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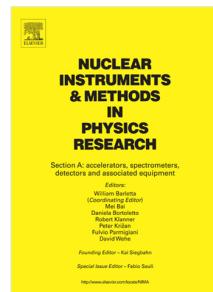
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¹ Evaluation of commercial nickel-phosphorus coating for
² ultracold neutron guides using a pinhole bottling
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¹⁸ **Abstract**

We report on the evaluation of commercial electroless nickel phosphorus (NiP) coatings for ultracold neutron (UCN) transport and storage. The material potential of 50 μm thick NiP coatings on stainless steel and aluminum substrates was measured to be $V_F = 213(5.2)$ neV using the time-of-flight spectrometer ASTERIX at the Lujan Center. The loss per bounce probability was measured in pinhole bottling experiments carried out at ultracold neutron sources at Los Alamos Neutron Science Center and the Institut Laue-Langevin. For these tests a new guide coupling design was used to minimize gaps between the guide sections. The observed UCN loss in the bottle was interpreted in terms of an energy independent effective loss per bounce, which is the appropriate model when gaps in the system and upscattering are the dominate loss mechanisms, yielding a loss per bounce of $1.3(1) \times 10^{-4}$. We also present a detailed discussion of the pinhole bottling methodology and an energy dependent analysis of the experimental results.

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²¹ **1. Introduction**

²² Ultracold neutrons have been recognized as an excellent tool for probing
²³ our understanding of the fundamental laws of nature, and new programs of
²⁴ research are fast-developing worldwide. Experiments are planned or have been
²⁵ carried out at UCN sources to measure the neutron's lifetime [1–6] and electric
²⁶ dipole moment [7–10], angular correlations in neutron β -decay [11, 12], and
²⁷ gravitational energy states [13, 14]. Many of these experiments are statistics
²⁸ limited and several new UCN sources [15–17] in development promise to deliver
²⁹ the required densities to achieve higher precision measurements. To deliver
³⁰ the required statistics for current and future experiments, the spallation UCN
³¹ facility at the Los Alamos Neutron Science Center (LANSCE) [18] is upgrading
³² several key components of their source [9], including the UCN guide system out
³³ of the source. The primary motivation for this upgrade is to achieve a UCN
³⁴ density > 10 UCN/cc, which will enable a future room temperature neutron
³⁵ electric dipole moment search with a goal sensitivity of 3×10^{-27} e-cm.

³⁶ UCN have sufficiently low kinetic energy that they can be totally internally
³⁷ reflected in material bottles. The interaction of the UCN and a material wall is
³⁸ described by the effective Fermi potential

$$V_F = V - iW = \frac{2\pi\hbar^2}{m_n} Na - i\frac{\hbar}{2} N\sigma v, \quad (1)$$

³⁹ where m_n and v are the mass and velocity of the neutron, N is the number
⁴⁰ density of the atoms in the material, a is the coherent neutron scattering length
⁴¹ of the material, and σ is the loss cross section (See e.g. Ref.[19] and the refer-
⁴² ences found therein). The real part of the Fermi potential gives the maximum
⁴³ energy of neutrons that can be confined, and the imaginary part describes the
⁴⁴ absorption and upscattering. The ratio of the imaginary and real parts of V_F ,
⁴⁵ $f = W/V$, is defined as the loss factor. The loss probability $\mu(E)$, as a function

46 of the UCN kinetic energy E , for a bottle of material potential V is given by

$$\mu(E) = 2f \left[\frac{V}{E} \sin^{-1} \left(\frac{E}{V} \right)^{1/2} - \left(\frac{V}{E} - 1 \right)^{1/2} \right], \quad (2)$$

47 after averaging over all angles of incidence [20]. Typically, the energy scale of V_F
 48 is 100's of neV with ^{58}Ni possessing the largest Fermi potential $V_F = 342$ neV
 49 among commonly available materials. A convenient quirk of nature sets the
 50 energy scale for interaction with magnetic fields, $V_{mag} = -\vec{\mu} \cdot \vec{B} \sim 60$ neV/T,
 51 and earth's gravity, $V_g = m_n g h \sim 100$ neV/m, on similar footing. Experiments
 52 have exploited these interactions to fully polarize a "beam" of UCN using a large
 53 magnetic field or significantly alter the energy of UCN by raising or lowering
 54 their apparatus a meter or two relative to the beam height.

55 In the LANSCE source, 6 m of electro-polished stainless steel tubes guide
 56 the UCN from the source volume to the experimental area. Stainless steel has
 57 an effective material potential of $V_F = 188$ neV. The current guide system has a
 58 loss-per-bounce probability of 5.2×10^{-4} [18], which includes the effect of gaps
 59 between the 1 m guide sections. Improving the transport from the source volume
 60 to the experimental hall will have a significant impact on achieving desired UCN
 61 densities.

62 Pure nickel coatings provide a high Fermi potential for bottling UCN; how-
 63 ever, the probability of spin-flip is unacceptably high for applications requiring
 64 polarized UCN. Nickel alloy coatings, such as nickel molybdenum or nickel-
 65 vanadium, have been shown to have high Fermi potential (210-220 neV), high
 66 specularity, and low spin-flip probability [21]. Electroless nickel plating has been
 67 used since the 1950's to provide robust corrosion resistant uniform coatings on
 68 conductive and non-conductive substrates. Unlike electro-plating, this method
 69 uniformly coats objects with complex geometries. When the phosphorus con-
 70 tent is in excesses of 10% by atomic composition the coating is non-magnetic
 71 at room temperature[22-24]. The combination of the robust nature of the coat-
 72 ing, the commercial availability, high Fermi potential, and the non-magnetic
 73 surfaces make nickel phosphorus (NiP) an ideal candidate for a UCN trans-
 74 port guide coating material. NiP coated guides have been in use at the UCN

75 source in Osaka for more than a decade [25]. Our group recently reported on a
 76 measurement of the spin-flip probability for UCN interacting with NiP coated
 77 stainless steel and aluminum neutron guides [26]. The results of that study
 78 show the spin-flip probability to be $\beta = 3.3^{1.8}_{-5.6} \times 10^{-6}$, which is sufficient for
 79 experiments requiring the transport and storage of polarized UCN.

80 In this paper we report on a suite of measurements of the bottle lifetime
 81 of NiP coated stainless steel guides performed at the PF2 facility at the Insti-
 82 tut Laue Langevin [27] and LANSCE UCN [18] sources. The purpose of the
 83 measurements was to evaluate the overall performance of the guide technology
 84 chosen for our new UCN source, including the surface properties for UCN as
 85 well as the guide coupling design. In Sec. 2 the results of a measurement of
 86 the real part of the Fermi potential and an estimate of the imaginary part are
 87 presented. Section 3 summarizes the guide preparation and coating. Section 4
 88 details the double pinhole method. Sections 5 and 6 present the measurements
 89 performed at LANL and the ILL, respectively. Section 7 provides a discussion
 90 of our results.

91 **2. Determination of the NiP Fermi Potential**

92 *2.1. Measurement of the Real Part of V_F*

93 The Fermi potential of NiP coated surfaces was measured using the time-
 94 of-flight spectrometer ASTERIX at the Lujan Neutron Scattering Center at
 95 LANSCE. ASTERIX views an intense polychromatic neutron beam through a
 96 neutron guide with a 36 cm^2 cross-sectional area and utilizes the wavelength
 97 band λ from 4 to 13 Å. The divergence of the neutron beam and its footprint on
 98 the beam at the sample can be controlled with two sets of slits. Two aluminium
 99 and two stainless steel coupons, with surface area of $50 \times 50 \text{ mm}^2$, were coated
 100 with $50 \mu\text{m}$ thick films of nickel phosphorus by Chem Processing, INC [28] us-
 101 ing the electroless process described in Section 3. The sample coupons had an
 102 average surface roughness of 16-32 micro-inches prior to coating. Neutron re-
 103 flectometry experiments were performed at the neutron incidence beam angle θ

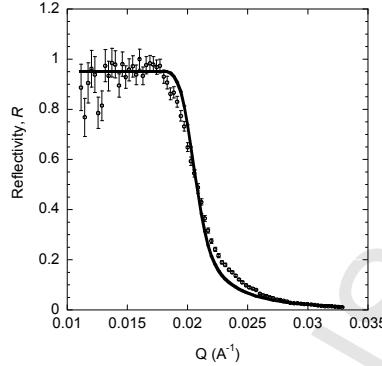


Figure 1: The fitted reflectivity curve (solid line) for the sample 3. The scattering length density of the film was $8.2(2) \times 10^{-6} \text{ \AA}^{-2}$.

104 of 0.7° . To minimize the effect of surface roughness, mostly from polishing and
 105 machining,² the beam was aligned along the polishing marks.

106 The obtained $I(\theta; \lambda)$ data was reduced and binned to obtain the reflected
 107 beam intensity, R , as a function of the neutron momentum transfer vector Q .
 108 R is normalized to the intensity of the incoming beam (and therefore equal ≈ 1
 109 below the critical momentum transfer vector, Q_c) and $Q = 4\pi \sin \theta / \lambda$. Fig. 1
 110 shows the reduced $R(Q)$ results for the NiP coating on a stainless steel sample.

111 The $R(Q)$ curves were fit using an open-source reflectivity package, MO-
 112 TOFIT, which runs in the IGOR Pro environment [29]. Using the Abeles matrix
 113 formalism, a theoretical reflectometry curve can be calculated and compared to
 114 the measured data. Both generic optimization and Levenberg–Marquardt non-
 115 linear least-square methods were employed to obtain the best fits with the low-
 116 est χ^2 values. The only two parameters varied in the fitting procedure were the
 117 scattering length densities of the thin coating films and the beam normalization
 118 parameters to account for imperfect beam normalization. The deviation of the
 119 data from the fit at the inflection points is due to imperfections in the sample

²Roughness here is defined as combined effect of sample surface deviation from mathematically sharp interface.

Table 1: Results of scattering length density measurement at the ASTERIX spectrometer. Four samples were tested: two aluminum coupons and two stainless steel coated with 50 μm of nickel-phosphorus. N_b is the measured scattering length density and V_F is the Fermi potential. The uncertainty in the measurement of the scattering length density was dominated by the surface roughness of the samples.

Sample	Description	$N_b \times 10^{-6} [\text{\AA}^{-2}]$	V_F [neV]
1	NiP on Al	8.15(20)	212.2(5.2)
2	NiP on Al	8.10(20)	210.9(5.2)
3	NiP on SS	8.20(20)	213.5(5.2)
4	NiP on SS	8.20(20)	213.5(5.2)

and does not affect the determination of the mean scattering length density used to calculate the Fermi Potential.

The values of the scattering length density obtained from the measured critical momentum transfer are shown in Table 1. The scattering length density N_b is related to the Fermi potential described in Eq. 1 as $N_b = Na$, the product of the number density and the scattering length. The Fermi potential obtained from the measured scattering length density is also listed in Table 1. The measured Fermi potential is consistent with our expectation based on the phosphorus content and density of NiP found in the literature.

2.2. Calculation of the Nickel Phosphorus loss factor

The imaginary part of the Fermi potential for the NiP coating can be estimated using Eq. 1 and summing over the constituent elements weighted by their density in the mixture,

$$W = \frac{\hbar}{2} \sum_i N_i \sigma_l^{(i)} v. \quad (3)$$

In this calculation the measured thermal neutron absorption cross section, σ_{abs} , is used for the loss cross-section, neglecting contributions from inelastic scattering ($\sigma_{abs}^{Ni} = 4.49$ b and $\sigma_{abs}^P = 0.172$ b) [30]. The phosphorus content of the coating is 10.5(25)% by weight. The NiP density is 7.8 g/cm³ [31]. Using the measured Fermi potential this results in estimates for the imaginary part of the

¹³⁸ potential of $W = 2.30(6) \times 10^{-2}$ neV and a loss factor of $f = 1.08(4) \times 10^{-4}$,
¹³⁹ where the uncertainty is dominated by the variation in the ratio of phosphorus
¹⁴⁰ to nickel in the coating.

¹⁴¹ **3. Guide Preparation**

¹⁴² The UCN guides were constructed from 7.62 cm OD, 7.29 cm ID Valex
¹⁴³ [32] 316L stainless steel alloy tubing with an internal surface finish of 10 Ra
¹⁴⁴ manufactured to Valex's 401SP specifications. Four guides were fabricated,
¹⁴⁵ denoted 1-4 in the following. Guides 1 and 2 were 70 cm long and guides 3 and
¹⁴⁶ 4 were 140 cm in length. In the new guide coupling design the tube was welded
¹⁴⁷ into a socket, and the surface was smoothed to minimize any surface impact at
¹⁴⁸ the weld joint, as shown in Fig. 2. In this configuration, guides mate to each
¹⁴⁹ other by inserting a thin coupling plate between them, as shown in Fig. 3. The
¹⁵⁰ flush surface is important to limit gaps, an important source of loss in UCN
¹⁵¹ transmission. After machining, the guides were cleaned with a warm water
¹⁵² (63° C) Alconox solution (1% by weight) and rinsed with de-ionized water and
¹⁵³ isopropyl alcohol. All guides were coated with a high phosphorus³ NiP mixture
¹⁵⁴ by ChemProcessing, INC. Guide 4 was electro-polished prior to coating ⁴. The
¹⁵⁵ coating process is an electroless chemical dip that uniformly coats all surfaces
¹⁵⁶ of the guide with 50 μm of NiP, described in more detail in Ref. [26]. This
¹⁵⁷ process exacerbates the inherent roughness of the substrate which can increase
¹⁵⁸ the probably of diffuse scattering. The coupling plate and pinhole flanges were
¹⁵⁹ coated in the same NiP as the guides.

¹⁶⁰ A previous iteration of the guide coupling design used ConFlat flanges to
¹⁶¹ mate the guides to each other. A ConFlat flange was welded to the outside of

³The phosphorus content in Chem Processing INC's high phosphorus coating is 10.5(25)% by weight.

⁴The Valex tubing used was electro-polished as part of the manufacturing process. The electro-polishing referred to here is after machining the guides to length and welding on the mating flanges.

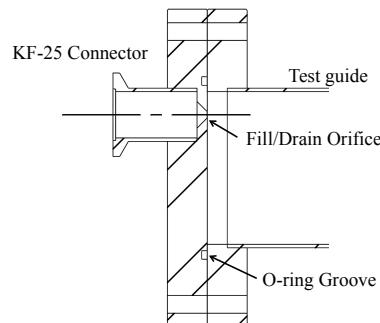


Figure 2: Schematic of the pinhole flange mating to the guide flange. A standard KF-25 nipple, welded to the flange, allows the test guide to be connected to a UCN guide or a ^{10}B ZnS:Ag detector assembly.

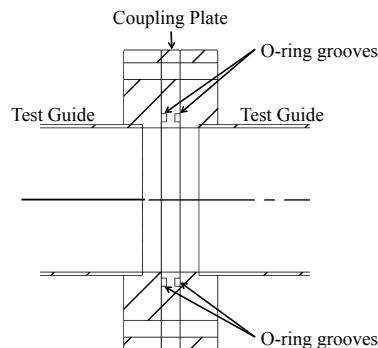


Figure 3: Schematic of test guides mated together using a coupling plate. Grooves cut in the coupling plate hold o-rings which seal against the flat surface of the flange. The coupling plate was coated in NiP.

162 the guide with the design goal that the flange's sealing surface and the end of
 163 the guide were coplanar, in order to minimize the gap between the guides. The
 164 width of the gap created in this design was measured for the flanges of four
 165 guides using Plastigauge ⁵. The results showed the ConFlat design had gaps
 166 varying between 100-340 μm at different positions around the same flange. The
 167 nonuniformity of the gaps was attributed to a slight misalignment between the
 168 flange and the end of the guide after welding.

169 **4. Experimental Method**

170 *4.1. Double pinhole approach to loss probability measurement*

171 Focusing on the specific example of this technique described in Brenner et al
 172 [33], a measurement of the loss probability was performed by trapping UCN in
 173 a bottle made of a test guide and shutters on the ends. The upstream shutter
 174 is opened and the downstream shutter is closed to fill UCN into the bottle.
 175 After the UCN density in the bottle saturates (when the fill rate is balanced by
 176 the loss rate), the upstream shutter is closed. Both shutters remain closed for
 177 a holding time varied from a few seconds to several hundred seconds and the
 178 surviving UCN are emptied and counted by opening the downstream shutter.
 179 The lifetime of the bottle is then determined from a double exponential fit to
 180 the number of unloaded UCN versus holding time, where the short τ_s and long
 181 τ_l time constants are attributed to the lifetimes of untrappable and trappable
 182 UCN. Untrappable UCN are assumed to have kinetic energy greater than the
 183 Fermi potential of the bottle walls and the loss rate of this class of UCN is
 184 dominated by the time required to have a near-normal incidence collision with
 185 the walls, which is influenced by the probability of diffuse scattering.

186 The measured long time constant τ_l is related to the bottle lifetime τ_{bottle}
 187 by

$$\tau_l^{-1} = \tau_\beta^{-1} + \tau_{bottle}^{-1} + \tau_{gap}^{-1} + \tau_{shutter}^{-1}, \quad (4)$$

⁵<http://www.plastigaugeuse.com>

188 where τ_{bottle} includes all possible loss contributions from the surface of the guide
 189 being tested, $\tau_\beta = 880.1(1.1)\text{s}$ is the neutron lifetime [34], τ_{gap}^{-1} the loss rate in
 190 the gaps between the guide and the shutter, and $\tau_{shutter}^{-1}$ is the loss rate on the
 191 shutter. A possible source of systematic uncertainty is then the reproducibility
 192 of the gap between the guide and the shutter from fill to fill.

193 In the measurements presented in Section 5 and Section 6 an alternative
 194 method of measuring the loss probability was adopted, using two fixed pinhole
 195 plates [35]. The bottle is created by fixing plates to the ends of the test guide,
 196 where each plate has a small hole accounting for $< 10^{-4}$ of the total surface
 197 area of the bottle. As shown below, in order for this method to be sensitive to
 198 the loss probability of the UCN guide surface, the loss probability through the
 199 holes, given by the ratio of the area of the holes to the surface area of the bottle,
 200 should be less than or equal to the expected material loss probability, which in
 201 this case is $\approx 10^{-4}$. A gate valve upstream of the bottle is used to control the
 202 flow of UCN to the entrance hole. Downstream of the bottle a detector is closely
 203 coupled to the exit hole to monitor the rate of UCN leaving the bottle. The gate
 204 valve is opened for a time t_{fill} equal to several bottle lifetimes to fill the bottle
 205 close to the maximum equilibrium density of UCN. After t_{fill} , the upstream
 206 gate valve is closed and the rate at which the density of UCN in the bottle
 207 decreases is monitored in the downstream detector. An absorber is mounted
 208 to the downstream side of the upstream gate-valve to prevent UCN that exit
 209 the bottle through the upstream hole from re-entering the storage volume. In
 210 the ideal case of perfectly lossless coating, no β -decay, and no gaps, the time
 211 dependence of the rate measured by the downstream detector would be similar
 212 to charging and discharging a capacitor, with a time constant governed by (for
 213 example, assuming an isotropic UCN velocity distribution, see [19] p. 91)

$$\tau_{hole} = 4V_{guide}/\langle v \rangle A_{hole}, \quad (5)$$

214 where V_{guide} is the volume of the guide, A_{hole} is the surface area of the hole,
 215 and $\langle v \rangle$ is the mean UCN velocity.

216 Now including the case in which there are other loss mechanisms, the bottle

²¹⁷ lifetime is determined from

$$\tau_d^{-1} = \tau_\beta^{-1} + \tau_{bottle}^{-1} + \tau_{gap}^{-1} + \tau_{hole}^{-1}, \quad (6)$$

²¹⁸ where τ_d is the measured drain time of UCN from the bottle after closing the
²¹⁹ upstream gate-valve. In these measurements the end plates were coated with
²²⁰ the same NiP as the guide and are considered part of the bottle surface area.

²²¹ *4.1.1. Extraction of the loss probability from double pinhole measurements*

²²² For simplicity, first consider a case where the UCN have a single velocity.
²²³ Then the UCN loss rate $R_L(t)$ at time t is given by

$$R_L(t) = \frac{A_{tot}}{4V} \bar{\mu} N(t)v = \frac{dN(t)}{dt}, \quad (7)$$

²²⁴ where A_{tot} is total inner surface area of the bottle, V is the volume of the bottle,
²²⁵ and $N(t)$ is the number of the stored UCN at time t . The loss per bounce
²²⁶ probability $\bar{\mu}$ can in general depend on v and can also contain contributions
²²⁷ from multiple loss mechanisms (including the pinhole contribution).

²²⁸ The rate $R_D(t)$ at which UCN are detected through a pinhole at time t is
²²⁹ given by $R_D(t) = \frac{A_{hole}v}{4V} N(t)$, where A_{hole} is the area of the pinhole. $dN(t)/dt$
²³⁰ can also be given by

$$\frac{dN(t)}{dt} = \frac{dR_D(t)}{dt} \frac{4V}{A_{hole}v}. \quad (8)$$

²³¹ Then the loss per bounce, the quantity of interest, is given by

$$\bar{\mu} = \frac{1}{R_D(t)} \frac{dR_D(t)}{dt} \frac{4V}{vA_{tot}}. \quad (9)$$

²³² Thus, if v is known, the detection rate of UCN through the pinhole (and its
²³³ time dependence) gives the loss probability per bounce.

²³⁴ In general, the UCN stored in a bottle have a velocity distribution. There-
²³⁵ fore, knowing the velocity distribution of the UCN inside the bottle is essential
²³⁶ in extracting the loss per bounce information. The UCN velocity distribution
²³⁷ itself can evolve while the UCN are stored. In the remainder of this section, we
²³⁸ discuss how we obtain the input velocity distribution and our evaluation of the
²³⁹ effect of the velocity distribution evolution.

240 4.1.2. Velocity distribution from UCN transport Monte Carlo

241 A UCN transport Monte Carlo simulation was developed at LANL to assess
 242 improvements to the guide system for the source upgrade. Material interactions
 243 in the simulation are parameterized by μ_{mc} , an energy independent loss-per-
 244 bounce probability, f_{mc} , a loss factor which is related to the loss-per-bounce
 245 probability by Eq. 2, and the probability of having a diffuse reflection, η_{mc} .
 246 Diffuse scattering occurs in the simulation when a random number sampled
 247 uniformly between 0–1 is less than η_{mc} . The direction of a UCN exiting a diffuse
 248 scatter is sampled from a Lambertian distribution [36]. These parameters can be
 249 set independently for each surface in the simulation. Parameters for the guide
 250 system coupling the UCN source to the experimental hall were determined by
 251 reconstructing timing distributions measured by A. Saunders *et al* [18]: $\mu_{mc} =$
 252 6.0×10^{-4} , $f_{mc} = 0$, and $\eta_{mc} = 5.2\%$. The energy dependent and independent
 253 loss probabilities can be used to describe guide sections that are dominated by
 254 gaps ($\mu_{mc} \neq 0, f_{mc} = 0$), sections where the material loss and gaps have similar
 255 weight ($\mu_{mc} \neq 0, f_{mc} \neq 0$), or gapless sections ($\mu_{mc} = 0, f_{mc} \neq 0$).

256 This simulation was used to determine the velocity distribution of UCN
 257 entering the test guide for the measurement done at LANL discussed in Sec.
 258 5. UCN with initial kinetic energy up to 400 neV are randomly sampled from
 259 a $N(v) \propto v^2$ distribution in the solid deuterium volume of the source and
 260 propagated through the guide system to the test guide. The velocity distribution
 261 $P(v)$ of UCN entering the test guide is recorded and fit to a piece-wise function
 262 with velocity v above and below $v_c = 5.7$ m/s

$$P(v) = \begin{cases} a(v - b)^c, & v \leq v_c, \\ de^{-vf}, & v > v_c, \end{cases} \quad (10)$$

263 shown in Fig. 4. The results of the simulation show that the initial v^2 distribu-
 264 tion is conditioned during transport and that velocity distribution entering the
 265 test apparatus is $\propto v^{2.7}$ with a mean velocity of $\bar{v} = 4.8(1)$ m/s. These results
 266 are consistent with previous analysis of UCN velocity spectrum [37]

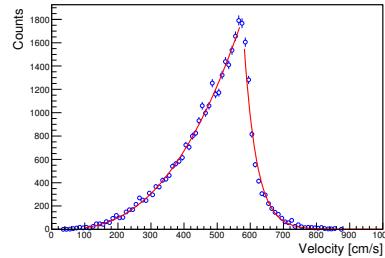


Figure 4: The simulated velocity distribution recorded at the entrance to the test guide for a guide system with an energy independent loss-per-bounce probability of 6×10^{-4} and nonspecularity of 5.2%. Simulated data are shown as open circles and the curves show the result of a fit to Eq. 10 (color online).

267 *4.1.3. Evolution of \bar{v} versus filling time and hole diameter*

268 Given that the rate at which UCN enter the bottle through a hole is proportional
 269 to the velocity, filling the bottle for a finite time will preferentially load
 270 faster UCN, in effect heating the spectrum. This effect is somewhat canceled
 271 by the fact that internal loss rate are also proportional to velocity. A series of
 272 transport simulations were performed where the filling time and aperture size
 273 were varied to investigate the amount of spectral heating.

274 Using the simulation parameters and input velocity spectrum from Section 4.1.2 UCN were simulated filling the bottle for $t_{fill} \in (50, 100, 200, 300, 400)$ s
 275 through apertures of $r \in (0.152, 0.318, 0.444, 0.635)$ cm. The bottle surfaces had
 276 a loss-factor of $f = 1.2 \times 10^{-4}$, Fermi potential of 212 neV, and nonspecularity
 277 of $\eta = 3\%$. A cut plane at the exit pinhole was used to record the velocity of
 278 UCN that would be detected in the experiment. UCN tallied at the exit plane
 279 are weighted by their probability to surviving until the time of detection due
 280 to β -decay. The results of these simulations are shown in Fig. 5. As expected,
 281 shorter fill time results in a hotter UCN spectra. Also, reducing the radius of
 282 the fill hole was observed to increase the mean velocity for a give fill time.

284 The measurements presented in the following section use apertures of $r =$
 285 0.152 cm and $r = 0.318$ cm and a filling time of 300 s. These simulation show
 286 that the difference in the internal mean velocity is on the order of a few cm/s or 1-

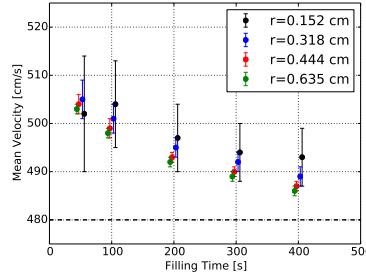


Figure 5: The mean velocity for UCN in the test bottle as determined by UCN transport Monte Carlo as a function of filling time for hole diameters of 0.304 cm, 0.636 cm, 0.888 cm, and 1.27 cm. Error bars are statistical (color online). Filling times of 50 s, 100 s, 200 s, 300 s, and 400 s were simulated and the results from the different hole diameters are shown with an offset around these times for clarity.

287 2% between measurements made with different aperture sizes. From simulation,
 288 the mean velocity of the internal spectrum was found to be 12(3) cm/s faster
 289 than the mean velocity of the input spectrum.

290 *4.1.4. Time evolution of \bar{v} in a closed bottle*

291 As noted by Tang, *et al*[26] the velocity distribution inside the bottle will
 292 evolve with time, cooling the spectrum as the faster UCN are lost through the
 293 exit hole or in interactions with the walls. The time dependence of the mean
 294 velocity can be calculated as

$$\bar{v}(t) = \frac{\int_0^{v_c} v \mathcal{P}(v) e^{-t/\tau(v)} dv}{\int_0^{v_c} \mathcal{P}(v) e^{-t/\tau(v)} dv}, \quad (11)$$

295 where v_c is the cutoff velocity, defined to be twice the maximum trappable
 296 velocity of the bottle. For this analysis the integration was cut off at $v_c \approx$
 297 12 m/s. In most cases relevant to UCN, $\mathcal{P}(v)$ is assumed to be the low energy
 298 tail of the Maxwell-Boltzmann distribution. $\mathcal{P}(v)$ is proportional to v^2 , which
 299 leads to an initial mean velocity of $\approx (3/4)v_{max}$. After substituting v^2 for the
 300 velocity distribution, Eq. 11 can be reduced to

$$\bar{v}(t) = \frac{e^{-btv_c} [-(btv_c)^3 - 3(bt v_c)^2 - 6btv_c - 6] + 6}{bt [e^{-btv_c} \{-(btv_c)^2 - 2btv_c - 2\} + 2]}, \quad (12)$$

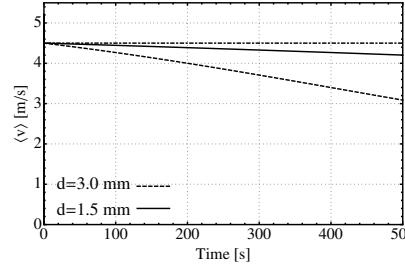


Figure 6: *Left* : The time dependence of the mean velocity from Eq. 12 for $\mathcal{P}(v) \propto v^2$ and a maximum velocity of 6.0 m/s for a 7.5 cm diameter, 100 cm long bottle with drain holes of radius 0.15 cm (solid) and 0.3 cm (dashed). The dot dashed line represents the mean velocity at $t=0$. Only losses due to the drain holes are included.

301 where $1/\tau(v) = bv$, $b = A/4V$ in the case where the hole represents the only loss
 302 mechanism in the bottle (τ given by Fig. 5). Figure 6 shows $\bar{v}(t)$ for $t \in (0, 500)$
 303 for a perfect bottle where the drain hole is the only loss. This analysis shows that
 304 treating the drain curve from a pinhole experiment with a static \bar{v} is insufficient
 305 to capture the dynamics in the bottle. Since the velocity distribution is not of
 306 the form $\propto v^2$, Eq. 11 will be numerically integrated in the following analysis.

307 *4.1.5. Relationship between detected rate and loss rate*

308 A possible ambiguity of the double pinhole approach is whether the measured
 309 rate of UCN exiting the bottle has a direct relationship with the loss mechanisms
 310 internal to the bottle. This was assessed using a transport Monte Carlo where
 311 the gap losses are represented as an energy independent loss-per-bounce prob-
 312 ability and an energy dependent loss for the NiP surface scaled by a loss-factor.
 313 The times UCN are lost on the surface of the bottle and the times UCN are
 314 detected are tallied. Fig. 7 shows that there is a direct correlation between the
 315 simulated rate of UCN capturing on a detector downstream of the pinhole and
 316 the change of the internal density of UCN. Therefore this method is sufficient
 317 for characterizing losses on the material walls of the guide.

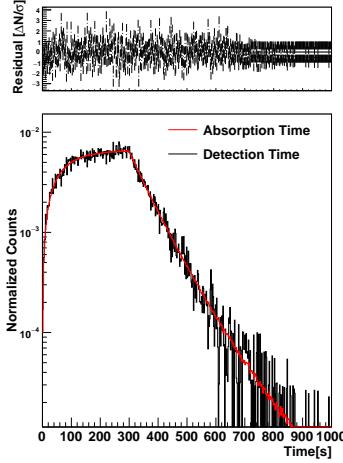


Figure 7: The normalized loss rate of UCN on the internal surfaces of the bottle (red line) is plotted against the normalized detection rate of UCN exiting the bottle (black line). Loss mechanisms accounted for in the simulation include: β -decay, an energy-independent loss-probability to account for gaps, and an energy-dependent loss-factor to represent interactions with NiP coated surfaces (color online).

318 **5. Measurements at LANL**

319 *5.1. Experimental Setup*

320 UCN are produced by cooling spallation neutrons generated by the pulsed
 321 800 MeV proton beam incident on a tungsten target at the LANSCE UCN
 322 source [18]. A cold moderator made of polyethylene cools a fraction of the
 323 spallation neutrons, and these cold neutrons, $E_k \approx 5$ meV, are down-scattered
 324 to UCN energies in a solid deuterium (SD_2) crystal at ~ 5 K. UCN produced in
 325 the SD_2 are confined by the Fermi potential of the ^{58}Ni coated source vacuum
 326 vessel. A series of stainless steel tubes couple to the source volume 1 m above
 327 the SD_2 volume and guide the UCN through the concrete biological shielding
 328 to the experimental area. This transit conditions the energy spectrum of UCN
 329 to the stainless steel Fermi potential, 188 neV.

330 The guide test apparatus was coupled to the outlet of the UCN source

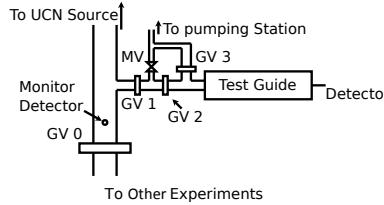


Figure 8: Transport and pumping manifold for the guide test apparatus. Detailed description in the text.

331 through a 2.54 cm aperture in the primary UCN guide as shown in Fig. 8.
 332 In this position the apparatus was mounted upstream of the superconducting
 333 magnet which pulls neutrons of one spin state through the vacuum separation
 334 foil such that the full output density of the source was accessible. Experiments
 335 mounted in this position run parasitically to the main UCN transport line and
 336 some system conditions were controlled by a higher priority experiment down-
 337 stream. UCN were loaded into the bottle through the pumping manifold, shown
 338 in Fig. 8. Valve GV1 separates the holding volume from the source. Valve GV2
 339 has a thin layer of TPX (polymethylpentene) on the downstream face which,
 340 when closed, will absorb and upscatter UCN exiting the test guide through the
 341 upstream pinhole. Valve GV3 allows the test guide to be continually pumped
 342 during the counting period. Valves GV2 and GV3 are controlled by an auto-
 343 mated valve control program and are always in opposite states.

344 Pinhole plates made from 304 stainless steel (Fig. 2) were mounted to both
 345 ends of the test guide to create a bottle. The interior face of the pinhole plate
 346 is flat and the exterior face has a 45° chamfer around the pinhole, which was
 347 positioned 2.54 cm off center. Two sets of pinhole plates were made, one with
 348 a 0.635 cm diameter hole and the other with a diameter of 0.318 cm. The up-
 349 and stream pinhole plates are mounted to the guide with a 180° rotation
 350 about the guide axis relative to each other, so that the pinhole was at 6 o'clock
 351 on the inlet side and 12 o'clock on the outlet. All pinhole plates were machined
 352 to a surface finish of 16 micro-inch or smoother and coated with NiP using the

353 same process as the test guides.

354 UCN leaving the pinhole on the downstream side of the guide transit through
 355 a 2.54 cm diameter outlet port to a ^{10}B coated ZnS:Ag foil detector [38]. UCN
 356 are captured in a 100 nm layer of ^{10}B coated on the ZnS:Ag substrate. The
 357 neutron capture on the ^{10}B produces an α and a ^7Li ion and one or both enter
 358 the ZnS:Ag substrate creating scintillation light which is measured by a 2.5 cm
 359 diameter PMT mounted outside of a vacuum window. Since the ^{10}B foil was
 360 inside the vacuum system the detection efficiency is relatively independent of
 361 UCN energy, unlike the ^3He gas detectors which require a 1 m drop to give UCN
 362 sufficient energy to penetrate an entrance window. The UCN density outside
 363 the test volume was also monitored with a similar ^{10}B coated ZnS:Ag detector,
 364 which was coupled to the main UCN guide through a 0.159 cm diameter hole.

365 *5.2. Data Collection*

366 The nominal data-taking cycle consisted of filling the bottle for 300 s and
 367 then draining for 300 s. These times correspond to several lifetimes for a material
 368 bottle with a nominal loss probability of $\mathcal{O}(10^{-4})$ containing UCNs with a mean
 369 velocity of ≈ 5 m/s and mean free path of ≈ 7.3 cm. During the fill GV0 was
 370 closed (Fig. 8) after 150 s to accommodate the experiment that was on the main
 371 port, which led to an increase in the UCN density outside the test port. This
 372 resulted in an increase in the measured rate in period (150-300 s) relative to the
 373 (0-150s) period, shown in Fig. 9 along with various fits discussed in the sections
 374 below. The drain time varied between 270 s and ~ 1000 s. Measurements with
 375 less than a 600 s fill and drain cycle were analyzed separately and averaged with
 376 the longer drain time measurements.

377 Signals from the PMT monitoring the ^{10}B ZnS:Ag foil were amplified with
 378 an Ortec 579 Fast Amp and analyzed by an Ortec 551 single channel analyzer.
 379 Timing pulses from the SCA were recorded by a Fast ComTec multichannel
 380 scaler. The measured background rate was typically 0.1 s^{-1} and was deter-
 381 mined from a fit for each data set. The peak signal rate when the UCN reach
 382 equilibrium density in guide was $30\text{-}40 \text{ s}^{-1}$ for the 0.635 cm diameter hole and

383 4-8 s⁻¹ for the 0.318 cm hole.

384 *5.3. Analysis*

385 The measured rate in the drain detector is fit to a double exponential plus
386 background term of the form

$$N(t) = N_s e^{-(t-\Delta t)/\tau_s} + N_l e^{-(t-\Delta t)/\tau_l} + B, \quad (13)$$

387 where $N_{s,l}$ are the weights of the short and long lifetime (τ_s and τ_l) components
388 of the UCN population in the bottle, $\Delta t = 300$ s is the start of the draining
389 period, and B is the background rate. The short lifetime term characterizes the
390 UCN which possess kinetic energy above the bottle potential, typically called
391 super-barrier UCN. Since it is the perpendicular component of the kinetic energy
392 that determines whether super-barrier UCN will penetrate the surface, they can
393 be trapped so long as they continue to scatter at grazing incidence. Diffuse
394 scattering on the bottle surface can lead to these UCN having closer to normal
395 incidence collisions with the wall and being lost. The long lifetime term is
396 therefore considered to represent the lifetime of trappable UCN in the bottle and
397 is the related to the loss-per-bounce probability of the material. The short and
398 long lifetimes determined by fitting the data with Eq. 13 are listed in Table 2.

399 The results of attempting to describe the unloading curve with a single time
400 constant are shown in Fig. 9. As expected, the single constant does not represent
401 the data, under predicting the rate at later times.

402 *5.3.1. Analysis 1: Double exponential fitting*

403 The bottle lifetime τ_{bottle} is related to the measured drain time, τ_d by

$$\tau_d^{-1} = \tau_{bottle}^{-1} + \tau_\beta^{-1} + \tau_{hole}^{-1} + \tau_{gaps}^{-1}, \quad (14)$$

404 where τ_β is the neutron lifetime, τ_{hole} is the drain time of the pinholes, and
405 τ_{gaps}^{-1} is the loss rate of UCN in gaps between the end of the test bottle and
406 pinhole plates or guide to guide mating. The drain time of UCN out of a hole
407 is $\tau_{hole} = 4V/(\langle v \rangle A_h)$, where V is the volume of the bottle, A_h is the area of

Table 2: The short and long lifetimes from a double exponential fit to the draining curve are listed for each guide and pinhole combination, Analysis 1. The listed loss-per-bounce probability does not correct for the unmeasured effect of the gaps in the system and therefore is the average system loss-per-bounce for the guide and oriface plates. Uncertainties for τ_s , τ_l , f , and l are the 1σ fit uncertainties. The resulting loss-factor and gap size from Analysis 2 are shown for individual fits (f, l) and the combined fit (f_c, l). Gap lengths that include 0 μm at 1σ are reported as 2σ upper bounds.

Guide	Pinhole [cm]	Analysis 1			Analysis 2			
		τ_s [s]	τ_l [s]	$\bar{\mu}$ [10^{-4}]	f [10^{-4}]	l [μm]	f_c [10^{-4}]	l [μm]
1	0.635	19.4(2.0)	36.2(0.6)	2.1(4)	0.9(1.0)	≤ 132	1.1(7)	67(63)
1	0.318	22.3(3.2)	55.8(1.4)	2.0(3)	1.0(1.1)	≤ 210		≤ 99
2	0.635	26.2(3.9)	48.3(2.1)	1.0(3)	1.1(3)	70(30)	1.5(3)	101(28)
2	0.318	26.4(5.5)	73.0(7.6)	1.4(3)	2.7(2)	≤ 27		31(24)
3	0.635	26.3(1.9)	67.8(2.5)	1.1(2)	1.8(2)	≤ 62	1.5(1)	45(9)
3	0.318	29.8(4.5)	85.8(8.7)	1.4(2)	1.1(2)	≤ 120		≤ 4
1+2	0.635	33.3(2.0)	58.8(1.6)	1.5(2)	2.3(2)	≤ 120	1.2(1)	18(7)
1+2	0.318	27.4(11.2)	74.7(4.0)	1.6(2)	1.9(4)	≤ 60		≤ 9

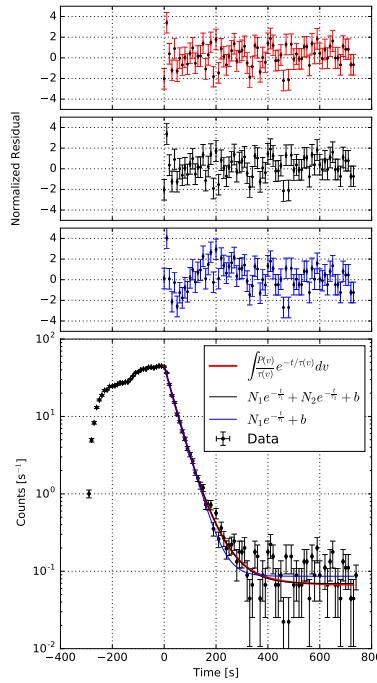


Figure 9: Fill and drain curve from guide 2 with the 0.635 cm diameter pinhole plates. The increase in count rate between the 0-150 s and 150-300 s period is due to the main gate valve downstream of the experimental setup being closed. Single and double exponential fits to the data are shown as solid blue and black lines respectively. The red line is the result of numerical integration analysis using Eq. 18 to fit the data. The upper panels show the normalized residuals of the data to each fit. (color online)

408 the hole, and $\langle v \rangle$ is the average velocity of the UCN in the bottle. The rate of
 409 loss due to gaps between the guide end and pinhole plate is estimated from

$$\tau_{gap}^{-1} = \epsilon \frac{\langle v \rangle A_{gap}}{4V} = \epsilon \frac{\langle v \rangle \langle d \rangle}{rl}, \quad (15)$$

410 where ϵ is the probability that UCN that find the gaps to be lost, $\langle d \rangle$ is the
 411 average gap size, r is the radius of the guide, and l is the length of the guide. If
 412 a gap is sufficiently diffuse the loss probability should approach 50%, however

413 for this analysis a conservative approach is taken and ϵ is set to 1 and $\langle d \rangle$ is the
 414 effective gap size.

415 The bottle lifetime can be written as

$$\tau_{bottle}^{-1} = \frac{\langle v \rangle A_T \bar{\mu}}{4V}, \quad (16)$$

416 where A_T is the surface area of the bottle and $\bar{\mu}$ is the loss probability averaged
 417 over the ensemble of UCN. Eq. 14 can be rewritten as

$$\tau_d^{-1} = \tau_\beta^{-1} + \frac{\langle v \rangle}{4V} (A_T \bar{\mu} + A_h). \quad (17)$$

418 Here $\bar{\mu}$ is assumed to be an averaged system loss rate incorporating loss on the
 419 bottle walls as well as loss due to gaps between the tube and the end plates.
 420 The Monte Carlo generated velocity distribution from Section 4.1.2 is used to
 421 determine the mean velocity $\langle v \rangle = 4.8$ m/s used in this analysis. The loss
 422 probability for the bottle system $\bar{\mu}$ can be determined using the measured τ_l
 423 and are listed in Table 2. The error-weighted mean loss-probability from the
 424 measurements performed on the four guides is $\bar{\mu} = 1.3(1) \times 10^{-4}$.

425 If the difference in τ_l measured in guide 3 and the guide 1+2 system is
 426 attributed to the additional two gaps created by the guide coupling plate the
 427 size of the internal gaps can be estimated. The total additional gap is $< 76\mu\text{m}$,
 428 assuming 100% loss in the gap.

429 *5.3.2. Analysis 2: Numerical integration fitting*

430 An alternative analysis method is to fit the data to

$$\frac{dN(t)}{dt} = N_0 \int_0^{v_{cut}} \frac{P(v)}{\tau(v)} e^{-t/\tau(v)} dv + B, \quad (18)$$

431 which incorporates the full velocity dependence of the Eq. 14 and the input ve-
 432 locity distribution $P(v)$. Instead of fitting the data to multiple time constants
 433 the fit parameters for Eq. 18 are the overall normalization N_0 , the loss-factor
 434 f , the total gap length l , and the background rate B . Because of the com-
 435 plicated dependence on energy in $\mu(E)$ Eq. 18 is numerically integrated up to
 436 $v_{cut} = 12$ m/s and evaluated at each time step.

437 Two approaches were used in this analysis. The results of each approach is
 438 summarized in Table 2. In the first approach Eq. 18 was fit to the data with
 439 N_0 , f , l , and B as free parameters, where all parameters were constrained to be
 440 positive. In this approach the loss-factor and gap size were found to be strongly
 441 correlated, but in some cases the fit algorithm failed to find a non-zero gap size.
 442 From averaging the results from the five data-sets with non-zero gap sizes, the
 443 mean loss-factor and gap length are $f = 1.4(1) \times 10^{-4}$ and $l = 50(22) \mu\text{m}$.
 444 In the second approach, the drain curves obtained for the two radii pinholes
 445 were simultaneously fit for each guide, constraining the loss-factor f_c to be a
 446 shared parameter and N_0 , l , and B as free parameters. This approach yielded
 447 an average loss-factor for the NiP coating of $\langle f_c \rangle = 1.36(7) \times 10^{-4}$.

448 **6. Measurements at the ILL**

449 *6.1. Experimental Setup and Data Collection*

450 The guide test apparatus was mounted on the TEST port of the UCN turbine
 451 PF2 at the ILL Grenoble, France [39]. Measurements performed at PF2 include
 452 the characterization of the UCN velocity spectrum, the relative transmission
 453 of electropolished and nonelectropolished NiP coated guides, and the double
 454 pinhole bottling lifetimes. All measurements were performed downstream of
 455 a $100 \mu\text{m}$ Al foil which separates the turbine vacuum from the experimental
 456 volume, blocking neutrons with kinetic energy below the aluminum Fermi po-
 457 tential $V_F^{Al} = 54 \text{ neV}$.

458 Ultracold neutrons were detected with the same ^{10}B ZnS:Ag foil detectors
 459 used in the LANL measurements. Signals from the photo-multiplier tube mon-
 460 itoring the foil were amplified with an Ortec 474 timing filter amp and recorded
 461 with CAEN DT5724 100 MHz, 14-bit analog to digital converter digitizer.

462 *6.1.1. Velocity Spectrum Measurements*

463 The velocity spectrum of UCN exiting the test port was measured using a
 464 rotating disc velocity selector [40]. The velocity selector has seven titanium discs

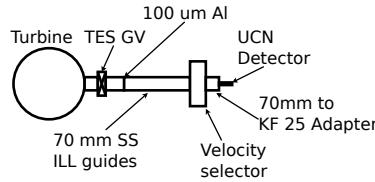


Figure 10: The velocity selector was connected to the turbine by a 1 m long 70 mm OD stainless guide after the coupler housing the $100 \mu\text{m}$ thick Al safety foil. On the downstream side a 22 cm long 70 mm OD to KF-25 adapter was used to attach the UCN detector.

465 with 26 evenly spaced 19° slits. The slits cover 50% of the surface area of each
 466 disc. The discs are positioned such that, when spinning at frequency f , UCN
 467 with velocity $v = f/5$ are transmitted. A diagram of how the velocity selector
 468 was attached to the beam line is shown in Fig. 10. The velocity spectrum was
 469 measured between 1-11.5 m/s in 0.5 m/s steps and the results are shown in
 470 Fig. 11. The results of this measurement confirm that UCN with velocities
 471 below ≈ 4.5 m/s are not transmitted through the aluminum safety foil. NiP
 472 coated guides having a Fermi potential of $V_F = 213$ neV can trap UCN with
 473 $v_{max} < 6.3$ m/s and therefore the majority of the neutrons at this position on
 474 the turbine are non-trappable. The measured velocity distribution was used as
 475 an input the Monte Carlo simulations of the test geometry.

476 *6.1.2. Relative Transmission Measurements*

477 The relative transmission through an electropolished and non-electropolished
 478 140 cm length of guide was measured for UCN with $v = 5.5$ m/s. The test
 479 configuration is shown in Fig. 12. Upstream of the velocity selector the incident
 480 flux of UCN is monitored through a 64 mm diameter hole in the wall the guide
 481 coupler. A 70 cm length of guide (guide 1) was mounted downstream of the
 482 velocity selector followed by a 140 cm length of guide. Both 140 cm guides
 483 were coated with $50 \mu\text{m}$ of NiP, however guide 4 was electropolished prior to

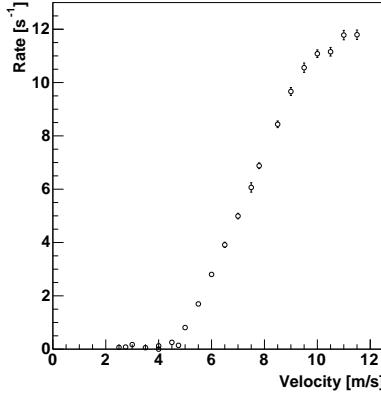


Figure 11: Velocity spectrum measured at the ILL TEST port using the TUM velocity selector.

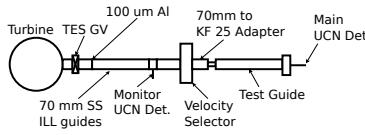


Figure 12: Guide configuration for the measurement of transmission through NiP coated guides.

a

484 coating⁶. The rate at the end of the guide was measured using a ¹⁰B ZnS:Ag
 485 detector mount on a 70 mm to KF25 adapter. A thin sheet of TPX with a 2.54
 486 cm diameter hole was fixed to the interior of the adapter to prevent bottling,
 487 preventing multiple bounces which distort the result.

488 The rate measured in the main UCN detector was normalized to the rate in
 489 the upstream monitor and the background was measured by closing the turbine
 490 shutter. The background-subtracted normalized rates were $2.4(3) \times 10^{-3}$ and

⁶All stainless steel guide stock arrived electropolished from the factory. The guide stock is then cut to length and flanges are welded to the ends. In this case guide 4 was electropolished after machining and welding whereas the other guides were not.

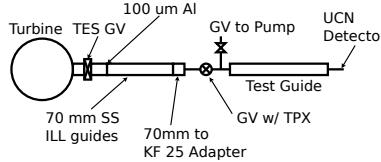


Figure 13: Bottling measurements were performed using a gate valve manifold system that couples to the feed guide and the pinhole plate with KF-25 connections. The gate valve "GV /w TPX" is opened to fill the test bottle and when closed introduces a TPX foil to the upstream side of the test guide, removing any neutrons escaping on that side. The gate valve "GV to Pump" allows continuous pumping of the test guide and its state is always opposite of the inline gate valve.

491 $1.4(3) \times 10^{-3}$ for the electropolished and non-electropolished guides respectively,
 492 corresponding to relative transmission of 55(14)%. The statistical uncertainty
 493 baseline measurement was too large to determine an absolute transmission, how-
 494 ever the relative measurement of guides 3 and 4 confirms the vendor's observa-
 495 tion that the NiP coating enhances inherent surface roughness, degrading the
 496 transmission. It has been noted that coating process used by other vendors does
 497 not amplify the roughness of the substrate [41].

498 *6.1.3. Bottle lifetime data collection*

499 The bottling lifetime was measured by filling the test guide through small
 500 aperture for a time $\Delta t = 200$ s and monitoring the count rate through a similar
 501 aperture on the downstream side of the test guide. The aperture plates were
 502 the same as those used Section 5. After filling, the gate valve was closed and
 503 the rate of UCN exiting the downstream aperture was monitored for 300 s. A
 504 time tag in the data, generated when the gate valve opens, allows the beginning
 505 of the filling period to be aligned in analysis. The 70 mm OD ILL guide from
 506 the turbine was connected to gate valve manifold system, via 70 mm to KF-25
 507 reducer, which allowed the flow of neutrons to be stopped and introduced a
 508 TPX foil on the upstream side of the test guide to remove UCN escaping from
 509 that side of the test guide. This prevented reloading of the test guide during

510 the draining period. Additionally, there was a second gate valve, not in the
 511 path of neutrons, which allowed continuous pumping on the bottle. Typically,
 512 the rate of UCN detected downstream of test guide was 10 s^{-1} when the bottle
 513 had reached its maximum density. Therefore, to achieve sufficient statistical
 514 precision each guide was measured for approximately eight hours or fifty fill and
 515 drain cycles.

516 *6.2. Analysis*

517 *6.2.1. Double Exponential Analysis*

518 The rate that UCN drain from the test bottle can be characterized to first
 519 order as

$$N(t) = \begin{cases} \sum_{i=l,s} (N_i(1 - e^{-t/\tau_i})) + B, & t < t_{fill} \\ \sum_{i=l,s} (N_i e^{-(t-t_{fill})/\tau_i}) + B, & t > t_{fill} \end{cases} \quad (19)$$

520 where $t_{fill} = 200 \text{ s}$, N_i is normalization parameter, τ_i is the time constant of the
 521 system, the indices l and s denote the long and short components of the time
 522 constant, $B = n_c R_{bkg}$ is the fixed background rate, where n_c is the number of
 523 fill and drain cycles, and the measured background rate $R_{bkg} = 0.006(1) \text{ s}^{-1}$
 524 for the 70 cm guides and $R_{bkg} = 0.0035(4) \text{ s}^{-1}$ for the 140 cm guides. In this
 525 analysis the filling and draining time constants were assumed to be the same,
 526 $\tau_F = \tau_D$. The short and long time constants are interpreted in the same way
 527 as in Section 5.3.1. The results of this analysis are summarized in Table 3 and
 528 the mean loss-probability is $\bar{\mu} = 1.6(2) \times 10^{-4}$.

Table 3: The results of fitting Eq. 19 to the fill and drain timing data and the loss per bounce probability are summarized below. Fit results which include 0 at 1σ are reported as 2σ upper bounds.

Guide	τ_s [s]	τ_l [s]	$\bar{\mu}[10^{-4}]$	f [10^{-4}]	l [μm]	\bar{f} [10^{-4}]
1	19.5(4.3)	45.3(20.1)	2.0(1.2)	≤ 2.0	178(23)	1.7(1)
2	26.4(8.3)	59.4(12.3)	1.4(0.4)	≤ 2.1	92.7(8.1)	1.0(1)
1+2	28.3(9.1)	57.5(18.2)	1.7(0.7)	≤ 2.3	≤ 460	1.2(1)
3	19.1(5.5)	62.2(8.9)	1.6(0.3)	1.2(1)	≤ 140	1.2(1)

529 *6.2.2. Numerical Integration Analysis*

530 The same analysis method used in Section 5.3.2 were applied to the ILL
 531 data using the measured velocity spectrum. Results for the fit parameters f
 532 and l for the two parameter fit and \bar{f} for the one parameter fit are summarized
 533 in Table 3. The entire data-set failed to converge on values for the loss-factor
 534 and gap length in the two parameter fit. To mitigate the instability of the two
 535 parameter fit, the gap length was fixed to zero and an effective loss-factor was
 536 used to fit the data. In this method the effect loss-factor is $\bar{f} = 1.3(1) \times 10^{-4}$.

537 **7. Discussion**

538 The results presented in Table 2 and Table 3 show that the system loss-
 539 probability for UCN interacting on stainless steel guides coated with $50 \mu\text{m}$

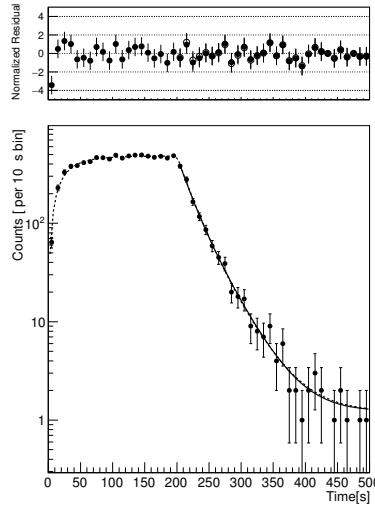


Figure 14: *Bottom:* Data for combination of guide 1 and 2 with the 1/4" pinhole plate is shown. The data is fit using Eq. 19 for the entire time range (dashed line) and over the draining time (solid line). *Top:* The fit residuals normalized by the statistical uncertainty of the bin are shown for a fit of Eq. 19 over the entire range (solid circles) and over the draining time (open circles). Error bars are 1σ .

540 of nickel phosphorus and coupled with the design described in Section 3 is
 541 $\bar{\mu} = 1.4(1) \times 10^{-4}$. This new guide coupling flange has shown to reliably and re-
 542 peatably join guides with minimal gaps, $\sim 100 \mu\text{m}$ improving, the bottle lifetime
 543 over previous designs using ConFlat flanges. Because the gaps can be reliably
 544 small this opens the possibility of using multiple sections of short guide to cover
 545 long distances which allows for the guides to be coated using methods that
 546 typically have length limitations (such as NiP and NiMo).

547 Analysis of ILL and LANL data-sets based on the method discussed in Sec-
 548 tion 5.3.1 produced consistent values for $\bar{\mu}$ after accounting for the different
 549 velocity distributions at each UCN source. Using a numerical approach to the
 550 analysis, this data resulted in a consistent loss-factor, $f_c = 1.3(1) \times 10^{-4}$ when a
 551 combined fit to the small and large pinhole drain curves was used. Nickel phos-
 552 phorus guide coating will provide a significant improvement in UCN density over
 553 stainless steel for experiments which fill a storage volume such as nEDM and
 554 neutron lifetime measurements. Results from the ILL indicate that the trans-
 555 mission through the electropolished nickel phosphorus coated guides are better
 556 than 85% at the 95% confidence level, but another measurement is required to
 557 determine the absolute transmission.

558 The pinhole bottling method of characterizing surfaces provides a comple-
 559 mentary approach to the gate valve bottling method with advantages and dis-
 560 advantages. In both methods knowledge of the initial velocity spectrum of UCN
 561 in the bottle and how that spectrum evolves with time is required to extract
 562 relevant surface parameters. One advantage of this method is that it requires
 563 no moving parts removing any uncertainties that arise due to mechanical repro-
 564ducibility. However, loading and draining the bottle through the static pinhole
 565 increases the time required to make a measurement and as guides improve (loss-
 566 factor decreases) the fill and drain cycle will take longer further increasing the
 567 time required.

568 Combining this work with the results of our earlier work [26] indicates that
 569 nickel phosphorus is a commercially available, low-depolarizing, low-loss coating
 570 for ultracold neutron transport guides. These qualities make NiP coatings ideal

571 for large UCN transport systems and have already been implemented in the
 572 upgrade of the LANL source. The electroless coating process allow parts with
 573 complex geometries to be uniformly coated.

574 **8. Conclusions**

575 We have measured the storage time of UCN in a nickel phosphorus coated
 576 bottle at two facilities with different neutron velocity distributions. The res-
 577 ults of these measurements are used to determine an effective loss-per-bounce
 578 probability in the bottle of $\bar{\mu} < 1.6 \times 10^{-4}$, which fulfills the transport require-
 579 ments for upgrade of the LANL UCN source. The results of this measurement
 580 demonstrate that the pinhole method is a reliable technique to characterize UCN
 581 surface properties. The Fermi potential of the NiP coating, $V_F = 213(5)$ neV,
 582 was measured at the ASTERIX time-of-flight spectrometer. Transmission meas-
 583 urements determined that although the guide is electropolished in production
 584 a second electro-polishing after welding the flanges on is required to maintain a
 585 specular surface.

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592 [1] D. J. Salvat, E. R. Adamek, D. Barlow, J. D. Bowman, L. J. Broussard,
 593 N. B. Callahan, S. M. Clayton, C. Cude-Woods, S. Currie, E. B. Dees,
 594 et al., Phys. Rev. C **89**, 052501 (2014).

595 [2] A. Serebrov, V. Varlamov, A. Kharitonov, A. Fomin, Y. Pokotilovski,
 596 P. Geltenbort, J. Butterworth, I. Krasnoschekova, M. Lasakov, R. Tal'daev,
 597 et al., Phys. Lett. B **605**, 72 (2005).

598 [3] A. Pichlmaier, V. Varlamov, K. Schreckenbach, and P. Geltenbort, Phys.
 599 Lett. B **693**, 221 (2010).

600 [4] S. Arzumanov, L. Bondarenko, S. Chernyavsky, P. Geltenbort, V. Morozov,
 601 V. Nesvizhevsky, Y. Panin, and A. Strepetov, Phys. Lett. B **745**, 79 (2015).

602 [5] S. Materne, R. Picker, I. Altarev, H. Angerer, B. Franke, E. Gutzmiedl,
 603 F. J. Hartmann, A. R. Müller, S. Paul, and R. Stoepler, Nucl. Instruments
 604 Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.
 605 **611**, 176 (2009).

606 [6] K. K. H. Leung, P. Geltenbort, S. Ivanov, F. Rosenau, and O. Zimmer,
 607 Phys. Rev. C - Nucl. Phys. **94**, 1 (2016), 1606.00929.

608 [7] C. Baker, Y. Chibane, M. Chouder, P. Geltenbort, K. Green, P. Harris,
 609 B. Heckel, P. Iaydjiev, S. Ivanov, I. Kilvington, et al., Nucl. Instruments
 610 Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.
 611 **736**, 184 (2014).

612 [8] J. M. Pendlebury, S. Afach, N. J. Ayres, C. A. Baker, G. Ban, G. Bison,
 613 K. Bodek, M. Burghoff, P. Geltenbort, K. Green, et al., Phys. Rev. D **92**,
 614 092003 (2015), 1509.04411.

615 [9] T. Ito, *Los Alamos National Laboratory LDRD Project #20140015DR*
 616 *Probing New Sources of Time-Reversal Violation with Neutron EDM*
 617 (2013).

618 [10] S. K. Lamoreaux and R. Golub, J. Phys. G Nucl. Part. Phys. **36**, 104002
 619 (2009).

620 [11] M. P. Mendenhall, R. W. Pattie, Y. Bagdasarova, D. B. Berguno, L. J.
 621 Broussard, R. Carr, S. Currie, X. Ding, B. W. Filippone, A. García, et al.,
 622 Phys. Rev. C **87**, 032501 (2013).

623 [12] L. Broussard, B. Zeck, E. Adamek, S. Baeßler, N. Birge, M. Blatnik,
 624 J. Bowman, A. Brandt, M. Brown, J. Burkhardt, et al., Nucl. Instruments

625 Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.
 626 (2016).

627 [13] V. V. Nesvizhevsky, H. G. Börner, a. M. Gagarski, a. K. Petoukhov, G. a.
 628 Petrov, H. Abele, S. Baeßler, G. Divkovic, F. J. Rueß, T. Stöferle, et al.,
 629 Phys. Rev. D **68**, 297 (2003), 0308108.

630 [14] T. Jenke, P. Geltenbort, H. Lemmel, and H. Abele, Nat. Phys. **7**, 468
 631 (2011).

632 [15] J. W. Martin, AIP Conf. Proc. **1560**, 134 (2013).

633 [16] A. Anghel, F. Atchison, B. Blau, B. van den Brandt, M. Daum, R. Doelling,
 634 M. Dubs, P. A. Duperrex, A. Fuchs, D. George, et al., Nucl. Instruments
 635 Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.
 636 **611**, 272 (2009).

637 [17] J. Karch, Y. Sobolev, M. Beck, K. Eberhardt, G. Hampel, W. Heil,
 638 R. Kieser, T. Reich, N. Trautmann, and M. Ziegner, Eur. Phys. J. A **50**,
 639 78 (2014).

640 [18] A. Saunders, M. Makela, Y. Bagdasarova, H. O. Back, J. Boissevain, L. J.
 641 Broussard, T. J. Bowles, R. Carr, S. A. Currie, B. Filippone, et al., Rev.
 642 Sci. Instrum. **84**, 013304 (2013).

643 [19] R. Golub, D. Richardson, and S. Lamoreaux, *Ultra-cold Neutrons* (Adam
 644 Hilger, New York, NY, 1991), ISBN 0-7503-0115-5.

645 [20] R. Golub and J. Pendlebury, Rep. Prog. Phys. **42** (1979).

646 [21] M. Daum, B. Franke, P. Geltenbort, E. Gutsmiedl, S. Ivanov, J. Karch,
 647 M. Kasprzak, K. Kirch, a. Kraft, T. Lauer, et al., Nucl. Instruments Meth-
 648 ods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip. **741**,
 649 71 (2014).

650 [22] E. Humbert and A. J. Tosser, J. Mater. Sci. Lett. **17**, 167 (1997).

651 [23] A. Berrada, M. F. Lapierre, B. Loegel, P. Panissod, and C. Robert, *J. Phys.*
652 *F Met. Phys.* **8**, 845 (1978).

653 [24] P. A. Albert, *J. Appl. Phys.* **38**, 1258 (1967).

654 [25] Y. Masuda, T. Kitagaki, K. Hatanaka, M. Higuchi, S. Ishimoto, Y. Kiy-
655 anagi, K. Morimoto, S. Muto, and M. Yoshimura, *Phys. Rev. Lett.* **89**,
656 284801 (2002).

657 [26] Z. Tang, E. Adamek, A. Brandt, N. Callahan, S. Clayton, S. Currie, T. Ito,
658 M. Makela, Y. Masuda, C. Morris, et al., *Nucl. Instruments Methods Phys.*
659 *Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.* **827**, 32 (2016),
660 1510.06490.

661 [27] www.ill.eu/pf2/.

662 [28] www.chemprocessing.com.

663 [29] A. Nelson, *J. Appl. Crystallogr.* **39**, 273 (2006).

664 [30] V. F. Sears, *Neutron News* **3**, 26 (1992).

665 [31] J. Sudagar, J. Lian, and W. Sha, *J. Alloys Compd.* **571**, 183 (2013).

666 [32] www.valex.com.

667 [33] T. Brenner, P. Fierlinger, P. Geltenbort, E. Gutsmiedl, A. Hollering,
668 T. Lauer, G. Petzoldt, D. Ruhstorfer, J. Schroffenegger, K. M. Seemann,
669 et al., *Appl. Phys. Lett.* **107**, 121604 (2015), 1502.06252.

670 [34] C. Patrignani, K. Agashe, G. Aielli, C. Amsle, M. Antonelli, D. Asner, and
671 H. Baer, *Chin. Phys. C* **40** (2016).

672 [35] M. Makela, *Priv. Commun.* (2014).

673 [36] S. Clayton, *Priv. Commun.* (2014).

674 [37] A. T. Holley, L. J. Broussard, J. L. Davis, K. Hickerson, T. M. Ito, C.-Y.
675 Liu, J. T. M. Lyles, M. Makela, R. R. Mammei, M. P. Mendenhall, et al.,
676 Rev. Sci. Instrum. **83**, 073505 (2012).

677 [38] Z. Wang, M. Hoffbauer, C. Morris, N. Callahan, E. Adamek, J. Bacon,
678 M. Blatnik, A. Brandt, L. Broussard, S. Clayton, et al., Nucl. Instruments
679 Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.
680 **798**, 30 (2015), 1503.03424.

681 [39] A. Steyerl, H. Nagel, F.-X. Schreiber, K.-A. Steinhauser, R. Gähler,
682 W. Gläser, P. Ageron, J. Astruc, W. Drexel, G. Gervais, et al., Phys.
683 Lett. A **116**, 347 (1986).

684 [40] J. Schroffenegger, Ph.D. thesis, Technische Universität Wien (2016).

685 [41] Y. Masuda, Priv. Commun. (2016).