Obtain the baseline characteristics - expected accuracy in proton beam polarisation determination for a given measurement time and pellet rate, systematic studies. Both tasks require the implementation of spin-spin dependent elastic scattering in Geant4/DD4HEP.

The deliverables from these key objectives by the end of the year would be as follows:

1. Demonstrate a working prototype of the pellet dripper system.
2. Preliminary results from in vacuum measurements of the produced pellets.
3. Results and updates from catalyst optimisation studies.
4. An initial design and implementation of the detector system, preferably in DD4HEP for ease of integration into the ePIC analysis framework.
5. Implementation of spin-spin dependent elastic scattering in chosen simulation package.

1.9 Budget

The budget for this 1 year project is relatively straightforward with a small number of items. The budget items are summarised in Table 2 below, workshop personnel costs are folded into equipment costs in the table, but are explicitly outlined below. We have assumed and included an exchange rate of £1:$1.30 in all provided figures. To deliver the first three objectives, as outlined in Section 1.8, a pellet system will need to be constructed. The equipment required to produce chemically hyperpolarised samples and measure their polarisation is already available. However, a vacuum pump and chamber to mimic beam vaccuum conditions and allow entrance of the liquid droplets is necessary. The chamber head will be made of steel (with viewing window) with the pellet drop component of the vacuum chamber likely to be comprised of (MRI compatible) carbon fibre or PTFE pipe through which the pellets fall ($4k). The chambers will be designed by the University of York (UoY) workshop and have all joins vacuum tested. In total, this will require 6 weeks of 1.0 FTE UoY workshop effort ($11.5k). An appropriate vacuum pump, connections and hoses, pressure gauges would also be required ($2.5k). A system to collect and evacuate the droplets after passing through the

MRI will also be required ($1k). A fast frame camera will image reflected laser light from the droplets (through the chamber viewing window) to measure their size ($1.5k).

For the pellet generation system, the main cost originates from the para-hydrogen generator which is provided in-kind by the UoY (system cost around £150k comprising an electrolyser to produce the hydrogen and a chiller to polarise). We request costs for the manufacture of a bespoke polarising magnet infrastructure to be used in polarising the droplets in arbritary directions, a set of nozzles, containment vessels, piezoelectric vibrator ($2k for magnet, total $2k for other items) and associated workshop costs (6 weeks with 1 FTE, $11.5k) for design and manufacture of the dropping system. We also request consumables for the creating the ChHP droplets ($3k for range of substrates, catalysts).

We request one year of a Masters/PhD student to carry out key tasks in the experimental aspects. These tasks would be carried out in laboratories at the University of York. As the project is interdisciplinary, we anticipate this will be a Chemistry student, likely supervised by Ducket, who will prepare the wide range of pellet samples in the program. This will be an attractive student project since it includes high-impact and publishable first measurements (e.g. properties of frozen ChHP samples).

To progress the simulation program (commensurate with the experimental program) in this one year time frame requires high-level simulation skills and detailed knowledge of EIC simulation. Due to the complexity of the simulations, which involve high-level adjustments to Geant4 to incorporate new polarised reaction routines, we crucially rely on PDRA level support. The experimental tests will also require a physicist experienced in the operation and analysis of MRI data to oversee the measurements. The PDRA candidate (Kay) has these skills. He is available and enthusiastic to work on the project.

A travel budget is requested to enable the PDRA, student and staff members to present at appropriate ePIC or EICUG meetings (5 person visits, $2k per visit, during the project).

1.10 Budget reduction scenarios

In a -20% budget scenario, we envisage that postdoc time allocated to simulations would be reduced. The reduction in the budget allocated to the PDRA in this scenario represents a reduction in their FTE assignment to this project. Detector simulations are contingent upon the satisfactory performance of the hyperpolarised pellet system, as such, it would be acceptable to potentially delay the completion of these simulations by reducing the time allocated to their completion. Student/staff travel to ePIC/EICUG meetings is also cut in this scenario. For a -40% budget scenario, the FTE allocations of the PDRA is reduced further. This scenario would represent almost cutting the simulation aspects of the project entirely. The student and equipment funding is unaffected in all scenarios as these are required for two major pillars of the funding. As such, both budget reduction scenarios would only impact deliverables 4 and 5 from Section 1.8. In subsequent years, we project that funding for the PDRA could be tapered off as the complex aspects of the simulation component conclude and the project shifts focus back towards construction, in this case of a detector system and a more mature pellet system.

Item	Type	100% Funding	80% Funding	60% Funding
Vaccuum chamber/system	Equipment	\$20,500	\$20,500	\$20,500
Dripper system to produce pellets	Equipment	\$18,500	\$18,500	\$18,500
Graduate student - 12 months 1 FTE, pellet system construction	Personnel	\$30,000	\$30,000	\$30,000
PDRA - Construction/Simulations - 12 months 0.5 FTE	Personnel	\$80,000	\$58,200	\$26,400
ePIC/EICUG Meeting	Travel	\$10,000	\$ 0	\$ 0
Total		**\$159,000**	**\$127,200**	**\$95,400**

Table 2: York FY24 Budget request money matrix, includes overheads.

1.11 Diversity, Equity and Inclusion

All staff at the University of York undergo training in DEI and recruitment will be carried out in this framework[1]. Additionally our group won significant funding (£0.48M) from the UK Science and Technology Facilities Council (STFC) Global Challenges Research Fund (Watts Co-PI STFC grant Nr. ST/S003118/1) to partner and provide local laboratory infrastructure, access to advanced simulation and high-level PhD training for students in the global south; including the Universities of Zululand and Western Cape in South Africa. We are fully engaged with the DEI agenda and will explore how this can be advanced with this project. As the project continues aspects suitable for engagement with global south researchers will be fully explored e.g. we have successfully supervised distance masters projects which have been successful in the past. Bashkanov, Zachariou and Watts also won Royal Society "exchange" funding which enabled a new collaboration and exchange visits with theorists from Khon Kaen University in Thailand. Collaborations in theoretical developments associated with this project will be explored with Khon Kaen and other groups in the global south.

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A Recent relevant R&D Results from the York ChHP Program

Recent work has been done on extending the lifetime of the chemical hyperpolarisation by investigating the effects of the storage field and the concentration of the catalyst on the relaxation time constant, T1. The chosen substrate for these experiments has been 3,5-dichloropyridine due to its long T1 as a result of having well-shielded protons.

The T1 time constant has been shown to be strongly dependent on the concentration of catalyst in solution. With a molar excess of substrate of over 120-fold relative to the catalyst the T1 for the para position proton has been shown to exceed 4 minutes, a 4x increase on that seen for a 5-fold excess as seen in Figure 3. The T1 increase for the doubly-represented ortho position protons is even more significant as they experience increased dipole-dipole interactions due to their proximity to the catalyst. Here the T1 experiences a 10x increase by going from a 5-fold excess to a 120-fold excess.

The dependence of the polarisation relaxation rate on the storage field has also been investigated in order to understand whether lifetime gains can be made this way. The T1 at different fields have been studied on 4 different spectrometers with fields of 1.4, 7.0, 9,4 and 11.7 T as seen in Figure 4. The lowest field was from the 60 MHz Spinsolve Carbon benchtop NMR and the higher fields were from 300/400/500 MHz Bruker NMR spectrometers.

It has been shown that there is little dependence on the polarisation lifetime with varying storage fields, showing that storage fields in excess of 1.4 T are unnecessary. The optimum polarisation transfer field (PTF) to protons with SABRE has been shown to be around 6 mT and thus further studies at lower fields will be attempted in order to determine whether a storage field distinct to the PTF is necessary.

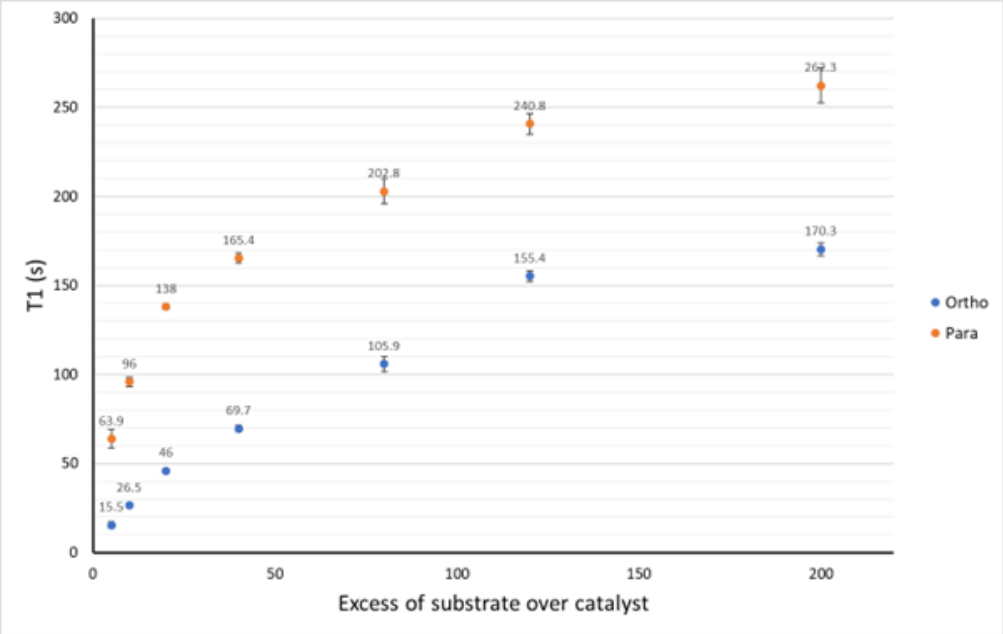

Figure 3: 3,5-dichloropyridine T1 with varying excess over IMes catalyst. Fixed concentration of 3,5-dcp (50mM) with varying concentration of IMes catalyst. T1s recovered via a sequence with 30, 10.35' flip angle pulses, with a 10s delay. Results are an average of 5 repeats, with the error bars representing 1 SD.

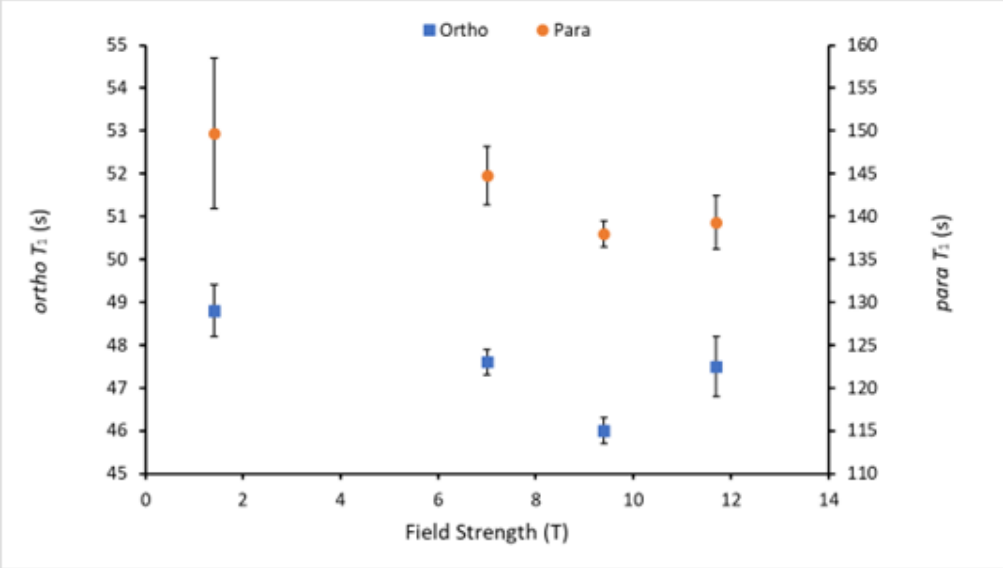

Figure 4: 3,5-dichloropyridine T1 with varying storage field. Fixed concentration of 3,5-dcp (50mM) with 2.5mM of IMes catalyst. T1s recovered via a sequence with 30, 10.35' flip angle pulses, with a 10s delay. Results are an average of 5 repeats, with the error bars representing 1 SD. For 1.41T 20 12.6' flip angle pulses were used with a 10s delay.