Investigation and Characterization of Sol-Gel Coated Target Cells to Improve ^{3}He Polarization

A thesis submitted in partial fulfillment of the requirement
for the degree of Bachelor of Science with Honors in
Physics from the College of William and Mary in Virginia,

by

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ABSTRACT

The internal structure of the neutron is of current interest in the field of nuclear physics. Presently, experiments at the Thomas Jefferson National Accelerator Facility explore the nature of the neutron using polarized ^{3}He target cells. The College of William and Mary now has the capability to produce polarized ^{3}He cells and characterize them using a nuclear magnetic resonance system. To effectively study the neutron, the ^{3}He in the target cell must achieve a high degree of polarization and remain polarized for a long period of time. Currently, target cells are made of aluminosilicate glass, which has the desired property of preventing the depolarization of ^{3}He nuclei. However, aluminosilicate glass is difficult to work with and costly. Ideally, a cheaper and more manageable type of glass, such as Pyrex, could be coated to create a better surface and thus increase the lifetime of the polarized ^{3}He. The purpose of this honors thesis is to test sol-gel coating techniques on target cells and measure the resulting lifetimes achieved. Our ultimate goal is to produce highly polarized cells with long lifetimes for use as targets in experiments at Jefferson Lab.

ACKNOWLEDGEMENTS

All of my thanks most certainly goes to Dr. Todd Averett, without whom this little endeavor would have been impossible. I learned more than I ever could have hoped, and I had an amazing time doing so. Todd, thanks so much for being a wonderful mentor over the past four years. You have to admit that there was never a dull moment! Rest assured that I certainly learned one thing: never buy stock in Pfeiffer.

CONTENTS

1. INTRODUCTION

Physicists at the Thomas Jefferson National Accelerator Facility are exploring the internal structure of neutrons through electron scattering experiments. To gather information on the spin properties of the neutron's substructure, a cell of spin aligned, or polarized, nuclei is used as a target for electron beam scattering experiments. Ideally, the target cell would contain polarized free neutrons. However, due to the free neutron's radioactive instability [1], researchers use polarized ^{3}He as an effective alternative.

Helium-3 is an acceptable substitute for free neutrons because it contains two protons of anti-parallel spin, and one neutron. The anti-alignment of the protons essentially cancels their spins, thus leaving the ^{3}He nucleus with the spin of the single neutron. Polarizing ^{3}He gives the nucleus and neutron the same known spin direction.

The ^{3}He is contained in a glass target cell (Figure 1). The upper chamber also contains rubidium, which is atomically polarized through a process called optical pumping. The polarized rubidium then interacts by spin exchange with ^{3}He, polarizing its nuclear spin. The bottom chamber contains only polarized ^{3}He, which is meant to interact with the electron beam. The end windows of the target chamber are extremely thin, approximately 150 microns, so that the electron beam's interaction with the glass is minimized.

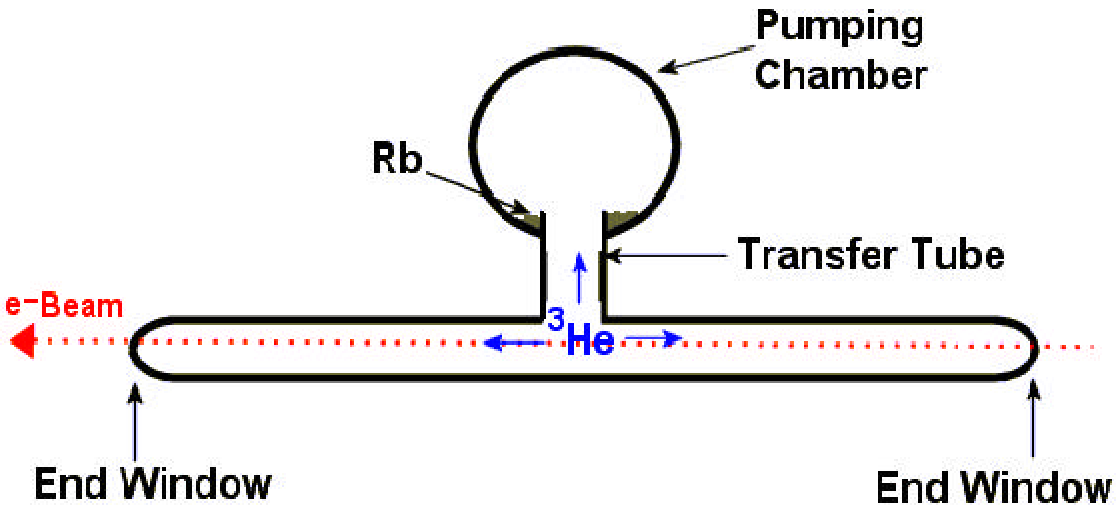

Figure 1: Typical Jefferson Lab target cell design. The pumping chamber contains rubidium used in the optical pumping polarization process. The lower chamber has thin cell walls, which the electron beam passes through, eventually colliding with ^{3}He atoms and scattering.

In the target cell, it is ideal to maintain a high degree of ^{3}He polarization that will be sustained over a long period of time, possibly days. However, there are numerous effects that can cause ^{3}He to depolarize. The most notable depolarization mechanism is the interaction of ^{3}He with the magnetic impurities of the glass cell walls upon collision. To minimize this effect, the cells are made of aluminosilicate glass, which is known to reduce depolarization effects and have a low porosity to ^{3}He [2]. However, this type of glass requires a costly and difficult glass blowing process.

Coating techniques can be used to create glass vessels that suppress the depolarization effects caused by cell wall collisions. Sol-gel, which produces a high purity glass in solution, is thought to be a suitable coating. Sol-gel begins with colloidal particles in a solution, or sol, and then becomes a gel of macroscopic material and dispersed liquid [3]. When the liquid evaporates, a smooth glass-like surface remains. Sol-gel coatings may be applied to target cells made of a less expensive and more manageable material, like Pyrex. The purpose of this honors thesis is to develop a system to characterize target cells and investigate the effectiveness of sol-gel coatings on improving the ^{3}He polarization and lifetimes in cells for experimental use at Jefferson Lab.

2. BACKGROUND

2.1 Polarization

The polarization of ^{3}He is a two-part process that involves the optical pumping of Rb and its spin exchange with ^{3}He. The ^{3}He target cell is placed in the target system seen in Figure 2.

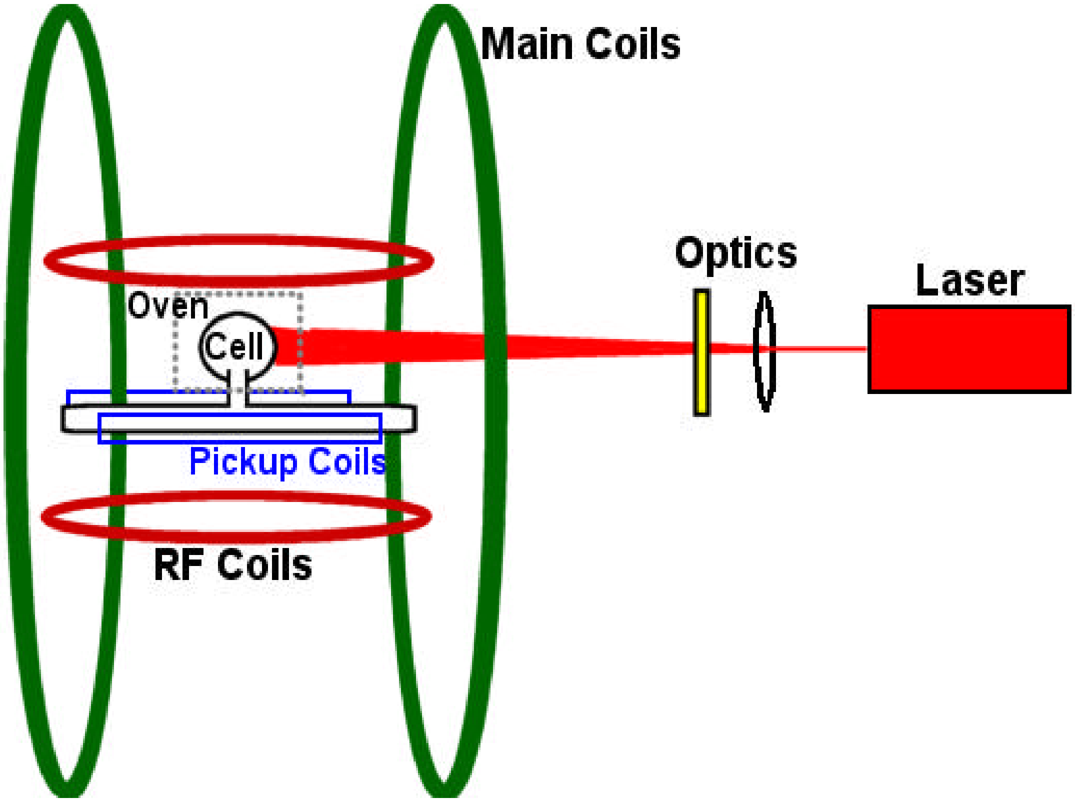

Figure 2: Schematic of the laboratory at William and Mary, which is used to polarize ^{3}He and take polarimetry readings. The pumping chamber of the cell is surrounded by an oven, which has an opening, allowing the laser beam to strike the cell. The main Helmholtz coils (green) create the magnetic holding field. The RF coils (red) create the RF field, and the pickup coils (blue) are placed next to the cell, where an EMF is induced by the precession of polarized ^{3}He nuclei during NMR.

2.1.1 Optical Pumping

Because it is difficult to directly polarize ^{3}He, the first step in the polarization process involves optically pumping the alkali metal, rubidium. The pumping chamber of the target cell is placed in an oven, causing the Rb to vaporize. The large Helmholtz coils, referred to as the main coils, create a magnetic field, which causes Zeeman splitting in the atomic spin states of Rb. Both the ground state, $5S_{1/2}$, and the first excited state, $5P_{1/2}$, are split into m = +1/2 and m = -1/2 states, as seen in Figure 3. A diode laser produces right-circularly polarized light at a wavelength of 795nm. Due to selection rules, this particular wavelength of light excites electrons from the $5S_{1/2}$, m = -1/2 state to the $5P_{1/2}$, m = +1/2 state. Collisional mixing between the Rb atoms causes the electrons to equally distribute themselves between the sublevels of the excited state. The electrons then decay to the ground state, and those that fall to m = +1/2 remain there, and will not be excited by the laser light again. The electrons that fall back into m = -1/2 of the ground state are quickly optically pumped back to the excited state, from which they will all eventually decay back to the $5S_{1/2}$ m = +1/2 state. However, when the electron de-excites, a photon of random polarization and propagation direction is released. These photons may be reabsorbed, giving the Rb atoms an undesired polarization. To prevent such radiative decays, a volume of diatomic nitrogen (about 60 torr at room temperature) is added to the target cell during filling. This molecule is capable of absorbing the energy of the de-exciting Rb electrons through collisions. Thus, the N_2 allows Rb to decay back to the ground state without emitting a depolarizing photon. Optical pumping under these conditions produces an atomically polarized Rb vapor.

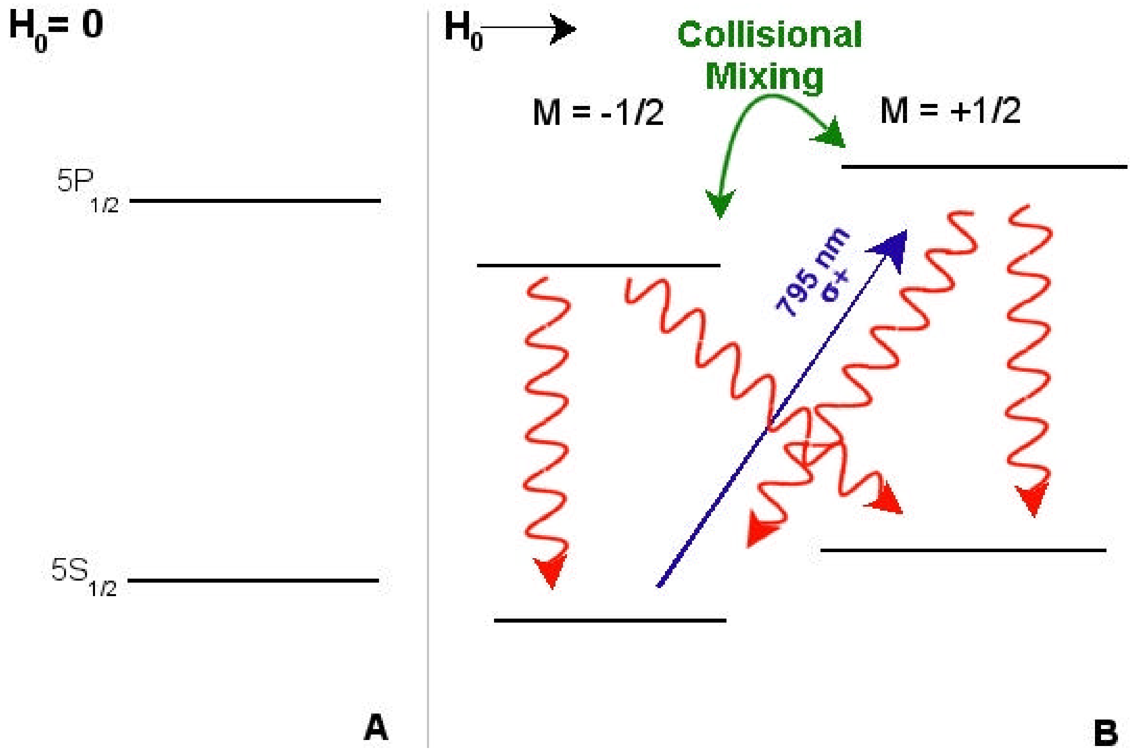

Figure 3: Optical pumping process. When a magnetic field (H_0) is applied, Zeeman splitting occurs in the ground state and first excited state. Electrons are excited to the $5P_{1/2}$ m = +1/2 state by 795nm laser light. Collisional mixing equally distributes the electrons between m = -1/2 and m = +1/2 of the excited state, from which they decay down to the ground state. Optical pumping continues until all electrons are in the $5S_{1/2}$ m = +1/2 state, where they have the same spin.

2.1.2 Spin Exchange

After the Rb vapor is atomically polarized, ^{3}He becomes polarized through a process known as spin exchange. Hyperfine interactions occur between the nucleus and electrons of the same atom. Spin exchange, however, is a hyperfine-like interaction, which occurs during binary collisions between the Rb atom and the nucleus of ^{3}He. The spin of the Rb atom is transferred to the ^{3}He nucleus, causing the Rb to depolarize (Figure 4) [4]. The Rb is then optically pumped back to the desired polarization state, where it is again ready for spin exchange. If no significant depolarization mechanisms exist, the ^{3}He remains polarized.

Figure 4: Rb atoms collide with ^{3}He atoms and transfer spin to the ^{3}He nuclei. This causes the Rb to depolarize, but it is quickly optically pumped into a polarized state where it is again ready for spin exchange.

2.1.3 Polarization Rate

Polarization is defined as:

$$P = \frac{N^{\uparrow} - N^{\downarrow}}{N^{\uparrow} + N^{\downarrow}} \tag{1}$$

in which $N^{\uparrow}$ represents the number of spins aligned parallel to an external magnetic field, and $N^{\downarrow}$ represents the number of the spins aligned antiparallel. Currently, the maximum absolute ^{3}He polarization measured at Jefferson Lab for cells at a density of about 9 amagats (1 amagat = 1 atm of gas at room temperature) is approximately 45%.

The polarization rate of ^{3}He can be described by the following equation

$$P_{He}(t) = \langle P_{Rb} \rangle \frac{g_{SE}}{g_{SE} + \Gamma}(1 - e^{-(g_{SE} + \Gamma)t}) \tag{2}$$

The polarization of ^{3}He at a given time depends on several factors [5]. The spin exchange rate, g_{SE}, is the rate of spin exchange between atomically polarized Rb and ^{3}He. This value depends on the Rb density in the target cell and the probability of spin exchange between Rb and ^{3}He. The factor Γ is the depolarization rate of ^{3}He, and $\langle P_{Rb} \rangle$ is the average Rb polarization in the target cell. Ideally, over a long period of time, ^{3}He would reach a maximum polarization given by

$$P_{^3He}^{\max} = \frac{g_{SE}}{g_{SE} + \Gamma} \left\langle P_{Rb} \right\rangle \tag{3}$$

With the density of polarized Rb maximized through continuous optical pumping, it is evident from equation (3) that the depolarization rate must be minimized in order to increase the polarization of ^{3}He.

2.2 Depolarization

In the absence of optical pumping, the loss of polarization in a ^{3}He target cell can be described by the equation

$$P_{He}(t) = P_{init} e^{-\Gamma t} \tag{4}$$

in which P_{init} represents the polarization when optical pumping ceases. The lifetime of the ^{3}He polarization is defined as $T \equiv 1/\Gamma$.

The relaxation of ^{3}He polarization is affected by numerous factors that are unavoidable but can be minimized. First, the electron beam causes a loss of polarization by ionizing incident ^{3}He atoms. However, this is not a major effect during experimentation if the beam currents are kept low. An inhomogeneous magnetic field can also cause depolarization. During NMR measurements, spins will precess around a magnetic field at different rates if the field is changing due to gradients, causing the ^{3}He to lose its polarization [6]. Nevertheless, using large Helmholtz coils can reduce the magnetic field gradients that cause such a depolarization [2]. The largest of the unpreventable sources of depolarization are the dipole-dipole interactions between colliding ^{3}He nuclei [2]. This factor limits the maximum achievable lifetime by the equation

$$\frac{1}{\Gamma} = \frac{744}{\left[{}^3He\right]} hrs \tag{5}$$

in which [^{3}He] is in amagats [7]. In the absence of all other sources of depolarization, $1/\Gamma$ is the maximum lifetime of the target cell.

Larger, yet more avoidable, effects on depolarization are caused by the collisions of ^{3}He atoms with paramagnetic impurities in the gas and the cell walls. Paramagnetic impurities cause the nuclear spin of the ^{3}He atom to change orientation [6]. Thus, a clean cell and gas are extremely desirable. Depolarization due to interactions with the cell wall can be reduced by careful cell production as well. The glass used for the target cells is chosen based on its low-porosity, smoothness, and purity [2]. Aluminosilicate glass has proven to be most suitable. This type of glass has low helium porosity and is also characterized by a clean smooth surface. Aluminosilicate glass has fewer impurities, and re-blowing the glass creates a surface with fewer imperfections and fissures that might trap ^{3}He atoms and cause them to lose their polarization. Aluminosilicate glass appears to be the perfect choice for a long lifetime target cell; however, this glass is expensive and difficult for the glassblower to work with. The desire to create a flawless surface has motivated research in sol-gel coatings, which may be a manageable and inexpensive solution to reducing depolarization rates.

2.3 Sol-Gel Coatings

Sol-gel reactions are frequently used for preparing glasses, gels, and ceramic powders [3]. Sol-gel chemistry is an easy way to produce high purity glass in solution at room temperature. The creation of sol-gel begins with a hydrolysis reaction, which produces a sol, and then a gel is formed through a condensation reaction. The macroscopic gel contains a liquid that, once evaporated, leaves a durable glass-like material.

To begin the hydrolysis reaction, $Si(OCH_2CH_3)_4$ (tetraethoxysilane, or TEOS) is mixed with H_2O. TEOS, a metal alkoxide that serves as a common organic precursor, is used because it reacts readily with water [3]. During the hydrolysis reaction, alkoxide groups (OCH_2CH_3) are replaced by hydroxides (OH), producing silicic acid ($Si(OH)_4$) and ethanol (CH_3CH_2OH) :

$$Si(OCH_2CH_3)_4 + 4H_2O \rightarrow Si(OH)_4 + 4CH_3CH_2OH. \qquad (6)$$

Next, silicic acid breaks down, producing Si-O-Si bonds and water. This is known as the condensation reaction:

$$2Si(OH_4) \rightarrow (OH)_3Si\text{-}O\text{-}Si(OH)_3 + H_2O. \qquad (7)$$

As this reaction occurs, a macroscopic gel is formed consisting of a network of silicon atoms bridged by oxygen atoms. This is an extremely slow reaction, and the addition of HCl serves as a catalyst to increase the reaction rate.

After coating a surface, the sol-gel solution must be densified. Densification is a heating process that drives off the water and alcohol that is trapped in the gel [3]. The evaporation of the liquids shrinks the gel, leaving a smooth glassy surface. At this stage, the sol-gel coating may crack due to the stresses imposed by the shrinking process. This strain on the gel can be prevented by slowly heating the coating during the densification process.

2.4 Polarimetry

A nuclear magnetic resonance system is used to test the polarization and lifetime of ^{3}He target cells. To characterize a ^{3}He target cell, the NMR system is used to perform a technique known as adiabatic fast passage (AFP) [2]. Once the ^{3}He nuclei are polarized, their magnetic moments are anti-aligned with the holding field created by the main coils [8]. An RF field, created by Helmholtz coils orthogonal to the main coils, is then applied at a frequency of 91kHz. This value is chosen because it is close to the Larmor frequency of the ^{3}He nuclei, which is described by the equation

$$\omega = \gamma B \tag{8}$$

Thus, the resonant frequency for ^{3}He is determined by the gyromagnetic ratio of the nucleus, γ, and the magnetic field, B [9]. The main holding field is ramped from 25G to 32G in order to make a passage through resonance. This sweep causes a passage, known as AFP, through the resonance of the ^{3}He nuclei [10]. The process is adiabatic such that it sweeps through the resonance at a slow rate to give the ^{3}He spins ample time to precess and remain aligned with the effective magnetic field, the sum of the static field and RF field. The sweep must also be fast enough that the nuclei do not depolarize during the passage through resonance. This AFP causes the magnetization direction of the ^{3}He nuclei to be reversed, and the spins are flipped. The main field is then ramped back down through resonance, flipping the ^{3}He spins back to their original polarization direction. The precession of the spins causes a changing magnetic flux within the pickup coils [11], which are perpendicular to both the holding field and the RF field. The induced EMF in the pickup coils can be detected as a voltage and is used to measure the relative polarization in the cell. In order to obtain an absolute polarization value, a water calibration must be performed on the target system. Water has a known equilibrium polarization when placed in a magnetic field, therefore its measured induced voltage can be compared to ^{3}He values, giving the absolute polarization of gas in the cell.

3 EXPERIMENTAL PROCEDURE

To test the effects of sol-gel coatings on target cell lifetimes, the following experimental procedure is followed. First, a sol-gel solution is made and used to coat target cells, which are then baked to densify the coating. After the cell is filled with ^{3}He, it is polarized and tested on the NMR system. The maximum polarization, lifetime, and depolarization rate are physical parameters that determine the performance of a target cell's polarization. Spin-up measurements, AFP loss measurements, and spin-down plots are used to examine the quality of the cell.

3.1 Sol-Gel Solution

3.1.1 Previous Research

In previous research by Mellor [12], the chemical composition for sol-gel coatings originally derived by Hsu et al was first considered (Table 1) [13].

Composition (mole % Al_2O_3)	TEOS (mL)	$Al(NO_3)_3 \bullet 9H_2O$ (g)	Water (mL)	Ethanol (mL)
20%	11	9.2	6	66.9

Table 1: Sol-gel solution derived by Hsu et al.

This solution contains aluminum nitrate, $Al(NO_3)_3 \bullet 9H_2O$, which is added to simulate the properties of aluminosilicate glass. The surface created by this composition is meant to have a desired thickness that can withstand repeated heating cycles without cracking. However, upon testing, the films produced were not consistent with the findings of Hsu et al [12]. Next, a more basic chemical composition of sol-gel, derived by Sakka et al, was tried [14]. Different ethanol dilutions of the solution were tested on glass slides, and a composition that provided the best compromise between cracking and the thickness of the coating was found (Table 2) [12]. The cracking was examined under a scanning electron microscope, and the thickness profile was measured.

TEOS (mL)	Water (mL)	Ethanol (mL)	HCl (mL)
23.4	29.7	43	0.9

Table 2: Basic chemical composition chosen for the sol-gel coatings.

While this composition and dilution proved successful on slides, the results were inconclusive as to the resulting surface's effectiveness on the polarization of a ^{3}He target cell.

3.1.2 Current Research

The research in this thesis began with the composition in Table 2. Sol-gel solutions diluted with extra ethanol (83mL and 123mL total) were also tested on cells. First, the chemicals are measured into an Erlenmeyer flask using a graduated plastic pipette. The water is added first, followed by the ethanol, TEOS, and HCl. The HCl is an aqueous solution of 70% water and 30% pure HCl. After the chemicals are added, the flask is covered with parafilm to prevent evaporation. This solution is then mixed using a magnetic stirrer at a medium speed for approximately four days.

3.2 Cell Coating

After the sol-gel solution has been mixed, the glass target cells can be coated. A glass tube (stringer) with four attachments for 1" diameter spherical cells was used (Figure 5).

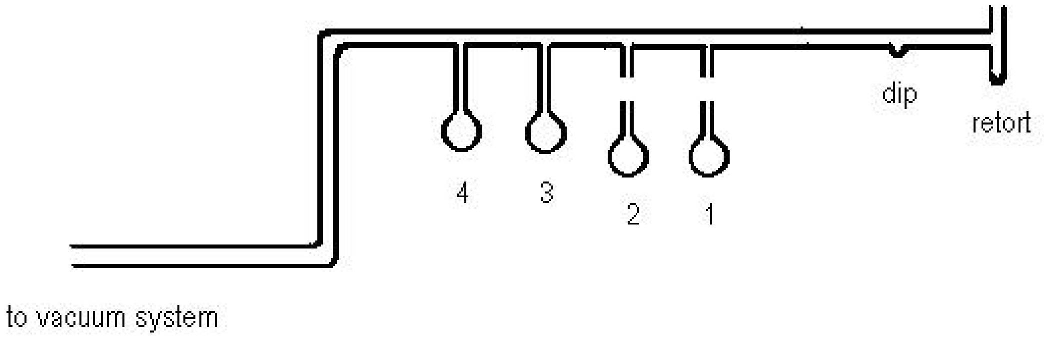

Figure 5: Schematic of the glass string to be attached to the vacuum system. Spheres to be coated with sol-gel (cells 1 and 2) are left unattached from the glass stringer until the coating process is complete. The Rb is sealed in the retort at the end of the stringer, and it is later moved to the dip.

The spheres to be coated with sol-gel were left unattached from the glass stringer. Using a pipette, the spheres are filled to the top with sol-gel solution, and they are left sitting upright and covered for approximately an hour. The solution is then poured out, and the cells are placed on a specially designed rack in the oven. The cells are positioned with the opening down to let the extra solution drain out during the densification process. The oven is set to 60°C for an hour to let the cells slowly dry before they are baked. The oven is then raised anywhere from 120°C to 150°C every hour until it is stable at 500°C for one hour. The oven is turned off, and the cells cool overnight. This slow heating process is used to prevent cracking in the sol-gel coating [3].

3.3 Cell Fill

To fill the cells with ^{3}He, the string is first attached to a vacuum system. A Rb ampoule is sealed in the retort at the end of the string, and the cells are then allowed to bake at a maximum of 470°C for approximately one week under ultra high vacuum in the 10^{-9} Torr range. During the bake-out, the exposed parts (outside the oven) of the glass string are heated with a torch three times a day. A few days into the baking process, the

Rb is moved out of the ampoule and into the dip, at which point the retort can be removed. Baking the cells and string ensures a clean pathway for the volume of ^{3}He and Rb needed to fill the cells.

After the bake-out, the oven is turned off, and the cells are cooled. They are then filled with a known volume of N_2 and ^{3}He and are cooled by liquid nitrogen. Finally, a torch is used to remove the cells from the string at the pull off point, and the cells are allowed to slowly return to room temperature.

3.4 Target Cell Characterization

To test the polarization and depolarization rates of the spheres, they are mounted in the target system. The set-up shown in Figure 2 is not capable of testing 1” spherical cells, so changes were made to the oven, and a new set of pick-up coils was built (Figure 6).

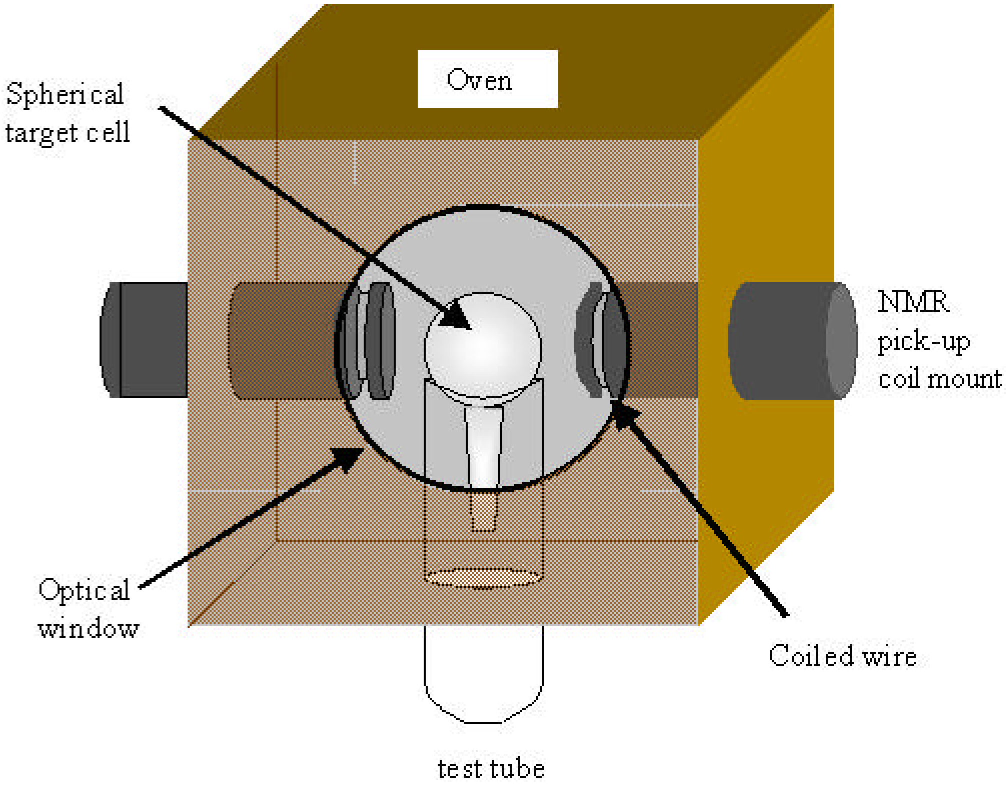

Figure 6: Set-up of the target system used to test 1” spherical cells. The cell sits on a test tube to center it in the optical window, through which the laser light passes. The pick-up coils contain 240 turns of wire, which are placed as close to the target cell as possible. The entire setup is enclosed in an oven that is used to vaporize the Rb for optical pumping.

The new upper pick-up coils were made by wrapping 240 coils of wire around 1” cylinders. The pick up coil mounts were then placed in tightly sealed holes on opposite sides of the oven, where they can be moved close to the target cell for alignment. The target cell sits on a test tube, which centers the sphere in the path of the laser beam.

After the target cell is placed in the system and the coils are aligned to reduce noise, the oven is heated to 170°C. The laser is turned on to begin the polarization process. Optical pumping is performed by using one laser at 30W, and then spin exchange occurs, polarizing the ^{3}He nuclei.

While the ^{3}He is polarizing, an AFP measurement can be performed and polarization data acquired. Voltage readings are taken from the pickup coils and plotted versus the holding field. A lock-in amplifier separates the signal and the noise into separate X and Y channels. The program can perform multiple sweeps at desired time intervals, which is ideal in gathering cell characterization data.

The plots produced by the NMR sweep program show two peaks, which are the products of the up and down sweeps through resonance (Figure 7).

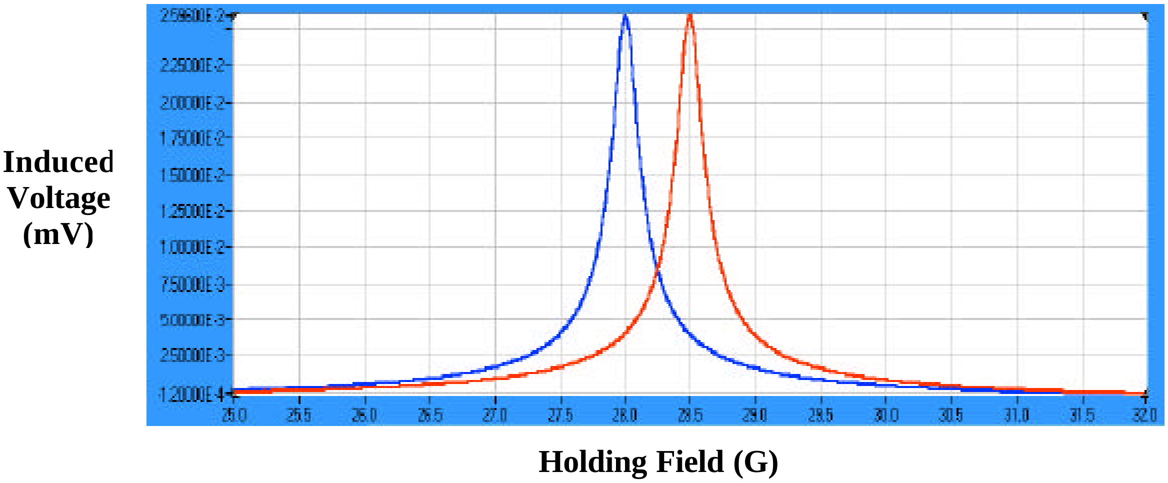

Figure 7: Output of a LabVIEW program. The voltage values (mV) induced in the pickup coils by the precessing ^{3}He spins are plotted as a function of the holding field (G). The two peaks represent the up and down sweeps through resonance. They are offset due to a time-lag in the electronics.

The peaks occur at the value of the holding field when the ^{3}He passes through resonance, and they show the relative polarization in mV. Though the resonance field value should be the same for the up and down sweep, the peaks occur at different field values due to a time-lag in the electronics. A fitting program is used to find the peak voltage. This procedure is followed for numerous measurements that are used to characterize a target cell.

3.4.1 Spin-Up

The spin-up is a procedure that helps determine the rate at which ^{3}He polarizes. After the cell is mounted in the oven and heated up, the lasers are turned on and the ^{3}He is left to polarize through optical pumping and spin exchange. To gather spin-up data, an NMR measurement is performed every 4 hours, and the up and down sweeps are analyzed. From the best-fit coefficients to the curve, the peak height of the relative polarization signal can be determined and plotted with the time of each sweep.

The shape of the spin-up curve reveals much about the nature of ^{3}He polarization. The curve rises sharply in the beginning, suggesting that ^{3}He polarizes quickly in the initial stages. Eventually, it begins to level off as the polarization reaches a maximum. The spin-up for a decent cell (Figure 8) can be fit to equation (2). A very poor cell will polarize and level off too sharply to be fit by this exponential equation.

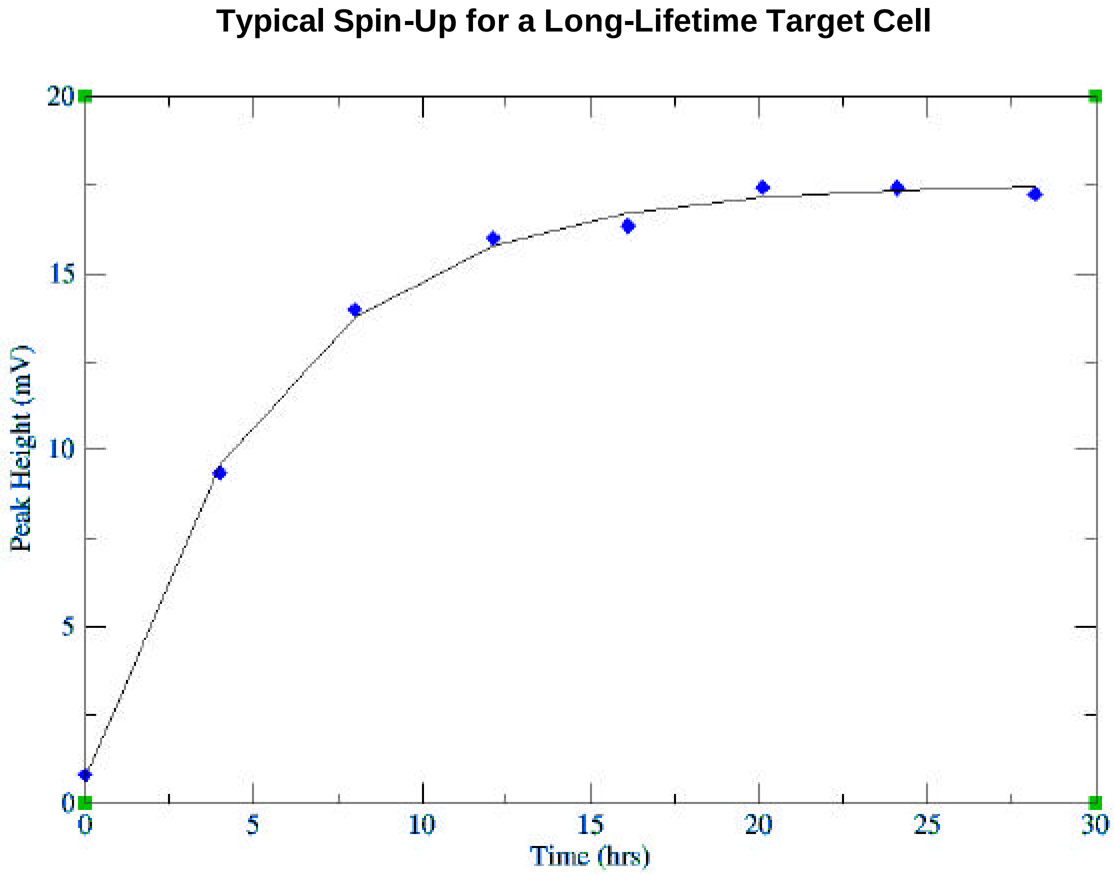

Figure 8: Spin-up plot for a typical target cell. The curve can be fit to equation (2), which describes the polarization of ^{3}He at any given time. This curve depends on the spin exchange rate, depolarization rate and the amount of polarized Rb present. The shape and successful fit of this curve suggest that the cell may have a suitable lifetime.

3.4.2 Spin-Down

The spin-down procedure is a method used to study the depolarization of ^{3}He and the lifetime of its polarization. After the cell reaches maximum polarization, the laser and oven are turned off, and the cell is allowed to depolarize. Again, an NMR sweep is taken every four hours, and the peaks are analyzed. The signal height is then plotted versus the time of the sweep.

The typical spin-down is a smooth exponential curve (Figure 9). The polarization decreases and levels off according to the depolarization rate of the particular cell. This curve can be fit to equation (4). Because Γ is the rate at which the cell depolarizes, its reciprocal, $1/\Gamma$, reveals the lifetime, in hours, of the ^{3}He polarization. However, the lifetimes measured from the raw NMR data must be corrected for AFP losses, which are explained in the next section.

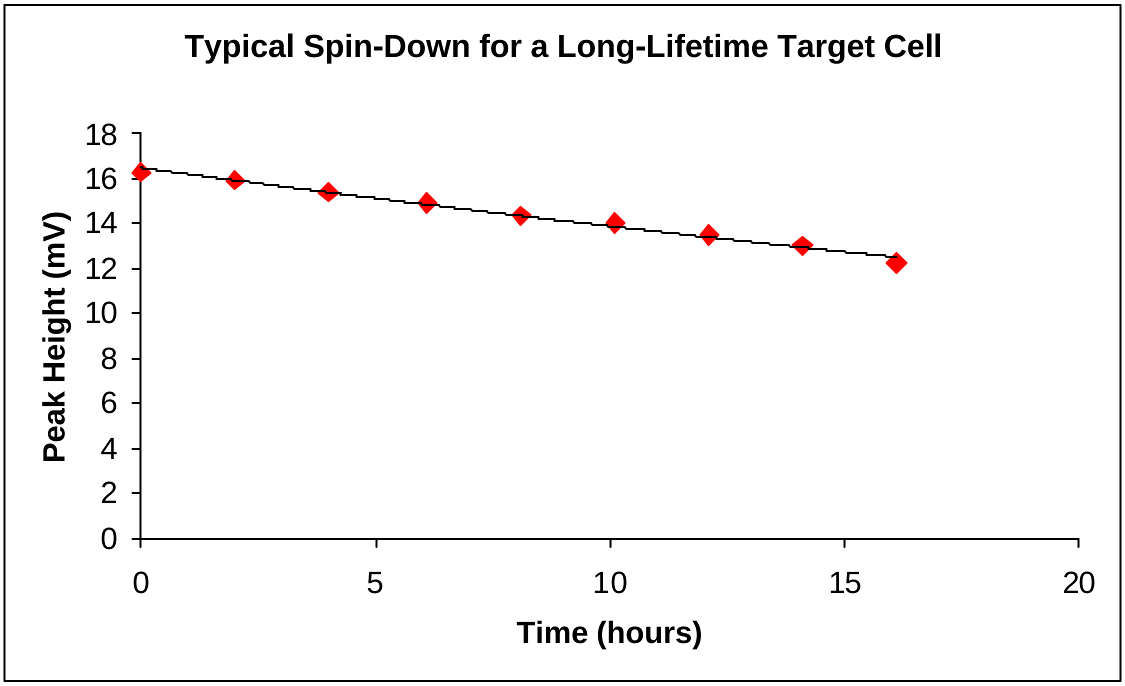

Figure 9: Spin-down plot for a typical long-lifetime cell. The polarization decreases at a very slow rate, which is comparable to an exponential decay. The spin-down data is fit to equation (4). The lifetime can be calculated using the fitted value of Γ. In this case, the lifetime, or $1/\Gamma$, is found to be 59.5 hours.

3.4.3 AFP Loss Test

When an NMR measurement is made, the spins of the ^{3}He nuclei are flipped. The adiabatic fast passage must be slow enough to allow for the ^{3}He nuclei to precess, but fast enough that it does not depolarize during the passage through resonance. However, during a measurement, some of the ^{3}He polarization is lost. This does not have a major effect on the spin-up curves, because measurements are being made while the cell is continually polarizing. However, for the spin-down curves, the AFP measurements cause additional depolarization that must be taken into account and corrected for. To measure the losses, five NMR sweeps are made consecutively on a cool, depolarizing cell. The average polarization lost between measurements, the AFP loss, is then used to correct the spin down curve (Figure 10). For a short-lifetime cell, depolarization happens too quickly to get decent AFP loss measurements. On the other hand, long-lifetime cells are depolarizing slowly, making them more sensitive to AFP losses.

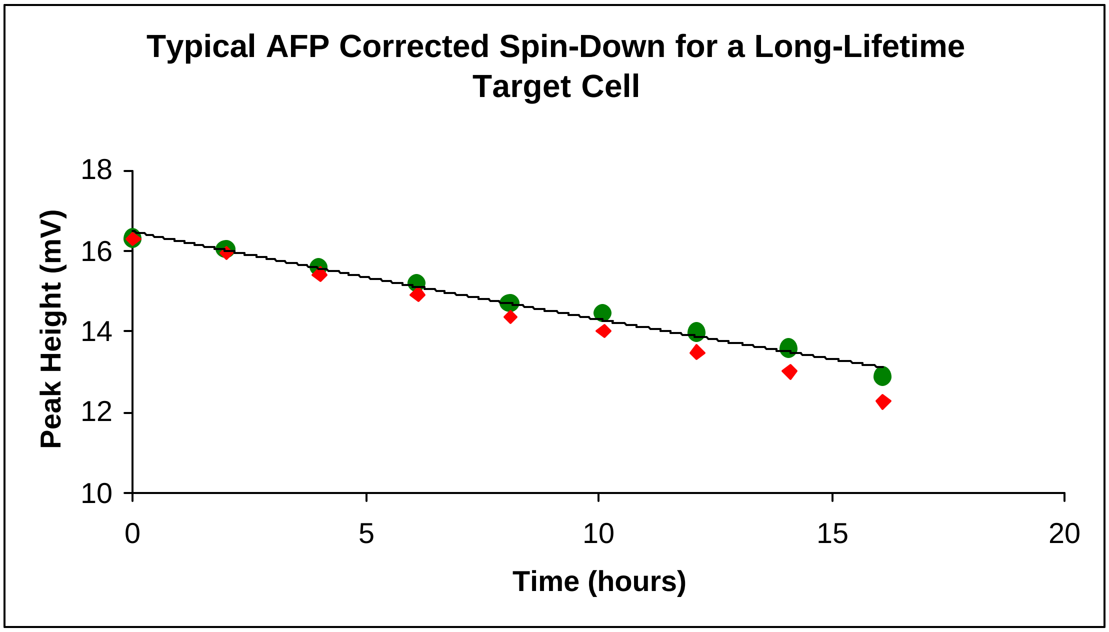

Figure 10: Plot of an AFP corrected spin-down for a long-lifetime cell. The diamonds (red) represent the raw spin-down data, and the circles (green) are the AFP corrections for each point. The AFP corrected points are fit using equation (4). The corrected lifetime of this cell, $1/\Gamma$, is calculated to be 72.1 hours, whereas the uncorrected lifetime is 59.5 hours. It is evident that AFP corrections over a long period of time are substantial for long-lifetime cells.

4 EXPERIMENTAL RESULTS

Three cell fills were performed this year, each producing four testable spherical cells. The results of the sol-gel coatings, cell fills, and NMR measurements follow. A summary of all of the data is given in Table 3.

4.1 Cell Fill 1 (12/3/02)

The first group of spherical cells consists of two cells coated with the sol-gel solution from Table 2, and two uncoated cells, all of which are made of an aluminosilicate glass, GE-180. After densification, the coated cells appeared to have a ring of thick sol-gel around the center of the sphere. This indicates that extra sol-gel remained in the cell when densification began, and therefore, more draining and drying time is necessary for a smooth coating. The cells were then attached to the stringer with a torch, where the coated cells are numbered **1.1** and **1.2**, and the uncoated cells are numbered **1.3** and **1.4**. The cell attachment was not extremely successful, causing the glass to be exposed to a sooty torch for a long period of time during the glass blowing. This glass stringer and cells were then baked at 470°C, and the cell fill was completed December 3, 2002.

Upon testing, the two uncoated cells have spin-up curves that level off at approximately 58 mV for cell **1.3**, as shown in Figure 11, and 35 mV for cell **1.4**. The spin-up curve for both cells is comparable to the typical spin-up of a good cell seen in Figure 8.

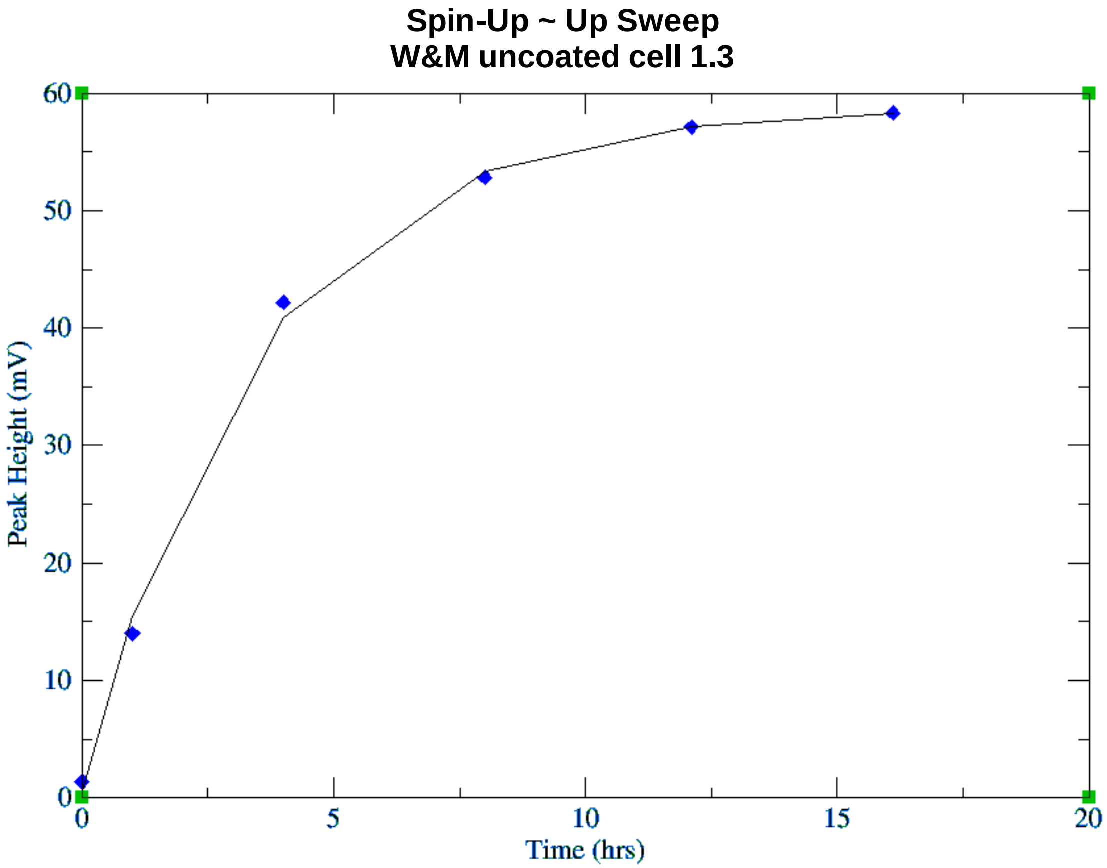

Figure 11: Spin-up plot of uncoated cell **1.3**. The cell polarizes quickly, but smoothly, and it levels off at a high peak voltage of about 58mV. The fit on this plot matches the shape of the typical spin-up curve seen in Figure 8.

The sol-gel coated sphere, **1.1**, polarizes quickly, but sharply levels off at a voltage of less than 10 mV (Figure 12). This curve differs greatly from the typical spin-up, and it cannot be fit to equation (2). The other coated sphere, **1.2**, maximized at a polarization of 52 mV, yet also has a sharp spin-up curve, similar to **1.1**. These were the first indications that the coated cells would not achieve a better lifetime than the uncoated cells.

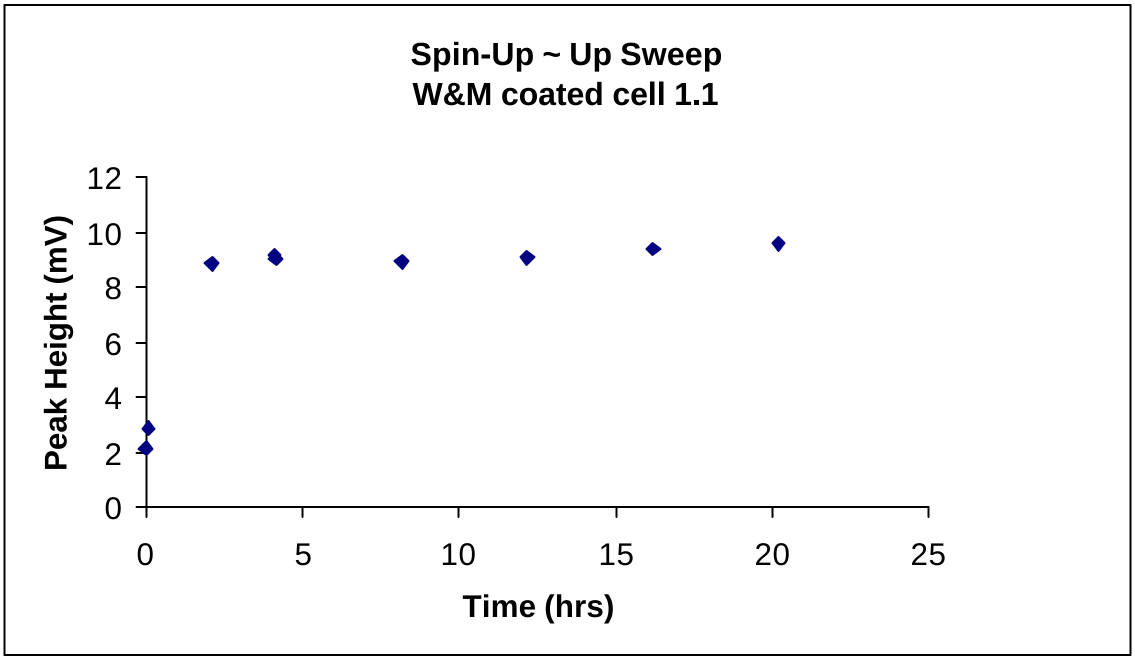

Figure 12: Plot of the spin-up for sol-gel coated cell **1.1**. The cell polarizes quickly, but then sharply levels off at a low peak height voltage. This curve cannot be fit with the spin-up equation.

The spin-down plots for cell fill 1 show disappointing lifetimes. The plot for uncoated cell **1.3** (Figure 13) shows a polarization lifetime of around 20 hours. In the absence of cell wall depolarization effects, a 1” spherical target cell filled with 3 amagats of ^{3}He should reach a maximum lifetime of 248 hours according to equation (5). Therefore, the poor lifetime of this uncoated aluminosilicate glass cell indicates that the entire cell fill was unsuccessful.

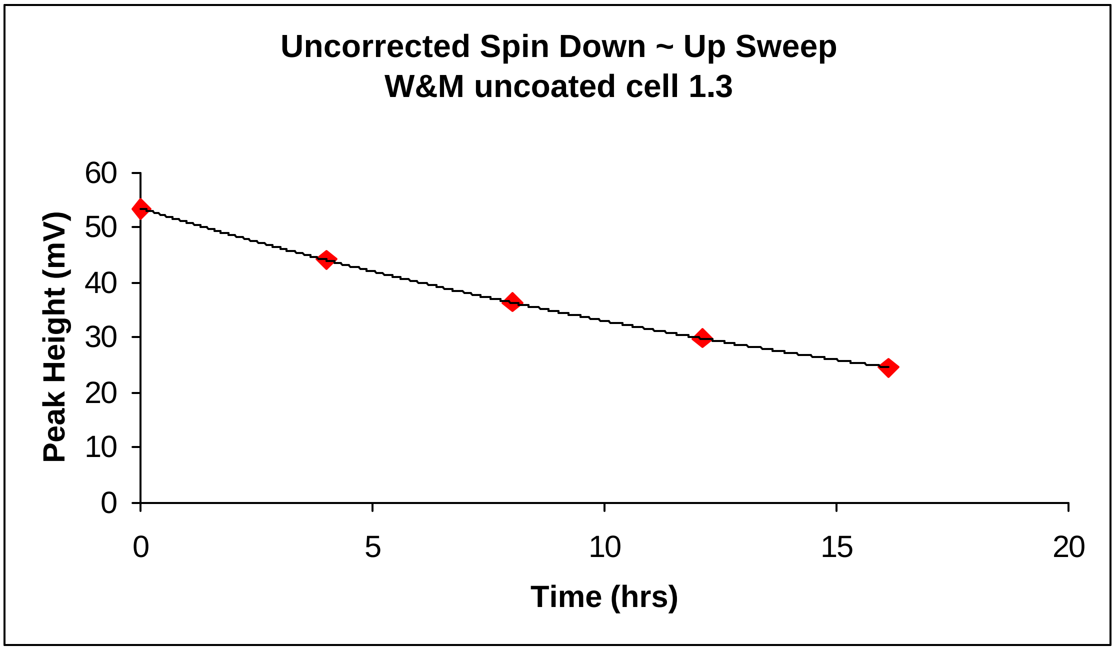

Figure 13: Spin-down plot for uncoated cell **1.3**. The polarization smoothly declines, and the lifetime of the cell, found using the exponential fit from equation (4), is 20.66 hours. However, this lifetime is low because it has not been adjusted for AFP losses.

For this cell fill, the uncoated spheres have average AFP loss percentages of approximately 0.5%. The spin-down of cell **1.3** is corrected by adjusting each data point for the AFP loss and then adding the losses from previous measurements. The corrected lifetime of cell **1.3** is almost 22 hours. Cell **1.4** has an even higher depolarization rate, and its ^{3}He polarization only had a corrected lifetime of about 5 hours.

Cell **1.2**, coated with sol-gel, depolarized more quickly than the uncoated cell, **1.3**. The spin-down plot shows a lifetime of about 17 hours when the data is corrected for the 0.2% AFP loss in polarization (Figure 14).

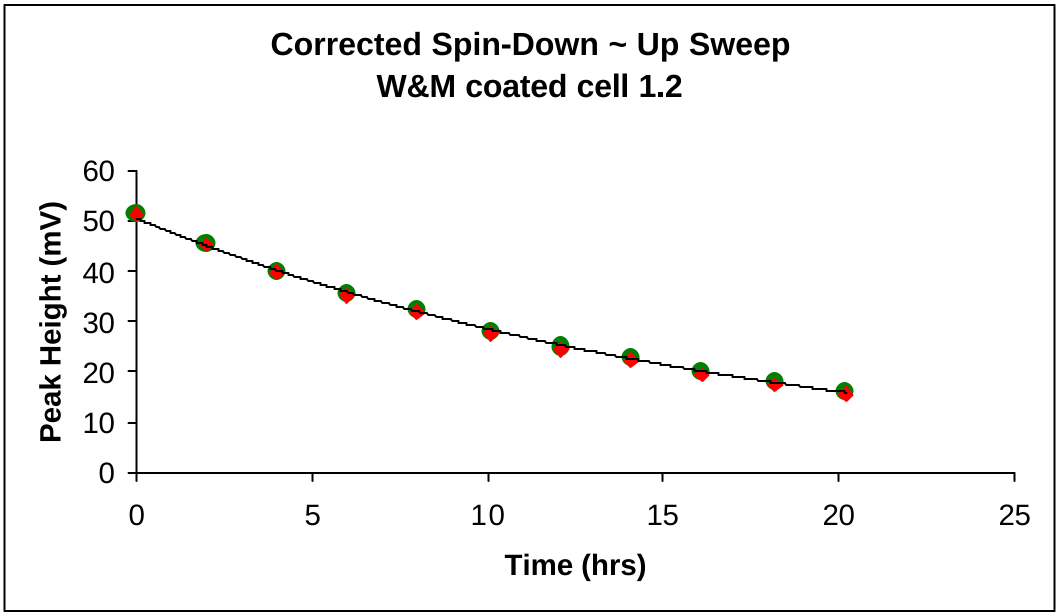

Figure 14: Spin-down plot of cell **1.2** with AFP loss corrections. The circles (green) represent the AFP corrected data, while the diamonds (red) represent the raw spin-down data. The corrected lifetime is 17.3 hours, which was calculated by fitting the AFP corrected data with the exponential found in equation (4).

Cell **1.1**, also coated, depolarized so quickly during testing that only two data points could be recorded in a two-hour period. Thus, the spin-down plot could not be fitted and the lifetime is not known. However, it is obvious that this coated cell has a significantly shorter lifetime than the uncoated cells. The coated cell has an extremely high AFP loss percentage of 3%, which again signifies that this coated cell has a much shorter lifetime than an uncoated cell. The low lifetime values of the coated and uncoated cells indicate that there is a problem with the cell filling procedure.

4.2 Cell Fill 2 (2/7/03)

The next group of spherical cells, also made of GE-180 glass, was coated using the same sol-gel chemistry and dilution as before. However, these cells were allowed to drain and dry longer, and after the densification process, their surfaces appeared to be very smooth. These cells were attached to the stringer by a glass-blower to minimize the torch and glass-working time. For this string, cell numbers **2.1** and **2.3** were coated, and

2.2 and **2.4** were uncoated. These spheres were then baked at a lower temperature of 420°C, and they were filled on February 7, 2003.

This cell fill produced 2 uncoated spheres with promising spin-up curves. Sphere **2.2** has a relatively smooth spin-up curve (Figure 15), which levels off at a high voltage of 58 mV and compares with the spin-up of the good cell seen in Figure 8. Cell **2.4** reached maximum polarization at 38 mV.

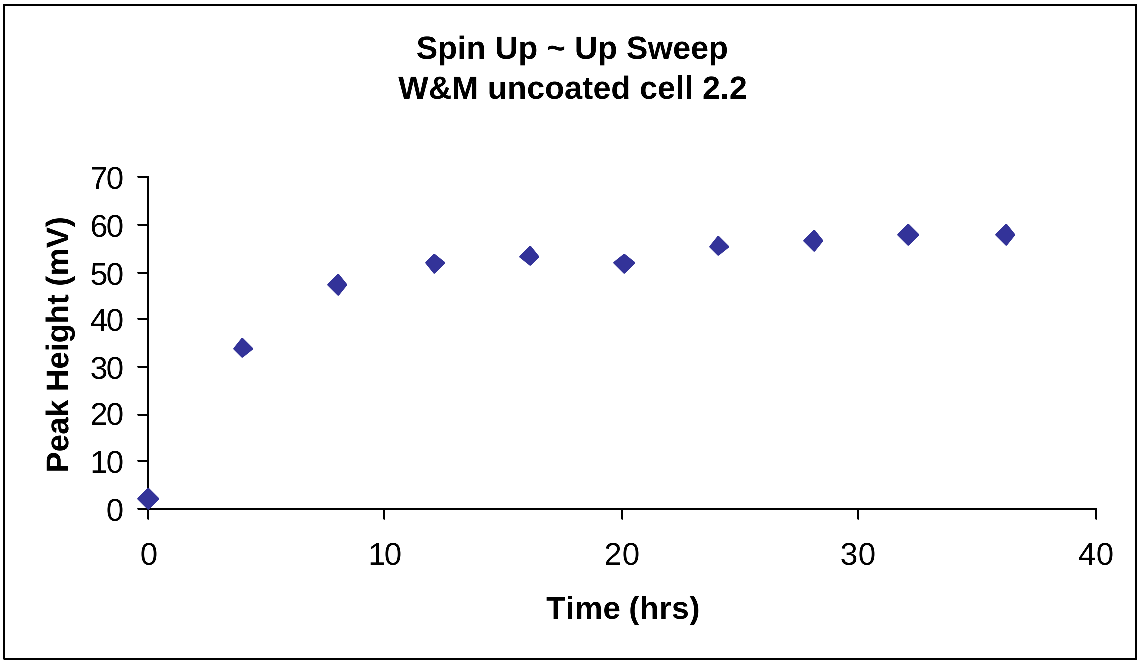

Figure 15: Spin-up plot of uncoated cell **2.2**. The cell polarized quickly, but smoothly, and it leveled off at a high peak voltage of about 58mV. This plot can be compared to the typical spin-up of a good cell seen in Figure 8.

The two coated spheres, **2.1** and **2.3**, reached slightly lower peak voltages of approximately 53 mV and 39 mV. The spin-up of cell **2.1** shows that the ^{3}He polarizes and saturates more quickly than in the uncoated cell (Figure 16). Again, the spin-up plots of the coated cells cannot be successfully fit to the proper equation, which indicates that they have short lifetimes.

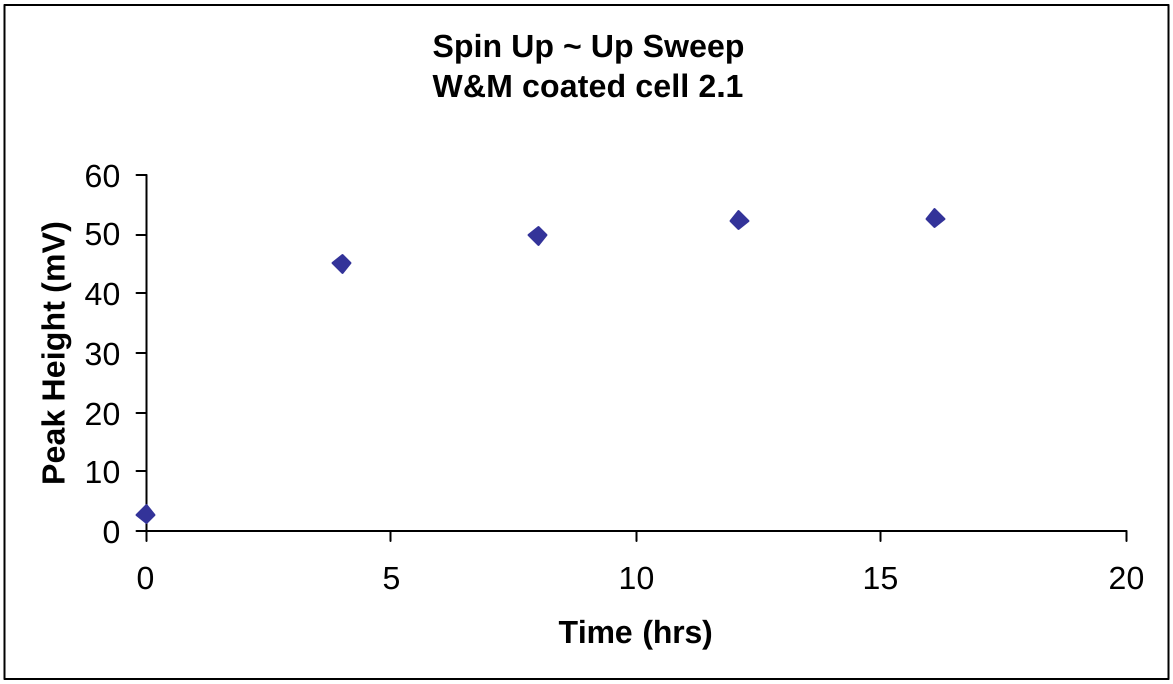

Figure 16: Spin-up plot of coated cell **2.1**. The ^{3}He polarizes more quickly and levels off more sharply, at 53 mV, than that of the uncoated cell.

Uncoated sphere **2.2** shows good spin-down results. It has a long lifetime of 187.4 hours, which is evident from its extremely slow depolarization during the time that data was taken for the spin-down (Figure 17). However, cell **2.4**, also uncoated, only has a raw lifetime of around 27.7 hours.

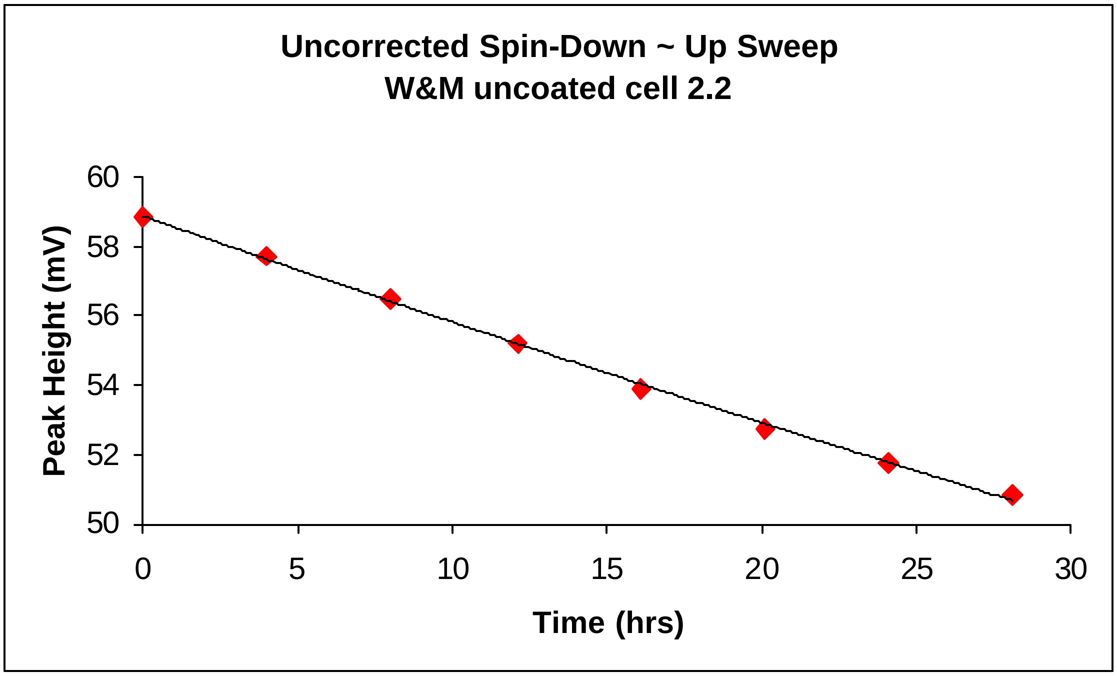

Figure 17: Spin-down plot for uncoated cell **2.2**. This cell depolarizes slowly, and the lifetime of the cell, found using the exponential fit from equation (4), is 187.40 hours. However, this lifetime is low because it has not been adjusted for AFP losses.

The lifetimes of the ^{3}He polarization were also adjusted for AFP losses. The lifetime of cell **2.2** rises to 220.1 hours with a low AFP loss of 0.3% (Figure 18). This result, comparable to the maximum lifetime value of 248 hours for 3 amagat spheres, indicates that the filling procedure itself was successful. The lifetime of sphere **2.4** was also corrected for its AFP losses of about 0.2%, giving a value of 28.4 hours.

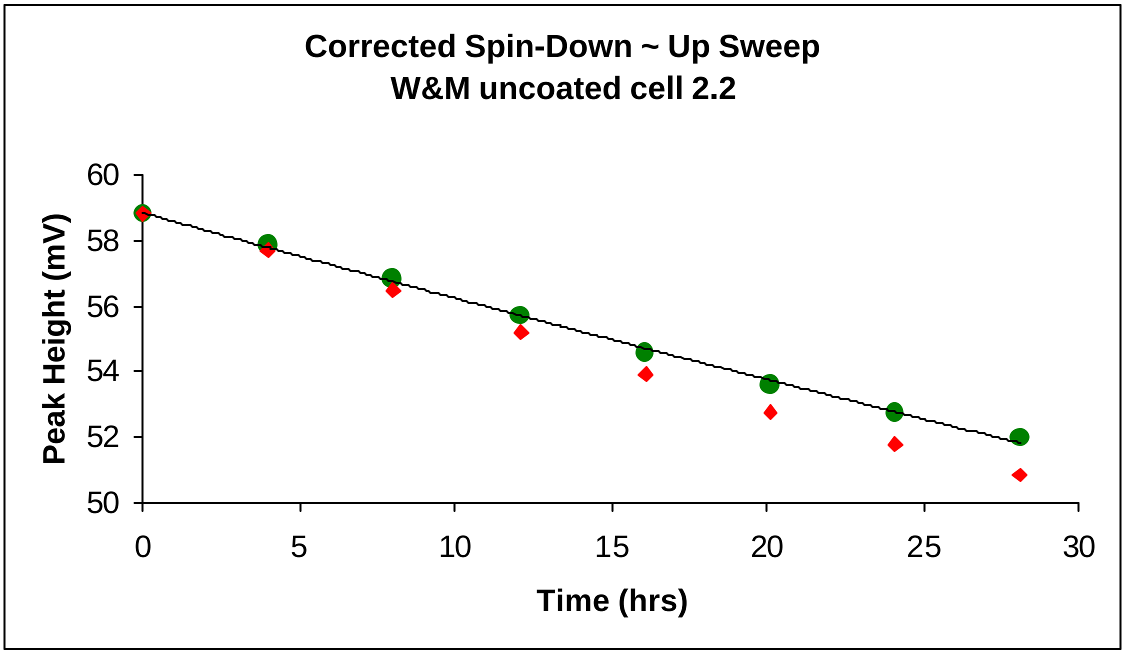

Figure 18: Spin-down plot of cell **2.2** with AFP loss corrections. The circles (green) represent the AFP corrected data, while the diamonds (red) represent the raw spin-down data. The corrected lifetime is 220.1 hours, which was calculated by fitting the AFP corrected data with the exponential found in equation (4). The sizable difference between the corrected and uncorrected data points shows how AFP losses affect the polarization in a long lifetime cell.

Unfortunately, the coated cells **2.1** and **2.3** have extremely short lifetimes of 5.76 and 5.58 hours. The spin-down for sphere **2.1** is a smooth exponential that shows a fast depolarization rate (Figure 19). The coated cells have very large AFP losses ranging from 0.75% to 1.6%, and their lifetimes are corrected to 6.47 and 5.93 hours.

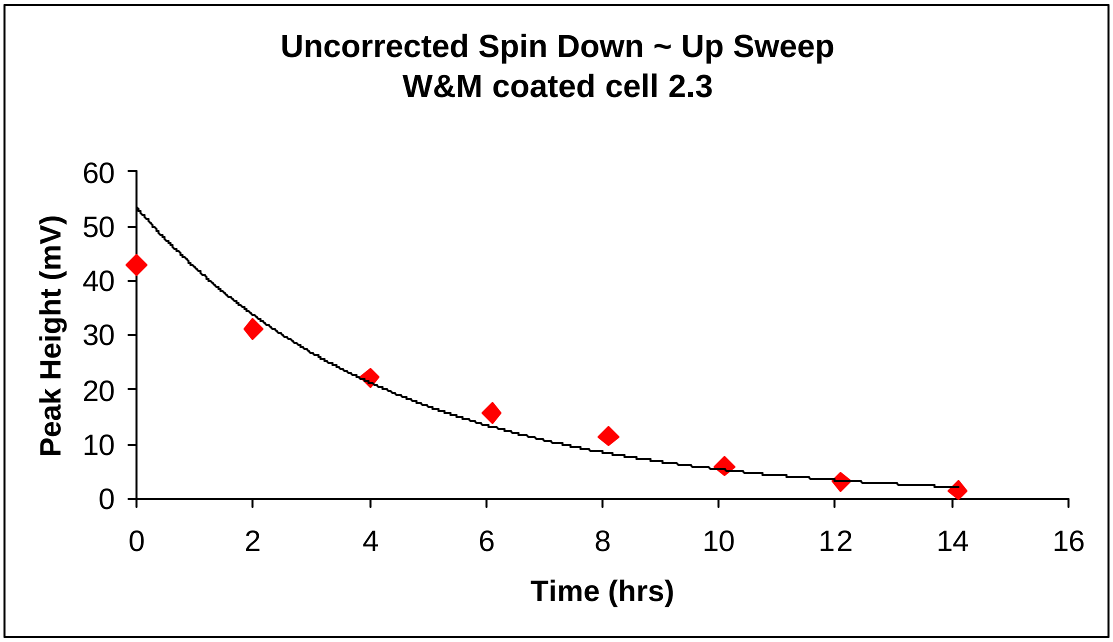

Figure 19: Spin-down plot for uncoated cell **2.3**. The polarization quickly declines, and the lifetime of the cell, found by using the exponential in equation (4) to fit the data, is 5.93 hours. However, this lifetime is low because it has not been adjusted for AFP losses.

4.3 Cell Fill 3 (4/03)

After testing the second group, it was decided that a more dilute sol-gel solution might coat the surface more smoothly with less build-up. In the final group of GE-180 glass spherical cells, three were coated with different sol-gel solutions. Cell **3.1** was coated using the same solution as before. Cell **3.2**, however, was coated with a sol-gel solution that was diluted with 83mL of ethanol, and cell **3.3** was coated with a solution diluted with 123mL of ethanol. Cell **3.4** was left uncoated. As with the second group of cells, the coated cells were given ample time to drain and dry before densification. Afterwards, cell **3.2** was the only one that appeared to have a slight ring of sol-gel around the sphere. These cells were also baked at 420°C and special care was taken as to not heat the coated glass with the torch while rubidium was chased into the spheres. These cells were filled on May 2, 2003.

Sphere **3.1** reached a maximum polarization of 17 mV. Like the spin-up plots of the coated cells from previous fills, this sphere polarized quickly and leveled off at a low maximum polarization. An extremely short lifetime of 1.8 hours was calculated. When corrected for the large AFP losses, which averaged around 3.3%, the lifetime rose to 2 hours. Like the other cells coated with this solution, **3.1** has a short lifetime and does not improve the depolarization rates of ^{3}He.

The sphere coated with a further diluted solution, **3.2**, revealed much better results when first tested. The cell's spin-up plot is comparable to Figure 8, and the polarization levels off at 67 mV. Using the fit from equation (4) for the raw spin-down data, a lifetime of 116 hours was calculated (Figure 20).

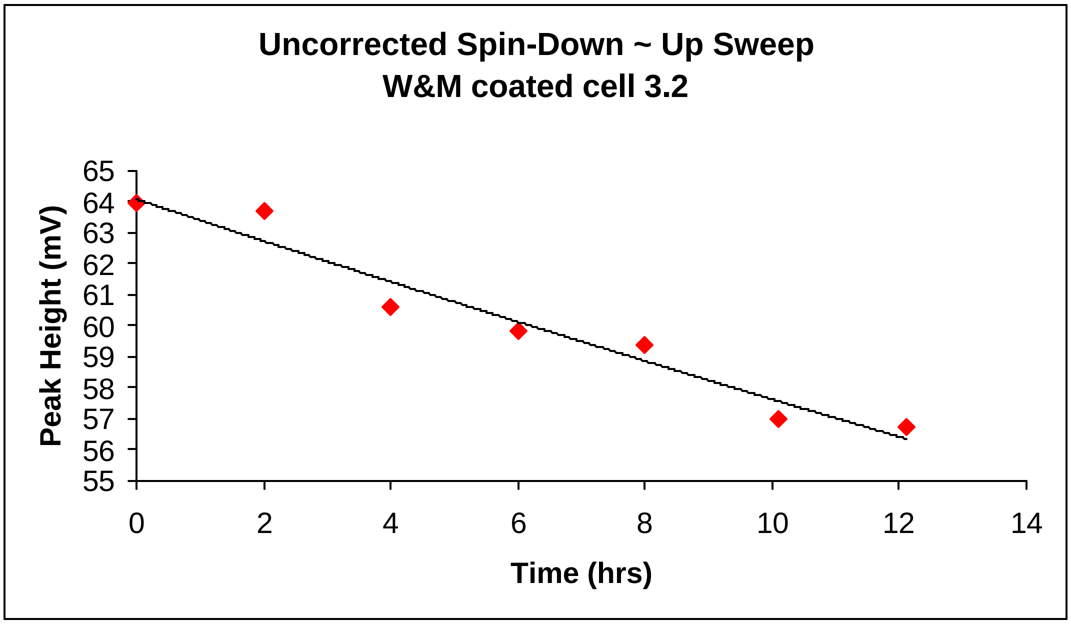

Figure 20: Spin-down plot for coated cell **3.2**. The polarization slowly declines, and the lifetime of the cell, found by using the exponential in equation (4) to fit the data, is 115.6 hours. However, this lifetime is low because it has not been adjusted for AFP losses. This plot shows an odd depolarization trend, therefore, a second characterization was performed and spin-down measurements were only made every 4 hours.

When corrected for the high AFP loss of 0.9%, a significant change in the lifetime was found. With a 214 hour lifetime, cell **3.2** is clearly a long-lifetime cell. From Figure 20, it is evident that an odd depolarization trend occurred when measurements were taken every two hours. Therefore, it was decided to retest the cell and take NMR measurements every four hours during the spin-down. On the second characterization, which took place three days later, a maximum polarization of 49 mV was reached. The polarization lifetime was greatly reduced to 13.2 hours, which was corrected to 14.2 hours using the AFP loss of 0.9%. Such a difference in lifetimes may imply that the sol-gel coatings are not durable, and they cannot withstand numerous heating and cooling processes.

The cell coated with the most diluted solution did not perform as well. Maximum polarization was reached at 38 mV, however, the ^{3}He depolarized quickly. A lifetime near 6.3 hours was corrected to 6.4 hours using the AFP loss of 0.3%.

Cell **3.4** is an uncoated sphere that revealed surprising results. A maximum polarization of only 1mV was reached, and the cell depolarized so quickly that data could not be taken for the AFP loss test or spin-down. This data may imply that the cell was lacking a sufficient amount of Rb or that the sphere was not filled with enough N_2 during the filling process.

Cell Fill	Sphere #	Coating	Ethanol (mL)	Maximum Polarization (mV)	Lifetime (hours)	AFP Loss (%)	AFP corrected Lifetime (hours)
1	**1.1**	sol-gel	43	10	*	2.95	*
	1.2	sol-gel	43	52	16.8	0.20	17.3
	1.3	none	--	58	20.7	0.60	21.7
	1.4	none	--	36	4.8	0.55	4.8
2	**2.1**	sol-gel	43	53	5.8	1.53	6.5
	2.2	none	--	59	187.4	0.30	220.1
	2.3	sol-gel	43	39	5.6	0.79	5.9
	2.4	none	--	38	27.7	0.24	28.4
3	**3.1**	sol-gel	43	17	1.8	3.30	2.0
	3.2	sol-gel	83	67	115.6	0.94	213.9
	(after 3 days)			49	13.2	0.94	14.2
	3.3	sol-gel	123	36	6.3	0.33	6.4
	3.4	none	--	1	*	*	*

Table 3: Summary of the data gathered from each of the three cell fills. (*) Cells **1.1** and **3.4** depolarized so quickly that sufficient data could not be collected for a spin-down plot. Therefore, a lifetime and AFP corrected lifetime could not be calculated due to the lack of data for a spin-down plot.

4.4 Error

There are many sources of error that may affect the experimental results. First, a fitting program is used to find the maximum peak height of each NMR sweep. Parameters, such as full-width-half-max and resonance field value, are adjusted in order to fit the data. Minor changes in the parameters cause only a slight change in the best-fit peak height. The resultant peak heights experience a 0.5% to 1.0% variation according to what values are chosen for the parameters. Fitting spin-down data is also a source of error. The deviation of the data points from the best-fit line can be examined through the chi-squared values. AFP losses also vary from point to point, and these losses pose as a source of error in the calculation of corrected polarization lifetimes. Long-lifetime cells are greatly affected by AFP losses, thus any deviations in the AFP loss would change the measured lifetime of the ^{3}He polarization. The AFP losses for cell **2.2**, the uncoated long-lifetime sphere, have a small standard deviation of 0.007492. The spin-down peak heights at positive and negative deviation were calculated and plotted against the average

AFP corrected data. When the AFP losses are corrected with one positive deviation, the exponential fit of the spin-down plot yields a new calculated lifetime of 221.0 hours (Figure 19).

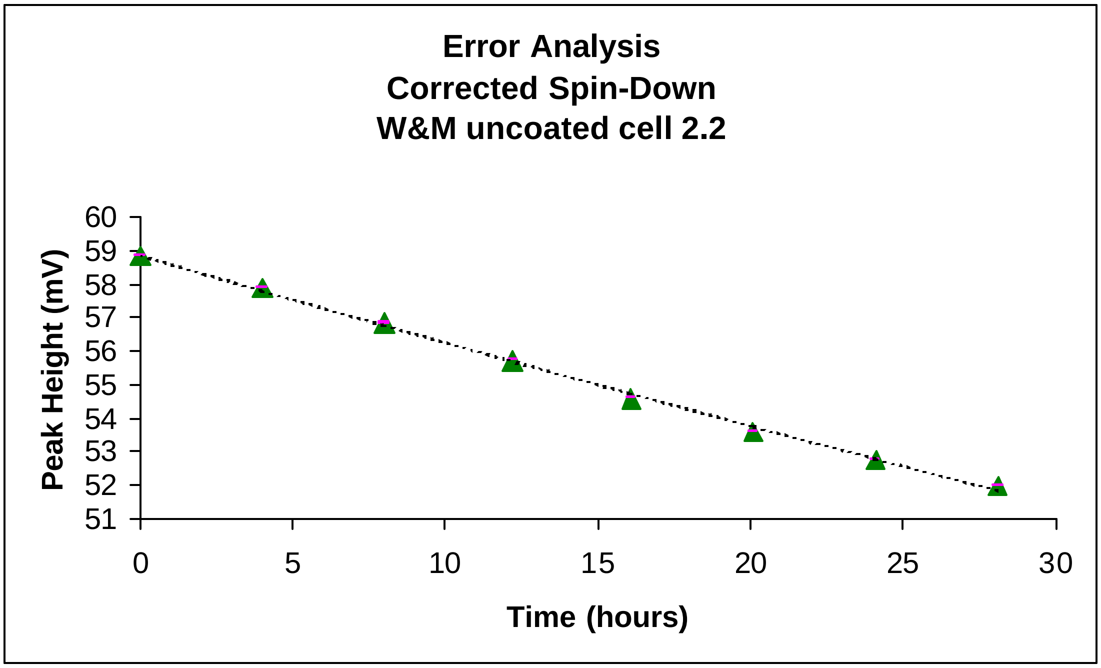

Figure 19: Spin-down plot corrected for AFP losses. The triangles (green) represent the corrections for the average AFP loss percentages. The bars (pink) represent the corrections for one positive standard deviation in the AFP losses. A new lifetime of 221.0 hours was measured when the error was taken into consideration.

Similarly, when the AFP losses are corrected with one negative standard deviation, a new lifetime of 219.2 hours was calculated (Figure 20).

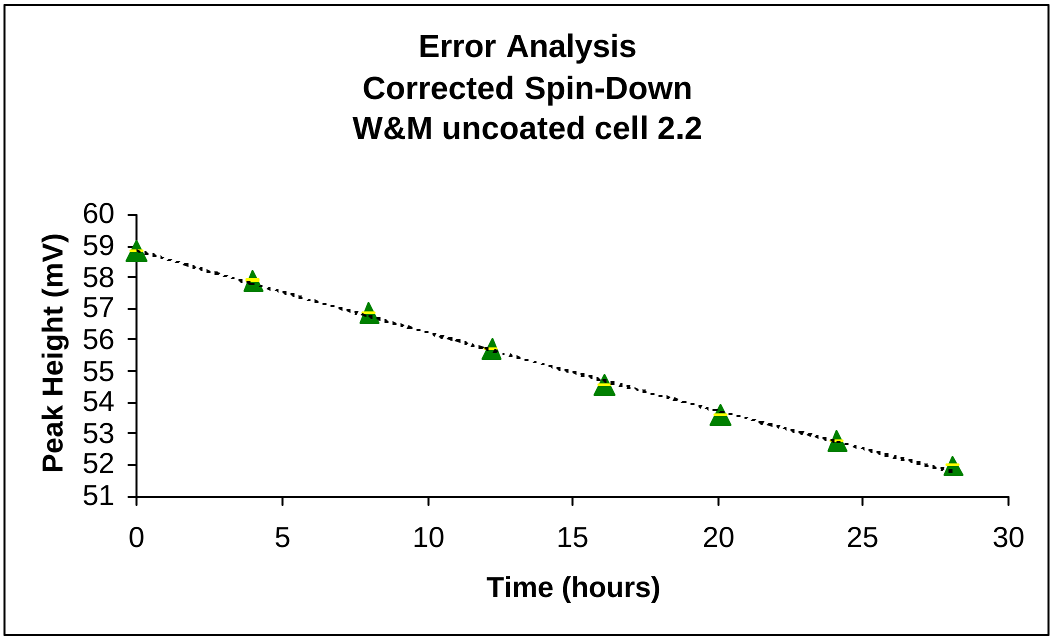

Figure 20: Spin-down plot corrected for AFP losses. The triangles (green) represent the corrections for the average AFP loss percentages. The bars (yellow) represent the corrections for one negative deviation in the AFP losses. A new lifetime of 219.2 hours was measured when the error was taken into consideration.

Overall, for the corrected lifetime, the error due to uncertainty in AFP losses for each measurement was calculated to be about 0.5%. Though the AFP losses in short-lifetime cells are larger and have greater uncertainty, the effect is less noticeable because the losses have significantly less time to accumulate due to the fast depolarization rate. Though peak height fitting techniques and curve fitting may induce slight errors, the deviation in AFP losses for each NMR measurement may be the largest source of error in the polarization lifetime values.

5 Conclusions and Future Research

In order to perform nuclear physics experiments with ^{3}He, a high degree of polarization, with a long lifetime, must be achieved. It is possible that coating a glass cell with sol-gel will leave a smooth surface without impurities and fissures, thus reducing the relaxation of ^{3}He polarization due to the atom's collisions with the cell wall. To test the benefits of sol-gel, cells were coated, filled, polarized, and characterized in an NMR system.

For this Senior Honors Thesis, I have tested three sets of four 1" spherical cells. Upon testing the first group of spheres, the sol-gel coated cells did not yield high polarizations or long lifetimes. This may have implications about the sol-gel theory or coating method, however, these results are extremely inconclusive because the uncoated cells generated disappointing results as well. They both achieved poor polarization and had extremely low lifetimes. It is possible that the glass string may have been contaminated by the torch and glass blowing procedures necessary for the attachment of the sol-gel coated cells. The fact that the entire string of cells rendered bad results implies that the cell filling techniques need improvement before any conclusive evidence can be gathered concerning sol-gel coatings.

Glass blowing and filling techniques were carefully reviewed for the next group of cells, and more telling results were gathered. The long lifetime of one of the uncoated spheres ensured that the cell fill was successful, and any results concerning the coated cells rested upon the effectiveness of the sol-gel. Unfortunately, the coated cells had short lifetimes, implying that the sol-gel coating worsened the depolarization of ^{3}He. This does not disprove the effectiveness of all sol-gel coatings. After the coated cells were characterized, their coating appeared to be yellow in color with noticeable lines showing an uneven surface. Again, it appears that the sol-gel was not drained and dried properly or the solution was too thick.

To test the effectiveness of different coating thicknesses, various dilutions of the basic sol-gel solution were tried on the final group of cells. The cells coated with the thickest and thinnest solutions had short lifetimes and spin-up plots similar to that of the previously tested coated cells. The sphere coated with the solution diluted with 83mL of ethanol yielded an extremely long lifetime. This cell's AFP corrected lifetime of 214 hours proves that it is possible to make a long-lifetime sol-gel coated cell. However, when the cell was characterized again three days later, the polarization lifetime was significantly reduced. It is possible that the coating is unable to withstand repetitive heating and cooling processes. This may crack the coating and significantly decrease its effectiveness on reducing depolarization. Also, upon heating, the vaporized Rb in the cell may be interacting with the sol-gel coating. Finally, the uncoated sphere was the most perplexing out of this cell fill. The cell barely polarized, and depolarization occurred so quickly that measurements could not be made. Cell **3.4**, like **1.4** and **2.4**, had a small lifetime, which may imply something about the cell filling procedure. As the last sphere in the row of cells on the stringer, the fourth cell may not be getting the necessary amount of Rb for the spin exchange process. Also, because the fourth cell is the last to be pulled off from the stringer, it may be lacking the volume of N_2 necessary to ensure maximum Rb polarization. Without the uncoated cell results from this cell fill, the true performance of the sol-gel coatings is inconclusive. It is unknown as to whether the long-lifetime coated cell actually showed an improvement in the lifetime of polarized ^{3}He.

Though experimentation with different dilutions may prove to be worthwhile, there are numerous other paths that may be taken to get a sol-gel coating that improves polarization lifetimes. First, the chemistry of the sol-gel solution itself may be changed. The addition of aluminum nitrate may be helpful in getting a coating that resembles the desired aluminosilicate glass. More dilutions and densification techniques may also be tested to ensure a smooth surface with a desirable thickness, which can be examined using a scanning electron microscope. Also, testing sol-gel coatings on different types of glass, such as Pyrex, may prove to be helpful. Improvements on cell filling techniques should also be explored. Throughout all of the experimentation with sol-gel coating techniques, the ultimate goal still remains to improve the lifetime of polarized ^{3}He for use in Jefferson Lab nuclear physics experiments.

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