

Study of Mass Transport Limited Corrosion with Rotating Cylinder Electrodes

An Overview of Theory and Practice

This technical note addresses two aspects of electrochemical testing using the Rotating Cylinder Electrode (RCE). First, the fundamental hydrodynamic behavior at a rotating cylinder is summarized, including equations which predict the mass transport limited corrosion current at an RCE. Second, a means of selecting the suitable rotation rate for an RCE test is discussed, with emphasis being placed on matching a particular rotation rate to a particular flow velocity in a smooth pipe. In addition, a bibliography of significant reports regarding the RCE is provided.

1. Introduction

Corrosion processes can accelerate significantly under extreme environmental conditions such as high temperature, high pressure, and turbulent fluid flow. When troubleshooting a field corrosion problem, a researcher often needs to return to the lab and reproduce the same (or similar) harsh conditions in a controlled setting. While familiar laboratory equipment for temperature control (ovens, water baths) and pressure control (autoclaves) is generally readily available and easy to use, recreating a fluid flow condition generally poses a larger challenge to the researcher.

Laboratory flow loop systems often require complex and expensive plumbing, maintenance, and calibration to reliably and reproducibly move fluid past a metal sample. The need for this type of large scale laboratory equipment can often be avoided by moving the metal sample with respect to the fluid instead.



Figure 1-1. Fully Assembled Pine Research Instrumentation 15 mm OD Rotating Cylinder Electrode System.



Figure 1-2. Assembled and Disassembled Views of the 15 mm OD Rotating Cylinder Electrode.

A convenient instrument¹⁻³⁵ for rapidly moving a metal sample with respect to a fluid is the Rotating Cylinder Electrode (RCE). This apparatus includes an electrode rotator, RCE electrode shaft, and accessories (see: Figure 1-1) capable of precisely adjusting the rotation rate of a vertically oriented shaft. A special tip capable of holding a cylindrical shaped metal sample is mounted at the lower end of the shaft. The tip is fashioned primarily from chemically inert and electrically insulating materials (such as PTFE, PCTFE, or PEEK), but buried within the tip is a metal shank which provides mechanical stability and also electrical contact with the metal cylinder sample, also called a metal coupon (see: Figure 1-2).

When immersed and rotated in a test solution, the hydrodynamic conditions generated by the RCE, even at low rotation rates, are generally quite turbulent¹⁻⁵. This makes the RCE an ideal probe for studying corrosion processes¹²⁻¹⁵ under turbulent conditions, but at low velocity. By adjusting the RCE rotation rate up or down (typically in the range from 200 to 4000 RPM), it is possible to tune the hydrodynamic conditions²⁰⁻³⁵ adjacent to the metal sample. The ideal goal is to adjust the rotation rate so that the laboratory fluid flow conditions match (or mimic) those found in the field. Once this is accomplished, the corrosion process can be monitored by classic mass loss methods or by electrochemical methods such as Linear Polarization Resistance (LPR)^{21,22} or Electrochemical Impedance Spectroscopy (EIS)^{14,24}.

2. Tuning Turbulent Flow

At very slow rotation rates, the solution near a rotating cylinder flows with a regular and smooth motion called laminar flow. As the rotation rate increases, the solution flow becomes more complex. While the layer of solution in direct contact with the cylinder continues to cling to the surface, the shear stress between this layer and layers further from the cylinder begins to spin off vortices. At this point, the solution flow transitions from laminar to turbulent flow, and as the rotation rate increases, the vortices themselves spawn further vortices.

The transition from laminar to turbulent flow is often characterized using the Reynolds Number (R_E) to quantify the ratio between inertial forces and viscous forces in a solution. For a rotating cylinder electrode¹⁻³ with outer diameter, d_{cyl} (cm), and radius, $r_{cyl} = d_{cyl}/2$, the Reynold's Number is

$$R_E = U_{cyl} d_{cyl} \frac{\rho}{\mu} \quad (1)$$

where ρ is the solution density (g/cm^3), and μ is the absolute viscosity of the solution ($g/cm s$). The linear velocity, U_{cyl} (cm/s), at the outer surface of the cylinder is given by

$$U_{cyl} = \omega r_{cyl} = \frac{\pi d_{cyl} F}{60} \quad (2)$$

where the rate can either be expressed as angular rotation rate, ω (rad/s), or as frequency, F (RPM). In general, for a rotating cylinder, when the Reynolds Number is greater than 200, then the flow is turbulent.

For all but the very slowest rotation rates, the turbulent condition is expected and desired. So, for a typical Pine Research RCE (15 mm OD, see Figure 1-2), rotation rates between 5 and 4000 RPM correspond to a range of Reynolds Numbers spanning several orders of magnitude (Table 9-1). The transition from laminar to turbulent flow occurs just above 20 RPM, when the Reynolds Number exceeds 200. It is worth noting that this transition occurs at a relatively small rotation rate, making the RCE an ideal tool for studying turbulent flow

at low velocity—precisely the condition frequently found in pipeline infrastructures. Higher turbulent velocities are also easily accessible at higher rotation rates.

3. Mass Transport

The turbulent flow at the RCE can bring material from the solution to the surface of the cylinder, and it can also carry material away from the surface. In the context of a corrosion study, the rate of mass transport to and from the metal surface is often the factor which governs the rate of corrosion. A familiar example would be a corrosion process which is limited by how fast oxygen can be transported from the solution to the metal surface.

Early reports by Eisenberg^{1,2} provide the most commonly accepted description for RCE mass transport. In particular, the mass transfer coefficient, K_m (cm/s), to a rotating cylinder is given by the following relationship:

$$K_m = S_H \frac{D}{d_{cyl}} = [0.0791 R_E^{0.7} S_C^{0.356}] \left(\frac{D}{d_{cyl}} \right) \quad (3)$$

where the diffusion coefficient, D (cm^2/s) is usually taken as the diffusion coefficient for the molecule or ion undergoing mass transport, and where S_H and R_E are the dimensionless Sherwood and Reynolds Numbers, respectively. The Schmidt Number, $S_C = \mu / \rho D$, is also a dimensionless number.

Combining equations (1) through (3), the overall mass transfer coefficient to an RCE can be expressed in one of three forms,

$$K_m = 0.0791 d_{cyl}^{-0.3} \left(\frac{\mu}{\rho} \right)^{-0.344} D^{0.644} U_{cyl}^{0.7} \quad (4a)$$

$$= 0.0487 d_{cyl}^{0.4} \left(\frac{\mu}{\rho} \right)^{-0.344} D^{0.644} \omega^{0.7} \quad (4b)$$

$$= 0.0100 d_{cyl}^{0.4} \left(\frac{\mu}{\rho} \right)^{-0.344} D^{0.644} F^{0.7} \quad (4c)$$

depending upon whether the rotation rate is expressed in terms of linear surface velocity (U_{cyl}) angular rotation rate (ω), or rotations per minute (F). Note that the form shown in equation (4a) is that which is most often found in the literature.

4. Wall Shear Stress

The turbulent flow at the RCE induces a wall shear stress on the surface of the cylinder. Again, Eisenberg's original reports^{1,2} offer a well accepted³⁵ equation for the wall stress, τ_{cyl} ($g/cm s$):

$$\tau_{cyl} = 0.0791 \rho R_E^{-0.3} U_{cyl}^2 \quad (5)$$

The wall shear stress for a typical Pine Research RCE tip ($d_{cyl} = 1.5$ cm) over a range of rotation rates is listed in (see: Table 9-1).

5. Electrochemical Measurements

When a rotating cylinder is used as the working electrode in a traditional three-electrode cell configuration, the corrosion behavior can be monitored^{34,35} by measuring the electric current at the cylinder. Electrical connection to the metal cylinder is accomplished by means of a brush contact on the rotating shaft. A potentiostat is employed to impose various potentials on the cylinder electrode while simultaneously measuring the current. The potential signal applied to the cylinder may be a very slow voltage sweep (e.g., Linear Polarization Resistance, LPR^{21,22}), or it may involve a high frequency sinusoidal signal (i.e., Electrochemical Impedance Spectroscopy, EIS^{14,24}).

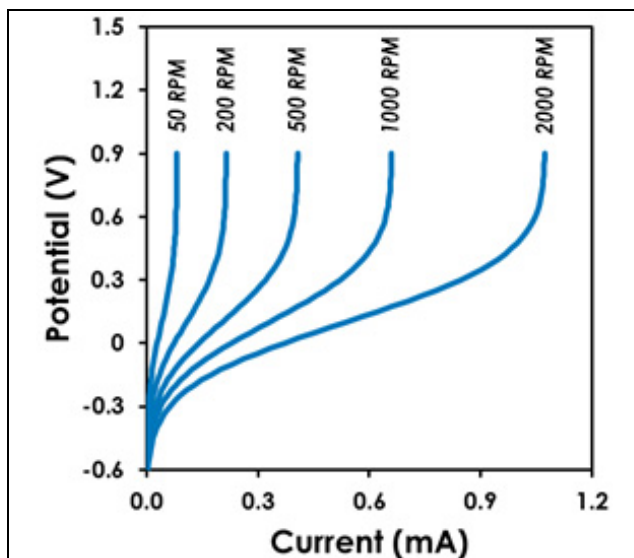


Figure 5-1. Example of a Series of LPR Scans Recorded at Various Rotation Rates.

Two other electrodes are also required to make an electrochemical measurement, a reference electrode (such as a silver/silver-chloride electrode) and a counter electrode. The counter electrode is often an even larger diameter cylinder, rod, wire loop, or flag placed in the solution so that it surrounds the rotating cylinder. This helps to assure uniform current density at the RCE during the test.

In general, the mass transport limited current density, j_{lim} (A/cm^2), observed in an electrochemical experiment is related to the mass transfer coefficient by the following relationship,

$$j_{lim} = \frac{i_{lim}}{A} = zFCk_m \quad (6)$$

where F is Faraday's Constant ($96484.6 C/mol$), i_{lim} (A) is the limiting current, and A (cm^2) is the area of the electrode. To make full quantitative use of this relationship, both the number of electrons exchanged, z , and the bulk concentration, C of the ion or molecule

involved in the electrochemical process must be known.

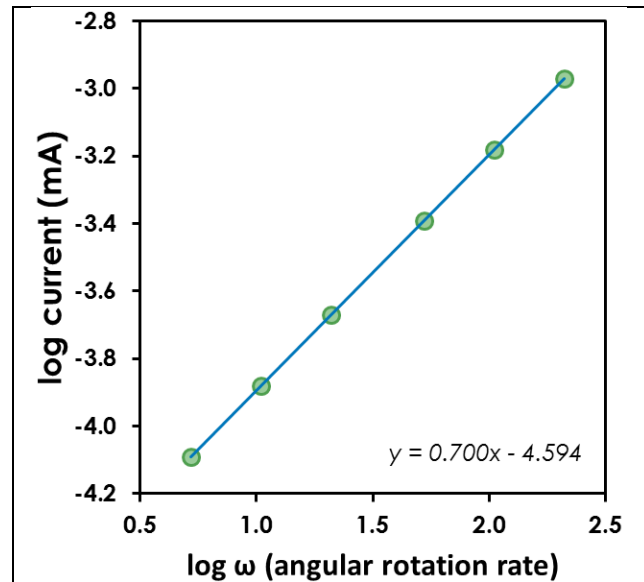


Figure 5-2. Logarithmic Plot to Test for Mass Transport Limited Corrosion Process.

Combining equations (4) and (6), the mass transport limited current density can be expressed as follows:

$$j_{lim} = 0.0791zFCd_{cyl}^{-0.3} \left(\frac{\mu}{\rho}\right)^{-0.344} D^{0.644} U_{cyl}^{0.7} \quad (7a)$$

$$= 0.0487zFCd_{cyl}^{0.4} \left(\frac{\mu}{\rho}\right)^{-0.344} D^{0.644} \omega^{0.7} \quad (7b)$$

Thus, if a corrosion process is limited by mass transport, it is expected that the limiting current (or limiting current density) will vary linearly with the rotation rate raised to the 0.7 power ($\omega^{0.7}$). Note that this behavior can be verified²⁸ even without explicit knowledge of z and C simply by conducting a set of measurements at several different rotation rates.

For example, consider a series of LPR scans performed over a range of rotation rates (see: Figure 5-1). As the rotation rate increases, so does the observed current. A log/log plot of the limiting current (or limiting current density) versus the rotation rate will reveal whether or not the observed current is mass transport limited (see: Figure 5-2). If the slope of a line drawn through the points on this plot is near 0.7, then this is good evidence that the corrosion process is limited by mass transport.

6. Modeling Pipeline Flow

A critical issue when attempting to use the RCE to match or mimic a field corrosion condition is choosing the proper rotation rate at which to perform electrochemical measurements. Several solutions to this problem have been proposed over the years²⁰⁻³⁵. Most involve operating the RCE at a rotation rate where the wall shear stress matches that found in the

field, or alternately, at a rate where the mass transport coefficient at the RCE matches that observed in the field.

The discussion here will be limited to the latter case, but at the outset, it is important to note that modeling a field corrosion situation in the laboratory involves some compromise and some assumptions. When an RCE is operated at a rotation rate which produces similar mass transport conditions to those found in the field, it is assumed²⁸ that the corrosion mechanism occurring in the field will be reproduced in the laboratory. However, it is not expected that the actual corrosion rate at the RCE will match that found in the field. There have been specific cases where the RCE failed²⁰ to reproduce the field corrosion condition. Particular attention is required when surface roughness^{7-10,28} influences mass transport. And lastly, there are practical limitations²⁹ on the range of pipe diameters accessible with the RCE method.

With these caveats in mind, a computational approach outlined in several reports by Silverman²¹⁻²⁹ (who, in turn, references reports by Wranglen³⁰, Holser³¹, Chen³² and Nestic³³) is summarized here. Consider turbulent flow through a smooth, straight pipe. If the flow rate through the pipe, U_p (cm/s), is known, then the target surface velocity, U_{cyl} (cm/s), at an RCE which produces a nearly equivalent mass transport condition can be estimated²⁸ as,

$$U_{cyl} = 0.1185 \left(\frac{\rho}{\mu} \right)^{1/4} \left(\frac{d_{cyl}^{3/7}}{d_p^{5/28}} \right) S_C^{-0.0857} U_p^{5/4} \quad (8)$$

where d_p (cm) is the diameter of the pipe. Using this relationship together with equation (2), it is possible to compute a target rotation rate for the RCE.

Equation (8) has too many parameters to plot convenient working curves on a graph. To convey some idea of the type of results produced using this equation, Table 9-2 lists the pipe velocity/rotation rate relationships for a typical Pine Research RCE ($d_{cyl} = 1.5$ cm and $A = 3.0$ cm²) operating in pure water. As an example, if water is flowing through a smooth 10" Schedule 40 pipe at 1.0 ft/sec, an RCE should be operated at about 131 RPM to match the conditions in the pipe.

Download a spreadsheet calculator from the Pine Research website to aid in these kind of computations: www.pineresearch.com.

7. Corrosion Instrumentation

Pine Research Instrumentation offers specialized tools for the study of mass transport limited corrosion. In addition to our potentiostats for measuring corrosion current and potential, we design and manufacture complete corrosion cell kits. These laboratory corrosion cells have been designed based on input we have received from researchers and practitioners in the corrosion community. The corrosion cell is designed to

be sturdy, long lasting, and easily integrated with other Pine products.



Figure 7-1. Components of the 15 mm OD RCE System.

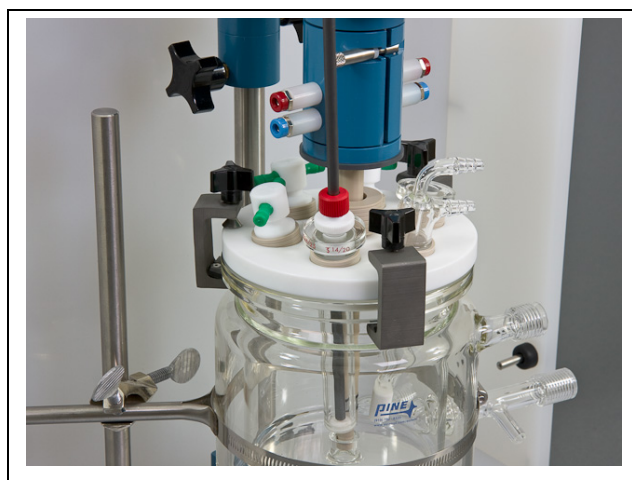


Figure 7-2. Assembled 15 mm OD RCE System with MSR Rotator.

The Pine Research classic RCE system was based on a 12 mm OD RCE (see: Table 10-1), but an improved system based on a 15 mm OD RCE is now available (see: Table 9-1) The Pine Research Instrumentation 15 mm OD Rotating Cylinder Electrode System has many features, such as:

- Reliable electrical contact between the shaft and the replaceable cylinder insert is accomplished using a spring-loaded ball plunger which pushes against the inside diameter of the cylinder insert.
- All shaft components are fabricated from a chemically resistant polymer, polyether ether ketone (PEEK), to protect the shaft from corrosive attack during testing. At elevated temperatures (up to 80 °C) this polymer has good mechanical stability.
- The 15 mm OD allows for greater wall shear at the cylinder surface for a given rotation rate (as compared to the traditional 12 mm OD design).

- The OpenTop cell features a removable lid for easy cleaning. The chemically resistant Teflon lid has six easily configurable cell ports with standard taper adapters for either 14/20 or 24/25 accessories. With these cell ports, the cell can be configured with a variety of accessories (condenser, pH measurement, thermowell, each sold separately) while still having enough ports for the reference and counter electrode.
- The OpenTop cell features a special recess in the bottom of the cell into which the lower end of the RCE shaft is inserted. This recess assists with aligning the shaft along the axis of the glass cell.
- Achievement of finer temperature control with a jacketed cell design.
- The one liter cell allows for larger solution volumes.
- A dual port purge accessory included with the cell permits the solution to be sparged and/or blanketed with a purge gas. In addition, the gas-purged bearing assembly (through which the rotating shaft enters the cell) has a separate purge port to allow a positive purge pressure to be maintained within the void space of the bearing itself.

Figure 7-1 and Figure 7-2 show the components of the 15 mm OD Rotating Cylinder Electrode System. The following additional items are necessary to perform corrosion based measurements:

- A potentiostat to measure corrosion current (Pine Research WaveDriver Series or WaveNow Series).
- An electrode rotator to achieve the desired wall shear stress (Pine Research MSR Electrode Rotator, see: Figure 7-3).

- Metal sample inserts for the RCE electrode (shown in Figure 1-2).
- A Reference electrode for potentiostat control (Ag/AgCl, Saturated Calomel, Hg/HgSO₄, or Hg/HgO).



Figure 7-3. Pine Research Instrumentation MSR Rotator.

8. Reprints

Reprints of this document are available upon request from:

Pine Research Instrumentation
2741 Campus Walk Avenue
Building 100
Durham, NC 27705
USA

<http://www.pineresearch.com>

Sales e-mail: pinewire@pineinst.com

9. Appendix A – 15 mm OD RCE Technical Information and data

The 15 mm OD RCE System uses a specially designed RCE shaft and metals inserts. As shown in Figure 9-1, the shaft length is insulated with PEEK for higher temperature tolerance. The metal cylinder insert fits onto the shaft between seals and is secured with a screw cap. The black seals ensure a tight seal to prevent solution leakage. The RCE shaft (15 mm OD) fits directly into a Pine Research Instrumentation MSR rotator.

The electrode shaft should never be cleaned or treated with acid. The shaft and internal hardware may corrode if exposed to corrosive solutions. Only use the RCE when fully assembled with seals, metal insert, and cap.



Figure 9-1. Assembled and Disassembled Views of the 15 mm OD Rotating Cylinder Electrode.

Rotation Rate F (RPM)	Rotation Rate ω (rad / s)	Surface Velocity* U_{cyl} (cm / s)	Wall Sheer Stress* τ_{cyl} (g / cm s ²)	Reynolds Number* Re (unitless)
5	0.524	0.39	0.0035	66
10	1.047	0.79	0.0113	131
20	2.094	1.57	0.0366	263
50	5.236	3.93	0.1737	657
100	10.47	7.85	0.5642	1315
200	20.94	15.7	1.8332	2629
250	26.18	19.6	2.6789	3287
500	52.36	39.3	8.7039	6573
1000	104.7	78.5	28.279	13146
2000	209.4	157	91.879	26293
3000	314.2	236	183.05	39439
4000	418.9	314	298.52	52586

Table 9-1. Hydrodynamic Computations for a Typical* 15 mm OD Pine Research Rotating Cylinder Electrode in Water.

*These quantities assume a typical Pine Research 15 mm OD RCE tip with outer diameter 1.5 cm rotating in water at 25°C. For pure water at 25°C, the density is 0.997 g/cm³ and the absolute viscosity is 0.00891 g/cm · s.

Pipe Velocity			Standard Schedule 40 Pipe Sizes (actual ID shown in centimeters)								
(ft/s)	(cm/s)	(mi/hr)	2 in	4 in	6 in	8 in	10 in	12 in	16 in	18 in	24 in
			5.25cm	10.23cm	15.41cm	20.27cm	25.45cm	30.32cm	38.1cm	42.88cm	57.48cm
0.1	3.0	0.07									
0.2	6.1	0.14	23	21	19	18	18	17	16	16	
0.3	9.1	0.20	39	34	32	30	29	28	27	26	25
0.4	12.2	0.27	55	49	46	43	42	40	39	38	36
0.5	15.2	0.34	73	65	60	57	55	53	51	50	48
0.6	18.3	0.41	92	81	76	72	69	67	64	63	60
0.7	21.3	0.48	111	99	92	87	84	81	78	76	72
0.8	24.4	0.55	131	117	108	103	99	96	92	90	86
0.9	27.4	0.61	152	135	126	120	115	111	107	105	99
1.0	30.5	0.68	174	154	143	136	131	127	122	119	113
2.0	61.0	1.36	413	366	341	324	311	302	290	284	269
3.0	91.4	2.05	685	608	565	538	517	501	481	471	447
4.0	122	2.73	982	871	810	771	741	718	689	675	640
5.0	152	3.41	1298	1152	1071	1019	979	949	911	892	846
6.0	183	4.09	1630	1447	1345	1280	1229	1192	1144	1120	1063
7.0	213	4.77	1976	1754	1630	1552	1491	1445	1387	1358	1289
8.0	244	5.45	2335	2073	1926	1834	1761	1707	1639	1605	1523
9.0	274	6.14	2705	2401	2232	2125	2041	1978	1899	1859	1764
10.0	305	6.82	3086	2739	2546	2425	2328	2256	2166	2121	2013
11.0	335	7.50	3477	3086	2868	2731	2623	2542	2440	2389	2267
12.0	366	8.18	3876	3441	3198	3045	2924	2834	2721	2664	2528
13.0	396	8.86		3803	3534	3366	3232	3132	3007	2944	2794
14.0	427	9.55			3878	3692	3545	3436	3299	3230	3065
15.0	457	10.23					3865	3746	3596	3521	3341
16.0	488	10.91							3898	3817	3622
17.0	518	11.59									3907
18.0	549	12.27									

Table 9-2. Rotation Rate Correlation for Water between a Typical* 15 mm Pine Research Instrumentation Rotating Cylinder Electrode and Smooth, Straight Pipe Flow.

*These quantities assume a typical Pine Research 15 mm OD RCE tip with outer diameter 1.5 cm rotating in water at 25°C. For pure water at 25°C, the density is 0.997 g/cm³ and the absolute viscosity is 0.00891 g/cm · s. Values that appear grayed out indicate this rotation rate incompatible with maximum or minimum rotation rates.

10. Appendix B – 12 mm OD RCE Technical Information and Data

Our classic 12 mm OD rotating cylinder electrodes have been used in oilfield corrosion inhibitor studies and other high velocity (high shear) corrosion experiments. While Pine Research Instrumentation will, of course, continue to support the 12 mm OD rotating cylinder electrode, our new customers are encouraged to consider our integrated 15 mm OD RCE system instead. The 12 mm RCE design features a PTFE electrode shroud around the shaft connector with PTFE washers as shown in Figure 10-1. The 12 mm RCE tip fits an E3 series shaft for the MSR rotator or the permanently installed shaft on the Pine CPR rotator. The 12 mm RCE tip is not compatible with the 15 mm RCE shaft.

The electrode shaft should never be cleaned or treated with acid. The shaft and internal hardware may corrode if exposed to corrosive solutions. Only use the RCE when fully assembled with seals, metal insert, and cap.

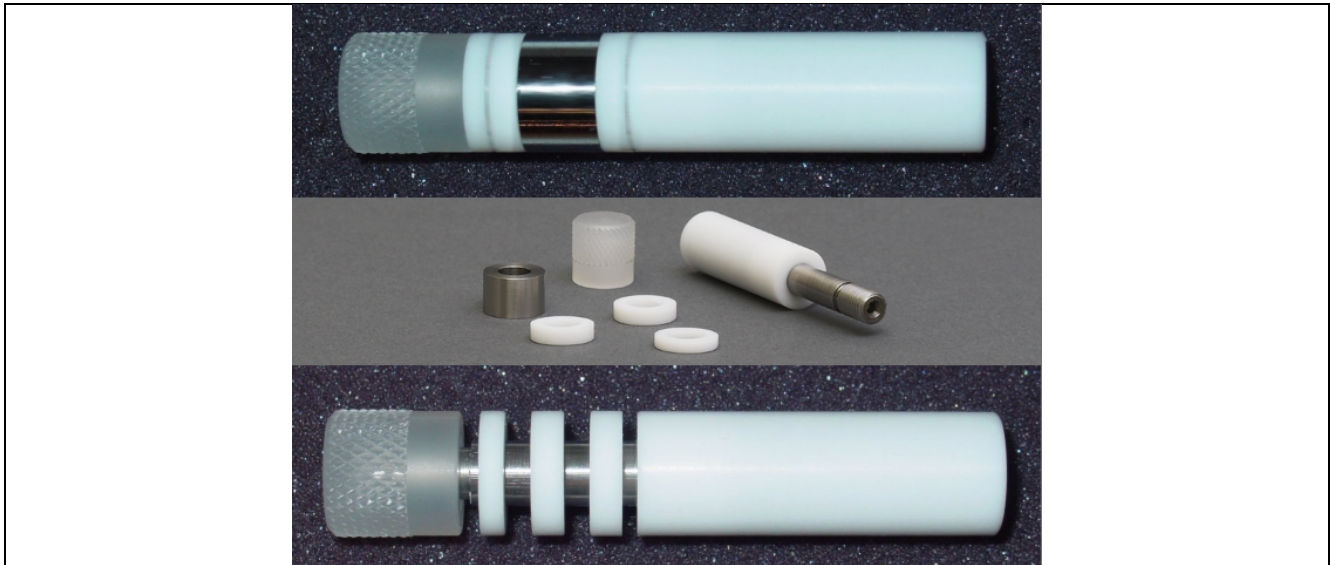


Figure 10-1. Assembled and Disassembled Views of the 12 mm OD Rotating Cylinder Electrode.

Rotation Rate F (RPM)	Rotation Rate ω (rad/s)	Surface Velocity* U_{cyl} (cm/s)	Wall Sheer Stress* τ_{cyl} (g/cm s ²)	Reynolds Number* Re (unitless)
5	0.524	0.31	0.0025	42
10	1.047	0.63	0.0082	84
20	2.094	1.26	0.0267	169
50	5.236	3.14	0.1270	422
100	10.47	6.28	0.4125	844
200	20.94	12.6	1.3402	1688
500	52.36	31.4	6.3631	4219
1000	104.7	62.8	20.674	8438
2000	209.4	125.7	67.169	16876

Table 10-1. Hydrodynamic Computations for a Typical[†] 12 mm OD Pine Research Rotating Cylinder Electrode in Water.

[†]These quantities assume a typical Pine Research 15 mm OD RCE tip with outer diameter 1.5 cm rotating in water at 25°C. For pure water at 25°C, the density is 0.997 g/cm³ and the absolute viscosity is 0.00891 g/cm · s. Values that appear grayed out indicate this rotation rate incompatible with maximum or minimum rotation rates.

Pipe Velocity			Standard Schedule 40 Pipe Sizes (actual ID shown in centimeters)								
(ft/s)	(cm/s)	(mi/hr)	2 in 5.25cm	4 in 10.23cm	6 in 15.41cm	8 in 20.27cm	10 in 25.45cm	12 in 30.32cm	16 in 38.1cm	18 in 42.88cm	24 in 57.48cm
0.1	3.0	0.07									
0.2	6.1	0.14	26								
0.3	9.1	0.20	44	39	36	34	33	32	31	30	29
0.4	12.2	0.27	63	56	52	49	47	46	44	43	41
0.5	15.2	0.34	83	74	68	65	63	61	58	57	54
0.6	18.3	0.41	104	92	86	82	79	76	73	72	68
0.7	21.3	0.48	126	112	104	99	95	92	89	87	82
0.8	24.4	0.55	149	132	123	117	113	109	105	103	97
0.9	27.4	0.61	173	153	143	136	130	126	121	119	113
1	30.5	0.68	197	175	163	155	149	144	138	135	129
2	61.0	1.36	469	416	387	368	354	343	329	322	306
3	91.4	2.05	778	691	642	612	587	569	546	535	508
4	121.9	2.73	1115	990	920	876	841	815	783	766	727
5	152.4	3.41	1474	1308	1216	1158	1112	1078	1035	1013	961
6	182.9	4.09	1851	1643	1527	1454	1397	1354	1299	1272	1207
7	213.4	4.77		1993	1852	1764	1693	1641	1576	1543	1464
8	243.8	5.45						1939	1862	1823	1730
9	274.3	6.14									
10	304.8	6.82									

Table 10-2. Rotation Rate Correlation for Water between a Typical[†] 12 mm Pine Research Instrumentation Rotating Cylinder Electrode and Smooth, Straight Pipe Flow.

[†]These quantities assume a typical Pine Research 15 mm OD RCE tip with outer diameter 1.5 cm rotating in water at 25°C. For pure water at 25°C, the density is 0.997 g/cm³ and the absolute viscosity is 0.00891 g/cm · s. Values that appear grayed out indicate this rotation rate incompatible with maximum or minimum rotation rates.

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