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PURE NIOBIUM AS A PRESSURE VESSEL MATERIAL

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ABSTRACT

Physics laboratories around the world are developing niobium superconducting radio frequency (SRF) cavities for use in particle accelerators. These SRF cavities are typically cooled to low temperatures by direct contact with a liquid helium bath, resulting in at least part of the helium container being made from pure niobium. In the U.S., the Code of Federal Regulations allows national laboratories to follow national consensus pressure vessel rules or use of alternative rules which provide a level of safety greater than or equal to that afforded by ASME Boiler and Pressure Vessel Code. Thus, while used for its superconducting properties, niobium ends up also being treated as a material for pressure vessels. This report summarizes what we have learned about the use of niobium as a pressure vessel material, with a focus on issues for compliance with pressure vessel codes. We present results of a literature search for mechanical properties and tests results, as well as a review of ASME pressure vessel code requirements and issues.

KEYWORDS: niobium, SRF, superconductivity, RF cavities, pressure vessel

INTRODUCTION

Physics laboratories around the world are developing niobium superconducting radio frequency (SRF) cavities for use in particle accelerators. These SRF cavities are typically cooled to low temperatures by direct contact with a liquid helium bath, resulting in at least part of the helium container being made from pure niobium. In the U.S., the Code of Federal Regulations allows national laboratories to follow national consensus pressure vessel rules or use alternative rules which provide a level of safety greater than or equal to that afforded by ASME Boiler and Pressure Vessel Code. Thus, while used for its superconducting properties, niobium ends up also being treated as a material for pressure vessels. Problems with the certification of pressure vessels constructed partially or completely of niobium arise due to the fact that niobium is not listed as an acceptable vessel material in consensus pressure vessel codes (e.g., not in ASME Boiler and Pressure Vessel Code, nor in European nor Japanese codes.) Within the ASME code, in particular, pure niobium is not approved for use in Division 1 or Division 2 vessels [1], and there are no mechanical properties available from code sources. Thus, showing "a level of safety greater than or equal to" that of the applicable standard involves establishing a safely conservative set of niobium mechanical properties for the vessel.

The best RF performance is associated with pure niobium having a very smooth surface, free of mechanical imperfections, free of non-niobium contaminants, and free of entrained hydrogen. Once a niobium SRF cavity has been formed and electron-beam welded, it typically is subjected to various chemical polishing, electropolishing, and heat treatment cycles. This processing is necessary to obtain the required surface conditions and to drive off the hydrogen. Determining the niobium properties in an SRF cavity is complicated by these chemical and thermal treatments applied to the material after forming and welding, as well as by variations among material batches, even from the same vendor.

We will look at pressure vessel compliance implications, review the niobium material properties for the as-received material, mechanical properties after forming and various chemical and heat treatments, and finally look at pressure vessel certification issues.

IMPLICATIONS OF PRESSURE VESSEL CODES

Cavity design that satisfies level of safety equivalent to that of a consensus pressure vessel code is affected by use of the non-code material (niobium), complex forming and joining processes, a shape that is determined entirely by cavity RF performance, a thickness driven by the cost and availability of niobium sheet, and a possibly complex series of chemical and thermal treatments.

FIGURE 1 illustrates an SRF cavity within a helium vessel. FIGURES 2 and 3 are photos of the niobium cavity and helium vessel which surrounds the cavity. This configuration has become standard for electron accelerators built and under development, including CEBAF (Jefferson Lab), Spallation Neutron Source (SNS) at Oak Ridge, TESLA Test Facility (TTF) at DESY, TESLA, XFEL at DESY, International Linear Collider (ILC), and Project X at Fermilab. Pressure from the liquid helium surrounding the niobium SRF cavity typically results in an internal pressure on the surrounding helium jacket and head components, and an external pressure on the niobium cavity itself. This pressure may occur both at ambient temperatures and at cryogenic temperatures. In either case, the niobium cavity is susceptible to buckling failure as well as tensile stresses in the end parts and joints to the surrounding helium vessel.

The ASME pressure vessel code allows two general approaches to pressure vessel design: design by specific, detailed rules (Division 1), and a more open "design by analysis" approach (Division 2).

Difficulties emerge with ASME code Division 1 (which provides specific rules for pressure vessel details) in two primary areas: 1) loadings other than pressure, and 2) geometries not covered by rules. Considering loads, Division 1 provides very little guidance for thermal contraction loads and the imposition of controlled displacements, both relevant to the design of SRF cavities. Considering geometries, the functional heart of a cavity assembly – the formed niobium shell - cannot be designed by Division 1 rules.

The typical approach to achieving a Division 1 vessel under these circumstances is to invoke the provision of paragraph U-2(g), which states: "This Division of Section VIII does not contain rules to cover all details of design and construction. Where complete details are not given, it is intended that the Manufacturer, subject to the acceptance of the Inspector, shall provide details of design and construction which will be as safe as those provided by the rules of this Division." In the case of the SRF cavities, the great majority of the design must be justified by U-2(g). Thus, a detailed analysis of the design of the niobium cavity is required.

In applying ASME code procedures, key elements demonstrating the required level of design safety are the establishment of a maximum allowable stress, and (for external pressure design) an accurate approximation to the true stress strain curve.

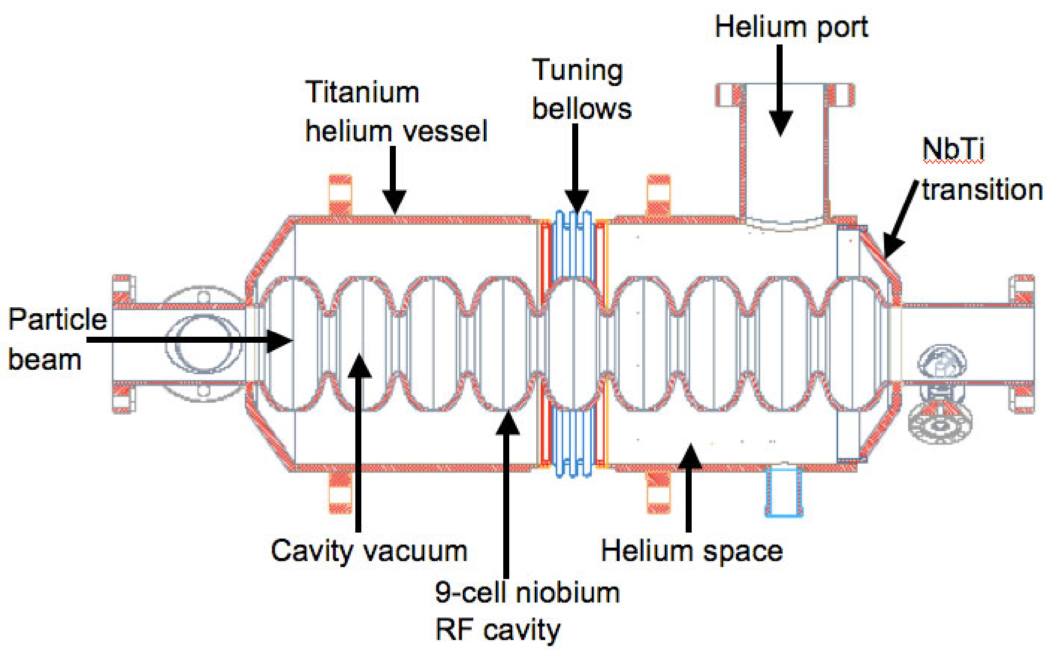

FIGURE 1. An example illustration of a niobium SRF cavity within a helium vessel.

FIGURE 2. A photo of a 1.3 GHz, 9-cell, niobium SRF cavity. Stiffening rings are seen (with hole for helium penetration) between cells. This niobium structure is externally cooled by and pressurized by the helium bath.

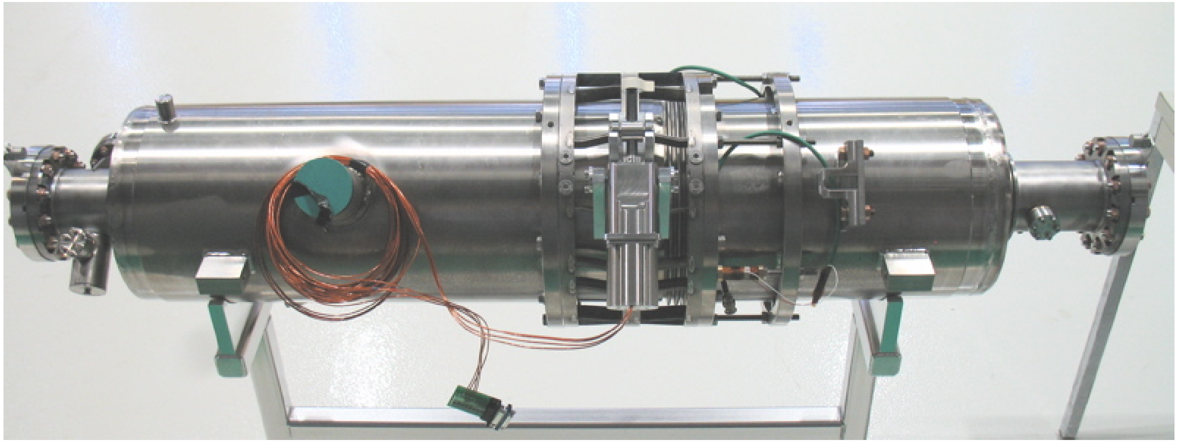

FIGURE 3. Photo of a titanium helium vessel which surrounds a 1.3 GHz, SRF cavity. A tuner mechanism which surrounds bellows is near the center of the cylindrical vessel.

NIOBIUM SPECIFICATION FOR SRF CAVITIES – INITIAL MATERIAL PROPERTIES

The initial niobium specification sets the limit of certain physical properties. Vendors often supply the material with some variations of mechanical properties within the specifications, mostly due to variable purity (often expressed in terms of the residual resistivity ratio, RRR) and grain size. TABLE 1 summarizes some mechanical property requirements from Fermilab's niobium specification for the as-delivered product (prior to the various subsequent processing steps).

TABLE 2 summarizes the allowable impurities in niobium from the DESY specification.

TABLE 1. Some requirements from Fermilab's niobium specification [2]

RRR	Yield Strength (MPa)	Tensile Strength (MPa)	Elongation	Vickers hardness (HV 10)	Grain size (μm)
300	48.2 (7000 psi) to 70.2 (10200 psi)	96.4 (14000 psi)	≥40% longitudinal, ≥35% transverse	≤ 60	≤ 64

TABLE 2. From the DESY specification, allowable impurities in niobium [3]

Concentration of impurities in ppm (weight)									
Ta	W	Ti	Fe	Mo	Ni	H	N	O	C
≤ 500	≤ 70	≤ 50	≤ 30	≤ 50	≤ 30	≤ 2	≤ 10	≤ 10	≤ 10

TABLE 3. Some examples of room-temperature properties of as-received high RRR niobium.

RRR	Yield Strength (MPa)	Tensile Strength (MPa)	Elongation (%)	Reference
492	73	199	57	4
260	51	145	44	5
310	76	142	56	6

TABLE 4. Averages from St. Louis Testing Laboratory report on as-received Wah Chang niobium purchased per the Fermilab specification [2], summarized in TABLE 1.

Sample	Test Temperature (K)	Yield Strength (MPa)	Tensile Strength (MPa)	Charpy impact (J)
Transverse	295	76.6	184.4	31.5
Longitudinal	295	64.0	185.1	27.9
Transverse	77	556.7	666.2	6.0
Longitudinal	77	577.8	673.7	4.6
Transverse	4.5	698.8	740.6	3.5
Longitudinal	4.5	698.8	745.1	3.0

TABLES 3 and 4 summarize as-received properties for niobium. This niobium is in the form of sheet which has been cold-worked. The cavity manufacturing processes, such as e-beam welding and heat treatment, can subsequently change the mechanical properties from the "as-delivered" condition as we discuss below.

MATERIAL PROCESSING AND CAVITY ASSEMBLY SUMMARY

Starting with RRR 250 - 300 nominal material as described above, the niobium cavity parts are cut and formed. Components are welded together, and various heat treatments and chemical treatments are done to enhance surface cleanliness and material purity, with the goals being RF performance and high thermal conductivity. The recipes for optimizing RF performance have been subjects of extensive R&D, so reports describe quite a large variety of heat treatment temperatures and durations.

Our specific interest was to understand the material properties of RRR niobium subjected to the following formation and processing steps:

1. Recrystalization at 875 K (600 C) – 1075 K (800 C)
2. Half cells are formed
3. Cavity is electron beam welded
4. Cavity external etch (buffered chemical processing, or BCP) 20 minutes
5. Internal bulk etch (BCP) 80 minutes
6. Bake (for hydrogen degassing) at 1075 K (800 C) for 2 hours
7. Internal light etch (BCP) 20 minutes
8. Test niobium cavity in liquid helium in vertical position
9. Assemble and weld the helium vessel around the cavity
10. Light internal cavity etch

FINAL POST TREATMENT MECHANICAL PROPERTIES

Given the as-received material, and the subsequent assembly and heat treating steps described above, the question arises as to what mechanical properties the designer should use for analysis and code compliance documentation. This section consists of some conclusions regarding the yield and ultimate stresses for pure niobium following a survey of the literature. Of particular interest and concern are the properties of formed and welded niobium following heat treatment at 1075 K (800 C) for 2 to 3 hours, which has been found to be effective for hydrogen degassing.

RRR, Heat Treatment, and Mechanical Properties

G. R. Myneni and H. Umezawa [5] reported on room temperature tensile tests of reactor grade niobium. The same reactor grade niobium was also post purified to improve the RRR. TABLES 5 and 6 summarize the mechanical properties of niobium before heat treating and after various heat treatments from this reference.

TABLE 5. Mechanical properties of reactor grade niobium [5]

Sample	RRR	Yield Strength (MPa)	Tensile Strength (MPa)	Elongation (%)
As received	68	110	226	50
1375 K (1100 C), 3 hr	57	76	183	50
1475 K (1200 C), 6 hr	119	64	165	50
1525 K (1250 C), 6 hr	210	56	120	53

TABLE 6. Mechanical properties of Tokyo Denkai high RRR niobium (strain rate in mechanical tests was 5.56 x 10^{-5} mm/mm per second) [5]

Niobium treatment	RRR	Yield Strength (MPa)	Tensile Strength (MPa)	Elongation (%)
As received	260	51	145	44
875 K (600 C)	300	48	145	48
1075 K (800 C)	350	39	131	47
1525 K (1250 C)	375	31	103	32

TABLE 7. Effect of 600 C and 800 C heat treatments on niobium [6]

Sample	Niobium treatment	Yield Strength (MPa)	Tensile Strength (MPa)	Elongation (%)
1	As received	65 – 85	134 – 177	54 – 61
1	875 K (600 C)	65 – 72	156 – 181	54 – 57
1	1075 K (800 C)	38 – 48	145 – 163	50 – 60
2	As received	90 – 97	161 – 164	41 – 51
2	875 K (600 C)	62 - 72	157	57 – 62
2	1075 K (800 C)	45 – 49	148	59
3	As received	186	214	46-60
3	875 K (600 C)	69 – 103	172	22 – 34
3	1075 K (800 C)	38	152 – 157	40

Note that the RRR improves with higher temperature heat treating, and the material softens as reflected in the reduced tensile and yield strengths. Since the RF cavities must retain their shapes in a stable manner, one does not want excessively low yield strength. So the goal of some of the R&D has been to find a heat treatment which sufficiently degasses hydrogen from the niobium but does not so severely anneal it as a higher temperature bake. The 1175 K (800 C) bake for 2 – 3 hours seems to work well and was used recently by Fermilab.

Myneni and Kneisel [6] reported material studies carried out for SNS. We summarize the results in TABLE 7. A range is shown for their various data samples except for those where the range was very small. These results illustrate the higher strengths which lead to their favoring a 875 K (600 C) bake over a 1075 K (800 C) bake. Fermilab, however, performed the hydrogen degassing at 1100 K (825 C), so we use the 1075 K data above as guidance for our material properties.

In our literature search, the lowest yield strength which we found reported in more than one test for niobium baked at 1075 K (800 C) was the last line of TABLE 7, 38 MPa. A representative but conservatively low tensile strength value for 1075 K (800 C) baked niobium is 130 MPa. We conclude that a conservative estimate for the room temperature yield and ultimate strengths of high RRR niobium, heat treated at 1075 K (800 C) for 2 hours, are 38 MPa yield and 130 MPa ultimate tensile.

A non-linear buckling analysis of the externally pressurized cavity requires a stress-strain curve for the as-treated niobium. We wanted a very conservative, representative stress-strain curve for niobium, rather than one from just one particular sample of material or from the literature. So a niobium stress-strain curve (FIGURE 4) was derived from formulas in Part 3 of Division 2 of the ASME code [1]. This curve is based on our conclusion of 38 MPa yield and 130 MPa ultimate tensile strength, the modulus of elasticity (1.05e5 MPa) and some parameters from code values for Ti and Zr, which are in the same family of metals. This curve is in good agreement with published stress-strain curves for the softest heat-treated niobium.

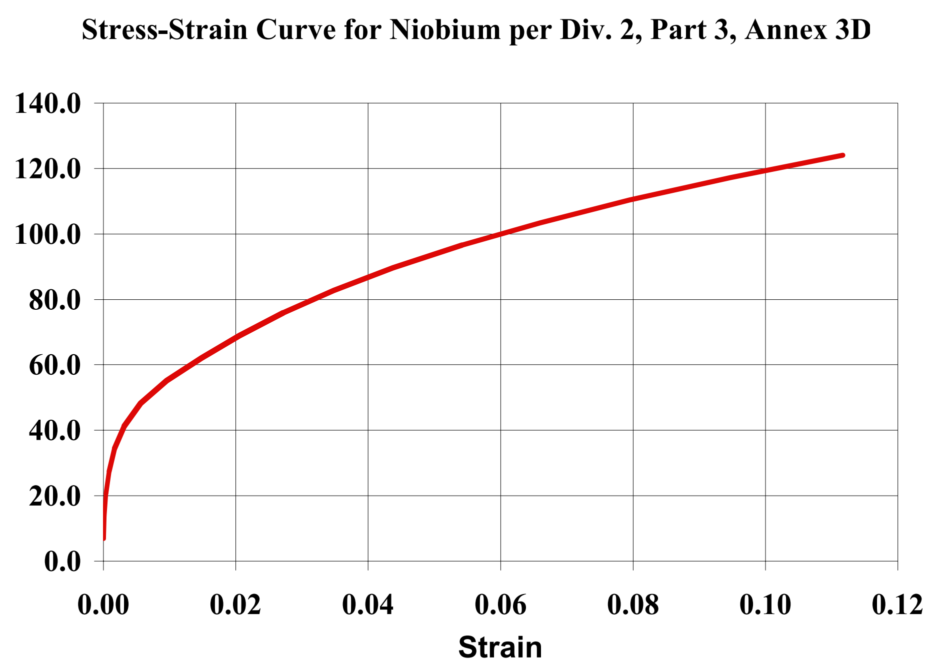

FIGURE 4. Niobium stress-strain curve for buckling analysis derived from parameters including our conclusions regarding yield and ultimate strengths for as-treated material.

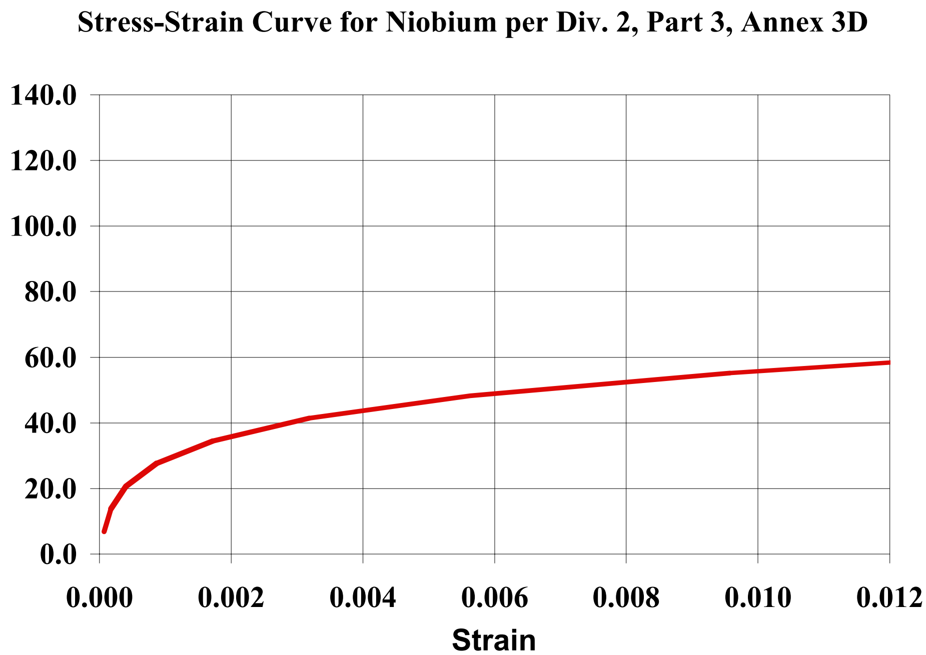

FIGURE 5. The same niobium stress-strain curve as in FIGURE 4, expanded for lower strain by a factor of 10.

Weld Design Issues

Several issues regarding weld design and weld certification arise in the fabrication of niobium RF cavities. The niobium cavities are electron-beam welded from formed half-cells. Electron beam welds in any material are required to be ultrasonically examined along their entire length per UW-11(e) of the ASME pressure vessel code, but not all welds are accessible for ultrasonic inspection.

A transition from the niobium cavity to titanium helium vessel typically involves welds from niobium to NbTi and NbTi to titanium. The ASME code prohibits welding titanium alloys to non-titanium materials, so these areas of transition will generally also be exceptions to the code.

Low Temperature and Weld Mechanical Properties for Niobium

In addition to the issue of the niobium cavity operating at a temperature as low as 2 Kelvin, the peak pressure may be significantly higher at low temperature than at room temperature. Room temperature pressures are determined by operational considerations such as cool-down and warm-up. For SRF cavity systems, this room temperature peak pressure may be as low as 2 bar differential. (2 bar absolute with vacuum in the cavity and vacuum around the helium vessel.) However, at liquid helium temperatures, a loss of insulating vacuum or cavity vacuum can cause rapid warming and pressurization of the helium. It may not be practical to provide vent lines large enough to limit the pressure to 2 bar, so a higher peak cold pressure must be verified as safe.

At 4.2 K the strength of niobium increases about a factor of 6 or greater, however the material becomes very brittle. High purity niobium (RRR=250) retains more ductility at 4.2 K than lower purity material (RRR=40) [7]. TABLE 8 contains a comparison of mechanical properties from Walsh, et. al. [7].

Table 8. Comparison of low temperature with room temperature properties of RRR = 250 niobium [7].

Temperature (K)	Niobium treatment	Yield Strength (MPa)	Tensile Strength (MPa)	Elongation (%)
295	Hot-rolled plate	67	172	57
77	Hot-rolled plate	618	642	30
4	Hot-rolled plate	658	929	16
295	e-beam weld	70	151	28
77	e-beam weld	445	639	13
4	e-beam weld	470	696	4.2

TABLE 9. Niobium mechanical properties conclusion from our literature search

				ASME Sec VIII, Div 1	
Yield strength (YS)	Tensile strength (TS)	approx RRR	heat treatment	allowable stress for non-ferrous metals	
				Take the smaller of:	
				2/3 x YS	TS/3.5
MPa	MPa			(MPa)	(MPa)
38	130	300	1100 K (825 C), 2.4 hr	**25**	37

One can see from the data in Table 8 the great increase in strength and reduction of ductility of niobium with low temperature.

The ASME Pressure Vessel Code, Section VIII, Div 1, UG-98(c) says, "Maximum allowable working pressure may be determined for more than one designated operating temperature, using for each temperature the applicable allowable stress value." [1] Thus, one may have a higher cold MAWP with the higher cold allowable stresses shown in TABLE 8.

ASME CODE ALLOWABLE STRESS

The last line in TABLE 9 is our conclusion from our literature search regarding conservative yield and ultimate strengths for 800 C heat treated niobium. Combining the information summarized above, we have for room temperature niobium, following an 800 C bake of 2-3 hours, an allowable stress for Section VIII, Division 1 as follows (explaining TABLE 9). From the ASME code (Table 1-100 in mandatory appendix 1, Section II, Part D), use the lowest of either TS/3.5 or 2/3 x YS. (Our niobium cavities do not have a longitudinal weld, so we will not take the 0.85 factor for welded pipe or tube.) So TS/3.5 = 130/3.5 = 37 MPa. YS x 2/3 = 38 x 2/3 = 25 MPa. Therefore, the allowable stress for 800 C baked niobium is S = 25 MPa.

The low temperature (80 kelvin or below) allowable stress is based on the tensile strength rather than yield strength due to the brittle nature of the low temperature material. A conservative result as can be seen from TABLE 8, allowing for some variability of samples, is 600 MPa/3.5 = 171 MPa, over six times the room-temperature value.

CONCLUSIONS

A search of the literature on pure niobium found that reported ultimate and yield strength for niobium are extremely variable, depending on various steps of forming, welding, and heat treating. Other sources which we consulted and which informed our assessment of a niobium as a pressure vessel material, in addition to those already quoted and cited, include the following references [8-15]. The result for analysis of pressure vessels made from niobium as treated for Fermilab RF cavities (the key step being a 2 hour, 1100 K bake) has been to use a very low, conservative yield stress estimate of 38 MPa, an ultimate strength of 130 MPa, and an ASME allowable room temperature stress of 25 MPa. We take credit for the increase in strength at low temperature in order to justify a higher maximum allowable working pressure at liquid helium temperatures. We conclude that a low temperature allowable stress of 171 MPa is a very conservative and reasonable value for vessel analysis. Finally, for a plastic analysis of buckling, a stress-strain curve based on these room-temperature yield and ultimate strengths is derived.

A companion paper describing Fermlab's proposed methodology for treating RF cavities as pressure vessels, "Guidelines for the design, fabrication, testing, installation, and operation of SRF cavities," was presented at this conference (paper C3-F-02) [16].

ACKNOWLEDGMENTS

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