Density of Gadolinium Nitrate Solutions for the High Flux Isotope Reactor

May 2009

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Nuclear Science and Technology Division

**DENSITY OF GADOLINIUM NITRATE SOLUTIONS
FOR THE HIGH FLUX ISOTOPE REACTOR**

P. A. Taylor and D. L. Schuh

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EXECUTIVE SUMMARY

In late 1992, the High Flux Isotope Reactor (HFIR) was planning to switch the solution contained in the poison injection tank from cadmium nitrate to gadolinium nitrate. The poison injection system is an emergency system used to shut down the reactor by adding a neutron poison to the cooling water. This system must be able to supply a minimum of 69 pounds of gadolinium to the reactor coolant system in order to guarantee that the reactor would become subcritical. A graph of the density of gadolinium nitrate solutions over a concentration range of 5 to 30 wt% and a temperature range of 15 to 40°C was prepared. Routine density measurements of the solution in the poison injection tank are made by HFIR personnel, and an adaptation of the original graph is used to determine the gadolinium nitrate concentration. In late 2008, HFIR personnel decided that the heat tracing that was present on the piping for the poison injection system could be removed without any danger of freezing the solution; however, the gadolinium nitrate solution might get as cold as 5°C. This was outside the range of the current density-concentration correlation, so the range needed to be expanded. This report supplies a new density-concentration correlation that covers the extended temperature range. The correlation is given in new units, which greatly simplifies the calculation that is required to determine the pounds of gadolinium in the tank solution.

The procedure for calculating the amount of gadolinium in the HFIR poison injection system is as follows.

1. Calculate the usable volume in the system
2. Measure the density of the solution
3. Calculate the gadolinium concentration using the following equation:

$$\text{Gd} \left(\frac{\text{lb}}{\text{ft}^3} \right) = \text{measured density} \left(\frac{\text{g}}{\text{mL}} \right) \times 34.681 - 34.785$$

4. Calculate the amount of gadolinium in the system using the following equation:

$$\text{Amount of Gd(lb)} = \text{Gd concentration} \left(\frac{\text{lb}}{\text{ft}^3} \right) \times \text{usable volume}(\text{ft}^3)$$

The equation in step 3 is exact for a temperature of 5°C, and overestimates the gadolinium concentration at all higher temperatures. This guarantees that the calculation is conservative, in that the actual concentration will be at least as high as that calculated. If an additional safety factor is desired, it is recommended that an administrative control limit be set that is higher than the required minimum amount of gadolinium.

1. BACKGROUND

In late 1992 Ken Morgan (Research Reactors Division) asked Paul Taylor (Chemical Technology Division, currently the Nuclear Science and Technology Division) to supply a graph of the density of gadolinium nitrate solutions over a concentration range of 5 to 15 wt%, which was later expanded to a maximum of 30 wt%, and a temperature range of 15 to 40°C. The High Flux Isotope Reactor (HFIR) was planning to switch the solution contained in the poison injection tank from cadmium nitrate to gadolinium nitrate. The poison injection system is an emergency system that can be used to shut down the reactor by adding a neutron poison to the cooling water. The density values were obtained from two published papers.^{1,2} The first paper had density values for solutions of gadolinium nitrate in water over the desired range of concentrations, but only at 25°C, while the second paper had data for the density of gadolinium chloride over the desired range of concentrations and temperatures. The ratio of density change from 25°C for each of the desired temperatures at each concentration was calculated from the gadolinium chloride solution data, and then these ratios were used to calculate the density of gadolinium nitrate solutions at these conditions by multiplying the density at 25°C by the appropriate ratio.

In late 2008, HFIR personnel decided that the heat tracing that was present on the piping for the poison injection system could be removed without any danger of freezing the solution; however, the gadolinium nitrate solution might get as cold as 5°C. This was outside the range of the current density-concentration correlation, so the range needed to be expanded.

The control limits for the gadolinium nitrate solution are based on the total quantity of gadolinium in the poison injection system tank. Because the density-concentration correlation was given in wt% gadolinium nitrate, the procedure to calculate the quantity of gadolinium in the tank was much more complicated than necessary. A copy of the current calculation worksheet is shown in Fig. 1. The height of solution in the tank is measured, and then the usable volume of solution is calculated (steps A and B). These steps would remain the same. A sample of the solution is taken, and the density (specific gravity) of the solution is measured with a hydrometer. The density-concentration correlation is used to determine the concentration of gadolinium nitrate, as wt% $\text{Gd}(\text{NO}_3)_3$ in the solution (step D). The density measurement is also used to calculate the weight of solution, using the volume of solution that was previously calculated (step C). The amount of gadolinium nitrate is then calculated from the weight of solution and the wt% $\text{Gd}(\text{NO}_3)_3$ (step E). Finally the weight of gadolinium is calculated from the weight fraction of gadolinium in $\text{Gd}(\text{NO}_3)_3$ (step F). Many of these steps could be avoided if the density-concentration correlation were given in units of lb Gd/ft^3 vs density, rather than wt% $\text{Gd}(\text{NO}_3)_3$ vs density.

Section 3 of this report describes the methods for calculating the new concentration-density correlation and gives the new procedure for calculating the amount of gadolinium in the tank.

A. Volume in Tank		
_____ in ÷ 12 in/ft	X 4.91 ft ²	= _____ ft ³ of solution
Adjusted level from Step 1.5		
B. Total Volume		
_____ ft ³	- 0.302 ft ³	= _____ ft ³
Volume in Tank (result from A)	Unusable Volume in System	Total Volume
C. Mass in Tank		
_____ ft ³	X _____ X 62.4 lb/ft ³	= _____ lbs of solution
(result from B)	Specific Gravity (from Step 1.8)	
D. Gram Conversion as Percentage		
_____	= _____ %	
Specific Gravity from Step 1.8		From Concentration vs Specific Gravity Figure
E. Specific Gravity to Mass of Gadolinium Nitrate		
_____ lbs	X [_____ ÷ 100]	= _____ lbs of Gadolinium Nitrate
(result from C)	(result from D)	
F. Mass of Gadolinium in the Solution		
_____ lbs	X 0.458	= _____ lbs of Gadolinium
(result from E)		

Fig. 1. Current worksheet for calculating the quantity of gadolinium in the poison injection system tank.

2. DESCRIPTION OF THE HFIR POISON INJECTION SYSTEM

Criterion 26 of the Nuclear Regulatory Commission General Design Criteria for Nuclear Power Plants, Appendix A of 10 CFR 50³, specifies redundancy in a reactivity control system. At HFIR this criterion is met through the use of control plates, the shim regulating cylinder, and the poison injection system. The poison injection system was provided as an alternate mechanism for ensuring nuclear shutdown in the event of very low probability events, which would render the movable shim-safety plates and shim-regulating cylinder ineffective. This system is maintained in the design, in combination with the shim-safety plates and shim-regulating cylinder systems, to provide diverse, redundant nuclear shutdown capability.

Use of the poison-injection system is restricted to conditions when it is impossible to maintain control of the reactor by manipulation of the control elements by any of the normal insertion methods. If all control elements became inoperable during normal power operation, the operating power level would steadily drop to compensate for fuel burnup, and a relatively small negative reactivity would be

sufficient to maintain the reactor subcritical following xenon decay. If the control elements should become inoperable when reactivity is being steadily increased due to xenon burnout following a scram and restart of the reactor, then (a) sufficient negative reactivity must be available in the poison injection system to shut down with the control elements fully withdrawn, and (b) the response time of the poison injection system must be short enough that the reactor can be shut down before the power is increased to a point that approaches the core thermal limits. This “xenon burnout” scenario was selected to provide the design basis for the system. The maximum negative reactivity required of the system was taken as the excess reactivity of a fresh core with control rods fully withdrawn, and the response time of the system to obtain full injection and mixing of the poison solution was taken as 2½ min. The release of soluble poison into the coolant system within 2½ min of a “servo insert error” alarm will shut down the reactor without reaching the core thermal limits, assuming the maximum rate of xenon burnout is consistent with the allowable restart power schedule.

The design of the poison injection system is as follows: The amount of neutron poison available shall be sufficient, when uniformly distributed in the primary coolant system, to completely poison a clean core with the control rods fully withdrawn. The neutron poison shall have a chemical form that is compatible with the materials used in the primary coolant system. The solubility and concentration of the neutron poison shall be such that it has adequate margin for solution under the normal ambient conditions present in the poison injection system. The poison injection system must be operable any time the reactor is operating at power levels greater than 8.5 MW.

The material selected to satisfy these criteria for use in the HFIR poison injection system was gadolinium nitrate ($\text{Gd}(\text{NO}_3)_3$). The amount of gadolinium material required to maintain subcriticality under the above conditions was calculated to be a concentration of 0.6 grams gadolinium per liter of water. This translates to be a minimum of 69 lb of gadolinium (not $\text{Gd}(\text{NO}_3)_3$) dissolved in the HFIR primary coolant system. The amount of $\text{Gd}(\text{NO}_3)_3$ dissolved in the HFIR poison injection system is verified monthly.

3. CALCULATION OF DENSITY-CONCENTRATION CORRELATION

Table 1 shows the data on the density of gadolinium nitrate solutions at 25°C from reference 1. The data covers the range of concentrations of interest; however, there is no information on the effect of temperature in this reference. The reference gives the concentrations in molal (moles/kg of solvent). The table shows the concentrations in additional units, including weight percent (wt% = g $\text{Gd}(\text{NO}_3)_3$ /100 g of solution) and lb Gd/ft³ of solution. To convert from molal to wt% $\text{Gd}(\text{NO}_3)_3$, the weight of $\text{Gd}(\text{NO}_3)_3$ in the solution is divided by the total weight of solution (water + $\text{Gd}(\text{NO}_3)_3$). The equation is as follows:

$$C_2(\text{wt}\%) = \frac{C_1 \left(\frac{\text{moles}}{\text{kg water}} \right) \times \text{MW of } \text{Gd}(\text{NO}_3)_3 \left(\frac{\text{g}}{\text{mole}} \right)}{\left(1000 \text{ g water} + C_1 \left(\frac{\text{moles}}{\text{kg water}} \right) \times \text{MW of } \text{Gd}(\text{NO}_3)_3 \left(\frac{\text{g}}{\text{mole}} \right) \right) / 100}$$

To convert from wt% $\text{Gd}(\text{NO}_3)_3$ to lb Gd/ft³ of solution, the weight fraction (wt%/100) is multiplied by the solution density and by the weight fraction of gadolinium in $\text{Gd}(\text{NO}_3)_3$. The equation is as follows:

$$C_3 \left(\frac{\text{lb Gd}}{\text{ft}^3} \right) = \frac{C_2(\text{wt}\%) \times \text{Density} \left(\frac{\text{g}}{\text{mL}} \right) \times 62.4 \left(\frac{\text{lb/ft}^3}{\text{g/mL}} \right) \times 0.458 \left(\frac{\text{lb Gd}}{\text{lb } \text{Gd}(\text{NO}_3)_3} \right)}{100}$$

Table 1. Density of gadolinium nitrate–water solutions at 25°C¹

Gd(NO₃)₃ concentrations		Gd conc. (lb Gd/ft³)	Density (g/mL)
(molal)	(wt%)		
0.1532	4.996	1.486	1.0409
0.2068	6.628	2.000	1.0559
0.2947	9.187	2.836	1.0802
0.4092	12.316	3.912	1.1113
0.5991	17.057	5.662	1.1615
0.7955	21.449	7.428	1.2118
1.0108	25.759	9.311	1.2648
1.2150	29.431	11.048	1.3135

Data on the density variation at various temperatures for gadolinium nitrate solutions was not available when the initial density calibration was done; therefore, published data for gadolinium chloride solutions² was used to estimate the density of gadolinium nitrate solutions at various temperatures. Table 2 shows the data from reference 2, which is in the form of coefficients for a power function equation.

Table 2. Coefficients for power function equation to calculate gadolinium chloride solution densities^a

[Gd Cl₃] (molal)	C₀	C₁	C₂	C₃	C₄	C₅
0.0978	1.0243	2.758E-05	-7.673E-06	5.884E-08	-3.832E-10	1.1630E-12
0.2368	1.0584	-1.761E-05	-6.806E-06	4.859E-08	-3.045E-10	8.9523E-13
0.4105	1.1002	-7.157E-05	-5.777E-06	3.618E-08	-2.075E-10	5.6368E-13
0.5894	1.1423	-1.197E-04	-4.996E-06	2.863E-08	-1.584E-10	4.1682E-13
0.7842	1.1874	-1.696E-04	-4.151E-06	1.917E-08	-8.740E-11	1.8198E-13
0.9947	1.2349	-2.147E-04	-3.565E-06	1.518E-08	-7.142E-11	1.5841E-13
1.2239	1.2854	-2.600E-04	-3.038E-06	1.258E-08	-7.384E-11	2.2978E-13

^aAdapted from Habenschuss and Spedding²

Note: Density = C₀ + C₁(T) + C₂(T)² + C₃(T)³ + C₄(T)⁴ + C₅(T)⁵, where T = Temperature (°C).

Table 3 shows the densities of gadolinium chloride solutions at various concentrations and temperatures, which were calculated from the equation and coefficients listed above. Table 4 shows the ratio of the density at each temperature to the density at 25°C at each concentration. These ratios were used to calculate the density of the gadolinium nitrate solutions at various temperatures, from the published densities at 25°C. The maximum difference between the densities at 5°C and 40°C was 1.1%, which occurred at the highest gadolinium chloride concentration, so temperature within this range has a relatively small effect on the density of the solutions. Since the density variation with temperature is small, and gadolinium nitrate and gadolinium chloride solutions should have similar temperature dependencies, using the measured gadolinium chloride temperature dependency to predict the temperature dependency is justified.

Table 5 shows the calculated densities for gadolinium nitrate solutions at various temperatures. The values were calculated from the published densities at 25°C, shown in Table 1, and the ratios shown

Table 3. Density of gadolinium chloride solutions

[Gd Cl ₃] (molal)	Density of solution (g/mL) at each temperature (°C)							
	5	10	15	20	25	30	35	40
0.0978	1.0243E+00	1.0239E+00	1.0232E+00	1.0222E+00	1.0210E+00	1.0195E+00	1.0179E+00	1.0160E+00
0.2368	1.0581E+00	1.0575E+00	1.0566E+00	1.0553E+00	1.0536E+00	1.0515E+00	1.0490E+00	1.0461E+00
0.4105	1.0997E+00	1.0989E+00	1.0978E+00	1.0964E+00	1.0947E+00	1.0927E+00	1.0903E+00	1.0876E+00
0.5894	1.1416E+00	1.1406E+00	1.1394E+00	1.1379E+00	1.1361E+00	1.1341E+00	1.1318E+00	1.1292E+00
0.7842	1.1864E+00	1.1853E+00	1.1839E+00	1.1823E+00	1.1805E+00	1.1785E+00	1.1763E+00	1.1738E+00
0.9947	1.2337E+00	1.2324E+00	1.2309E+00	1.2292E+00	1.2273E+00	1.2252E+00	1.2229E+00	1.2204E+00
1.2239	1.2840E+00	1.2825E+00	1.2808E+00	1.2790E+00	1.2770E+00	1.2748E+00	1.2725E+00	1.2700E+00

Table 4. Ratios of gadolinium chloride solution densities at each temperature, compared with density at 25°C, for each concentration

[Gd Cl ₃] (molal)	Ratio of density at each temperature to the density at 25°C							
	5	10	15	20	25	30	35	40
0.0978	1.0032	1.0028	1.0021	1.0012	1.0000	0.9986	0.9970	0.9952
0.2368	1.0043	1.0037	1.0028	1.0016	1.0000	0.9980	0.9957	0.9929
0.4105	1.0045	1.0038	1.0028	1.0016	1.0000	0.9981	0.9960	0.9935
0.5894	1.0048	1.0039	1.0029	1.0015	1.0000	0.9982	0.9962	0.9939
0.7842	1.0050	1.0040	1.0029	1.0015	1.0000	0.9983	0.9964	0.9943
0.9947	1.0053	1.0042	1.0029	1.0015	1.0000	0.9983	0.9964	0.9944
1.2239	1.0055	1.0043	1.0030	1.0016	1.0000	0.9983	0.9965	0.9945

Table 5. Calculated densities of gadolinium nitrate solutions at various temperatures (from references 1 and 2)

[Gd (NO ₃) ₃] (molal)	Density of solution (g/mL) at each temperature (°C)							
	5	10	15	20	25	30	35	40
0.1532	1.0448	1.0443	1.0435	1.0424	1.0409	1.0391	1.0371	1.0347
0.2068	1.0605	1.0599	1.0589	1.0576	1.0559	1.0538	1.0513	1.0484
0.2947	1.0850	1.0843	1.0833	1.0819	1.0802	1.0781	1.0757	1.0729
0.4092	1.1163	1.1155	1.1144	1.1130	1.1113	1.1092	1.1068	1.1041
0.5991	1.1671	1.1661	1.1648	1.1633	1.1615	1.1594	1.1571	1.1544
0.7955	1.2179	1.2167	1.2153	1.2136	1.2118	1.2097	1.2074	1.2049
1.0108	1.2714	1.2701	1.2685	1.2667	1.2648	1.2626	1.2603	1.2577
1.2150	1.3207	1.3191	1.3174	1.3155	1.3135	1.3112	1.3088	1.3062

in Table 4. Where the gadolinium chloride concentrations were close to the gadolinium nitrate concentrations, the ratios in Table 4 were used directly; otherwise, interpolated values were used to calculate the densities of the gadolinium nitrate solutions.

Figure 2 shows a graph of the densities of gadolinium nitrate solutions at a range of concentrations and temperatures. The concentrations are plotted as lb Gd/ft³ of solution. The variation in density caused by the different temperatures is relatively small, with a maximum variation of 1.1% between 5°C and 40°C. A new paper was published in 2005⁴ that gave the density of gadolinium nitrate solutions over a limited range of concentrations and temperatures. The solutions used in these experiments contained a low concentration of nitric acid in addition to the gadolinium nitrate. The data from reference 4 is shown in Fig. 3 and Table 6. In Fig. 3, the densities for the lowest two concentrations of the data from Table 5, which overlap the new data in Table 6, are also plotted for comparison. The two sets of data match very well at the lower temperatures, but there is a slight difference at 40°C at the higher concentration. This new data did not extend to the higher concentrations that are used in the poison injection tank at HFIR, or down to the lower temperatures that are desired. Therefore, it is used as a comparison with the densities calculated using the gadolinium chloride temperature variation data, to validate the information in Table 5.

Table 7 shows a comparison of the predicted densities of gadolinium nitrate solutions at the same concentrations using the data from references 1 and 2, where the temperature variation is based on the gadolinium chloride solution data, and the data from reference 4. The new paper⁴ did not use exactly the same gadolinium nitrate concentrations at each temperature, so interpolation was used to get consistent data, at the same concentrations that were used for the original data from references 1 and 2, for comparison. The results show that there is a small difference in the predicted densities between the two data sets, particularly at the higher temperatures. The predictions using the data from references 1 and 2, which cover that range of concentrations and temperatures needed by HFIR, give

Fig. 2. Gadolinium nitrate solution densities, from references 1 and 2, at various concentrations and temperatures.

Table 6. Density of gadolinium nitrate solutions at various temperatures⁷

Concentration (molal)		Temperature (°C)	Density (g/mL)
Gd(NO ₃) ₃	HNO ₃		
0.01918	0.00037	15	1.004828
0.03873	0.00075	15	1.010576
0.05782	0.00111	15	1.016163
0.07778	0.00150	15	1.021992
0.09778	0.00188	15	1.027791
0.11819	0.00228	15	1.033684
0.13862	0.00267	15	1.039590
0.15900	0.00306	15	1.045392
0.18085	0.00348	15	1.051642
0.20228	0.00390	15	1.057810
0.01904	0.00037	25	1.002723
0.03814	0.00073	25	1.008284
0.05790	0.00111	25	1.014003
0.07749	0.00149	25	1.019642
0.09775	0.00188	25	1.025448
0.11793	0.00227	25	1.031202
0.13846	0.00267	25	1.037041
0.15997	0.00308	25	1.043140
0.18082	0.00348	25	1.049031
0.20228	0.00390	25	1.055063
0.01931	0.00037	40	0.997779
0.05811	0.00112	40	1.008876
0.07803	0.00150	40	1.014527
0.09801	0.00189	40	1.020169
0.11642	0.00224	40	1.025348
0.13779	0.00265	40	1.031339
0.15840	0.00305	40	1.037089
0.18068	0.00348	40	1.043292
0.20228	0.00390	40	1.049282
0.01931	0.00037	40	0.997779

Fig. 3. Gadolinium nitrate solution densities, published⁴ and calculated.^{1,2}

Table 7. Densities of gadolinium nitrate solutions

Gd(NO ₃) ₃ (molal)	Temp. (°C)	Density (g/mL)		Variation (%)
		Refs. 1 and 2	Ref. 7	
0.1532	15	1.0435	1.0437	0.02
0.1532	25	1.0409	1.0412	0.03
0.1532	40	1.0347	1.0356	0.09
0.2068	15	1.0589	1.0592	0.03
0.2068	25	1.0559	1.0565	0.06
0.2068	40	1.0484	1.0507	0.22

a slightly lower density at each concentration. The difference may be the result of the extra concentration of nitric acid in the solutions used for the reference 4 data, or just experimental measurement variations between the two sets of data. Since the density differences at 25°C, where the gadolinium nitrate solution densities were measured in reference 1, follow the same pattern as the data at the other temperatures, which were predicted from the gadolinium chloride data in reference 2, there is no evidence that using the temperature variations from the gadolinium chloride solutions is the cause of the differences.

The reference 4 data gives slightly higher gadolinium nitrate concentrations than the reference 1 and 2 data at each temperature and concentration. For a given measured density, the correlation from the reference 1 and 2 data (Table 5 and Fig. 2) would predict a lower gadolinium nitrate concentration than the reference 4 data. Therefore, using the correlation from the reference 1 and 2 data would give the lowest predicted gadolinium concentration for a given density measurement for the solution in the HFIR poison injection system, which would be conservative for the HFIR application and guarantee that the actual amount of gadolinium in the system is not less than that calculated.

The existing procedure for determining the amount of gadolinium in the HFIR poison injection system uses a graph of density vs concentration at one temperature, which is based on the original data at 15°C (Fig. 4). Using the data at the lower temperature predicts the lowest gadolinium concentration for a given measured density, which is conservative. The data in Fig. 4 has a built-in safety factor of about 10% compared with the actual data calculated from references 1 and 2. This safety factor helps guarantee that the actual amount of gadolinium in the poison injection system is higher than the calculated amount; however, it also means that the graph loses the direct tie to the published data. For the new extended temperature range, using the density vs concentration data for the lowest expected temperature will give the lowest amount of gadolinium for any measured density. Therefore, the density data at 5°C should be used for calculating the amount of gadolinium in the poison injection system, which will guarantee that the actual concentration is at least as high as the predicted concentration. Figure 5 shows a graph of solution density vs gadolinium concentration (as lb Gd/ft³ of solution) and also shows the linear regression correlation and R-squared value for this data.

Fig. 4. Graph of wt% gadolinium nitrate vs density, from HFIR procedure.

Fig 5. Graph of gadolinium concentration vs density at 5°C, from references 1 and 2.

The suggested new procedure for calculating the amount of gadolinium in the HFIR poison injection system is as follows.

- 1 Calculate the usable volume in the system
2. Measure the density of the solution
3. Calculate the gadolinium concentration using the following equation:

$$\text{Gd} \left(\frac{\text{lb}}{\text{ft}^3} \right) = \text{measured density} \left(\frac{\text{g}}{\text{mL}} \right) \times 34.681 - 34.785$$

or by reading the value from the Fig. 5

4. Calculate the amount of gadolinium in the system using the following equation:

$$\text{Amount of Gd (lb)} = \text{Gd concentration} \left(\frac{\text{lb}}{\text{ft}^3} \right) \times \text{usable volume} (\text{ft}^3)$$

If an additional safety factor is desired, it is recommended that an administrative control limit, which is higher than the required minimum amount of gadolinium, be set, rather than changing the graph in Fig. 5 or the corresponding equation.

For a typical density of 1.2 g/mL in the poison injection system, the equation above would give a gadolinium concentration of 6.83 lb/ft³. The graph in the HFIR procedure would give a concentration of 18.5 wt% = 6.24 lb Gd/ft³. Because of the safety factor built into the old graph, this result is about 8% lower than the actual concentration.

4. REFERENCES

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