

A Brief User's Guide to the Excel[®]-Based DF Calculator

Fuel Cycle Research & Development

***Prepared for
U.S. Department of Energy
Material Recovery and Waste Form
Development Campaign***

RT Jubin

30 September 2015

FCRD-MRWFD-2015-000627

ORNL-SPR-2015/493



DOCUMENT AVAILABILITY

Reports produced after January 1, 1996, are generally available free via US Department of Energy (DOE) SciTech Connect.

Website <http://www.osti.gov/scitech/>

Reports produced before January 1, 1996, may be purchased by members of the public from the following source:

National Technical Information Service
5285 Port Royal Road
Springfield, VA 22161
Telephone 703-605-6000 (1-800-553-6847)
TDD 703-487-4639
Fax 703-605-6900
E-mail info@ntis.gov
Website <http://www.ntis.gov/help/ordermethods.aspx>

Reports are available to DOE employees, DOE contractors, Energy Technology Data Exchange representatives, and International Nuclear Information System representatives from the following source:

Office of Scientific and Technical Information
PO Box 62
Oak Ridge, TN 37831
Telephone 865-576-8401
Fax 865-576-5728
E-mail reports@osti.gov
Website <http://www.osti.gov/contact.html>

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

SUMMARY

To understand the importance of capturing penetrating forms of iodine as well as the other volatile radionuclides, a calculation tool was developed in the form of an Excel[®] spreadsheet to estimate the overall plant decontamination factor (DF). The tool requires the user to estimate splits of the volatile radionuclides within the major portions of the reprocessing plant, speciation of iodine and individual DFs for each off-gas stream within the Used Nuclear Fuel reprocessing plant. The impact to the overall plant DF for each volatile radionuclide is then calculated by the tool based on the specific user choices. The Excel[®] spreadsheet tracks both elemental and penetrating forms of iodine separately and allows changes in the speciation of iodine at each processing step. It also tracks ³H, ¹⁴C and ⁸⁵Kr. This document provides a basic user's guide to the manipulation of this tool.

The DF Calculator uses two types of building blocks. The first is input blocks in which the user can insert information on the feed to the system, splits in various unit operations, and capture efficiencies for each stream of interest. The second class of blocks is the stream composition blocks or results blocks. Based on the user input, the quantity of each species reaching the stack is calculated, and an overall plant DF computed. Graphic output is also provided for each species in terms of the contribution of each of the five off-gas streams that contribute to the overall plant gaseous emissions.

CONTENTS

SUMMARY	iii
ABBREVIATIONS AND ACRONYMS	vi
1. INTRODUCTION	1
2. DESCRIPTION OF DF CALCULATOR	2
2.1 Feed Block	3
2.2 Head-end Split to the Cell Off-Gas	3
2.3 Head-End Cell Off-Gas DF.....	4
2.4 Dissolver Split to the Cell Off-Gas	5
2.5 Dissolver Off-Gas Split to the Cell Off-Gas	6
2.6 Dissolver Off-Gas DF	7
2.7 Dissolver Cell Off-Gas DF.....	8
2.8 Split in Solvent Extraction and Other Processes to the Vessel Off-Gas	8
2.9 Vessel Off-Gas DF	9
2.10 Split in Waste System to the Waste System Off-Gas.....	10
2.11 Waste System Off-Gas DF	11
2.12 Overall Plant Off-Gas DF	12
3. CONCLUSIONS	15
4. BIBLIOGRAPHY	17

FIGURES

Fig. 2-1. Typical off-gas streams that could be present in a notional UNF reprocessing plant.	2
Fig. 2-2. Input Block to DF Calculator.	3
Fig. 2-3. Head-end Split portion of DF Calculator.	4
Fig. 2-4. Head-end Off-Gas treatment portion of DF Calculator.	5
Fig. 2-5. Dissolver Split to Off-gas portion of DF Calculator.	6
Fig. 2-6. Dissolver Split to Off-gas and DOG abatement portion of DF Calculator.	7
Fig. 2-7. Dissolver Cell Off-Gas abatement portion of DF Calculator.	9
Fig. 2-8. Split to the Vessel Off-Gas portion of DF Calculator.	10
Fig. 2-9. Vessel Cell Off-Gas abatement portion of DF Calculator.	11
Fig. 2-10. Split to the Waste System Off-Gas portion of DF Calculator.	12
Fig. 2-11. Waste System Off-Gas abatement portion of DF Calculator.	13
Fig. 2-12. Vessel Cell Off-Gas abatement portion of DF Calculator.	14
Fig. 2-13. Distribution of iodine sources by major process stream.	14
Fig. 2-14. Distribution of complex iodine sources by major process stream.	15
Fig. 3-1. Overall layout of the DF Calculator Tool.	16

ABBREVIATIONS AND ACRONYMS

CH ₃ I	Methyl iodide
COG	Cell off-gas
DF	Decontamination factor
DOG	Dissolver off-gas
HOG	Head-end off-gas
SOG	Shear off-gas
UNF	Used nuclear fuel
VOG	Vessel off-gas
WOG	Waste solidification off-gas

A BRIEF USER'S GUIDE TO THE EXCEL[®]-BASED DF CALCULATOR

1. INTRODUCTION

Four radionuclides have been identified as being sufficiently volatile in reprocessing of nuclear fuel that their gaseous release needs to be controlled to meet U.S. regulatory requirements (Jubin et al. 2011, 2012). These radionuclides are ³H, ¹⁴C, ⁸⁵Kr, and ¹²⁹I. Of these, ¹²⁹I has the longest half-life and greatest potential biological impact. Accordingly, control of the release of ¹²⁹I is most critical with respect to U.S. regulations for the release of radioactive material in stack emissions. Current U.S. Environmental Protection Agency regulations governing nuclear facilities (40 CFR 190) state that the total quantity of radioactive materials entering the general environment from the entire uranium fuel cycle, per gigawatt-year of electrical energy produced by the fuel cycle, must contain less than 5 mCi of ¹²⁹I.

In a previous document, the needs for treating off-gas streams from the reprocessing of used nuclear fuel (UNF) and the regulations that set the limits for releases from such a facility were discussed (Jubin et al. 2012). These regulations ultimately dictate the decontamination factor (DF) that must be met in the facility. In a subsequent report (Jubin et al. 2013), the role of off-gas pathways was examined since the gaseous radionuclides can partition to various different reprocessing off-gas streams. Based on this report and subsequent analysis (Jubin, et al. 2015), the DF for ¹²⁹I could be 1000 to 3000. The required DF for ³H could be as high as 200, depending on the age of the fuel processed. The DF for ⁸⁵Kr could be up to ~35, also depending on fuel age. The required DF for ¹⁴C is 1 in many cases (no treatment required) but could be higher in specific cases.

Iodine removal has always been primarily focused on the head-end off-gas operations. However, in light of the estimated iodine emissions control efficiencies of up to 99.9% or higher needed to meet U.S. regulatory requirements, the control of iodine in the dissolver off-gas (DOG) stream is insufficient. Operating experience and tests with actual nuclear fuel show that iodine evolution from the dissolver solution into the dissolver off-gas ranges between 95 and 99%. The 2013 study identified the major iodine release pathways that would require treatment to effectively control the release of iodine to the stack:

- DOG, including the off-gas from the fuel shearing operation;
- Vessel off-gas (VOG), including the vents from the process operations and tanks within the facility;
- Off-gas from liquid waste solidification (WOG), including the production of waste forms from the primary off-gas system traps. Also included in this stream would be the treatment activities on used solvent and on the solidification of discarded used solvent. Vents from the waste storage tanks in this section of the plant must also be connected to an iodine trap system;
- Dissolver Cell off-gas (COG).

A recent report by Bruffey et al. (2015) points out that the study of inorganic iodide in off-gas systems has been almost exclusively limited to I₂, and the focus of organic iodide studies has been CH₃I. This report specially focused on those iodine species that have the potential to be poorly sequestered with traditional capture methodologies. These forms have historically been known as *penetrating* iodine-bearing species, and if they are, in-fact, significantly more difficult to trap and retain, the overall plant DF could be adversely affected.

2. DESCRIPTION OF DF CALCULATOR

To understand the importance of the distribution of the volatile radionuclides among the various off-gas streams within a UNF Reprocessing Plant and the impacts of the treatment capabilities employed on each stream on the overall plant DF, a spreadsheet tool was developed using Excel[®]. The Excel[®] spreadsheet tracks both elemental and penetrating forms of iodine separately and allows the user to input an estimate of the speciation of iodine at each processing step. The spreadsheet does not have an internal capability to estimate the speciation of iodine at each processing step, i.e., this is solely a user input. It also tracks ³H, ¹⁴C and ⁸⁵Kr. Figure 2-1 shows the expected pathways within a reprocessing facility considered by the DF Calculator.

The DF Calculator uses two types of building blocks. The first type is an input block in which the user enters one of three categories of information required by the DF Calculator. The first category of information is the specification of the feed to the system. The second category is information about splits in various unit operations. The third category of information that is input is specification of the capture efficiencies for each stream and component of interest. These input blocks are indicated by the blue title block (see Fig. 2-2). Entries can be made for each of the five species of interest: Iodine as elemental iodine, iodine as a complex species, krypton, Carbon-14, and tritium. The second class of blocks is the stream composition block or results block. These blocks are designated by the green title block, and the user should never enter any data into these output blocks because these are all calculated values. The following sub-sections will describe each of the blocks in more detail.

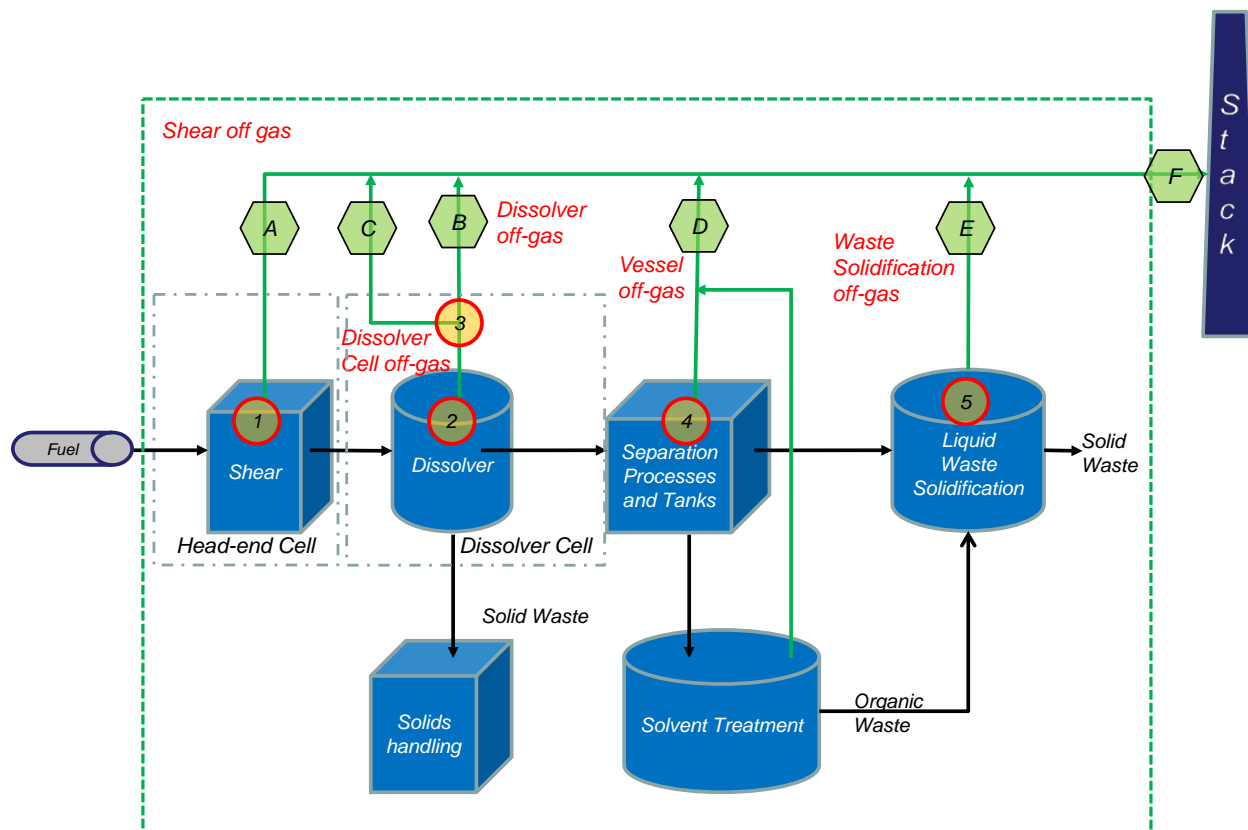


Fig. 2-1. Typical off-gas streams that could be present in a notional UNF reprocessing plant.

2.1 Feed Block

The feed block is shown in Fig. 2-2. The values in this block are the starting point for all calculations. The DF calculator as designed and implemented is “units neutral” in that it does not show or convert units on individual species that are input. Whatever the user determines as a useful unit of measure associated with the numeric input value will be assumed for all subsequent calculations without any units conversion. As an example the user could choose to use a value of 100 for each of the species of interest as a stand in for percentage, and all of the output block should then be interpreted to be a percentage of the species fed to the facility. Similarly, the user could input the actual grams or Curies of an element per tonne of fuel processed and the results would be in the same units.

One item of particular note is the Complex Iodine value in the Fuel Block. This should be left blank for the fuel fed to the reprocessing plant. The complex iodine species are assumed to be formed in the dissolver and/or subsequent steps.

Fuel	
Iodine (I2)	100
Complex Iodine	
Kr	1000
C-14	1000
Tritium	1000

Fig. 2-2. Input Block to DF Calculator.

2.2 Head-end Split to the Cell Off-Gas

This block is shown in Fig. 2-3. The number “1” in the blue title block associates this block with the shear block also labeled “1” in Fig. 2-1. In this block, the user specifies the percentage of the stream entering the block (the Fuel Block) that is split off to the Head-end Cell Off-Gas Stream (the results block directly above the “Head-end split to the Cell Off-Gas” block). In this example, the user has estimated that 0.01% of the iodine and 5% of the Kr will report to the Head-end Cell Off-Gas stream, and the results are shown in that block. The remaining volatile radionuclides will remain in the fuel and are shown in the results block to the right, the “Fuel to Dissolver. This block will be discussed in Sect. 2.4.

		Head-end Cell Off-gas			
		Iodine	0.01		
		Complex Iodine	0.00		
		Kr	50.00		
		C-14	20.00		
		Tritium	1.00		
Fuel		1. Head-end split to Cell Off-gas		Fuel to Dissolver	
Iodine (I2)	100	Iodine (%)	0.01%	Iodine	99.99
Complex Iodine		I converted & volatilized (%)	0.00%		
Kr	1000	Kr (%)	5.00%	Kr	950
C-14	1000	C-14 (%)	2.00%	C-14	980
Tritium	1000	Tritium (%)	0.10%	Tritium	999

Fig. 2-3. Head-end Split portion of DF Calculator.

2.3 Head-End Cell Off-Gas DF

This block is shown in Fig. 2-4. The letter “A” in the blue title block associates this block with the location of a potential abatement system on the off-gas line from the shear block also labeled “A” in Fig. 2-1. In this input block, the individual DFs for each species of interest are entered. The DF is a ratio of the amount of a species entering the abatement system divided by the amount of the same species in the effluent from the system. Thus a DF of 100 indicates that 99% of the species is removed by the abatement system. The results block located above the Head-End Cell Off-Gas DF block shows the impact of the abatement system on the stream entering (the results block below). In this example no abatement is assumed (that is, a DF of 1), and the head-end off-gas in and out values are the same. This results block will be summed in the Treated Plant Off-gas block as part of the Overall Plant DF calculations discussed in Sect. 2.12.

This stream could physically represent several streams in a reprocessing plant. It could represent the off-gas from the shear itself, the shear off-gas (SOG), or it could be the leakage from the shear to the head-end cell. Whichever stream the user chooses to represent is left to the user’s discretion. The user simply inputs the appropriate splits and extent of abatement employed.

Emissions from Treated Head-end Cell Off-gas	
Iodine	0.01
Complex Iodine	0
Kr	50
C-14	20
Tritium	1
A. Head-end Cell Off-gas DF	
Iodine (DF)	1
Complex Iodine (DF)	1
Kr (DF)	1
C-14 (DF)	1
Tritium (DF)	1
Head-end Cell Off-gas	
Iodine	0.01
Complex Iodine	0.00
Kr	50.00
C-14	20.00
Tritium	1.00

Fig. 2-4. Head-end Off-Gas treatment portion of DF Calculator.

2.4 Dissolver Split to the Cell Off-Gas

This block is shown in Fig. 2-5. The number “2” in the blue title block associates this block with the dissolver block also labeled “2” in Fig. 2-1. In this block, the user specifies the percentage of the stream entering the block (the Fuel to Dissolver Block) that is volatilized in the dissolver and is split off to the “Volatilized in Dissolver” results block located directly above the “Dissolver Split to Off-gas” block. This split to be volatilized is entered as the percentage of the species in the input to this block. Note that this is not the percentage of the total amount entering the plant in the Fuel Block. In this example, 96% of the iodine and 99% of the Kr will be volatilized along with 96% of the ¹⁴C and 1% of the tritium.

In the dissolver, a portion of the iodine may also react to form more complex iodine species that in turn may be more difficult to trap. To address this case, the user can specify a percentage of the iodine that is volatilized to be “converted” to this complex iodine form. The sum of the two iodine forms is 96% of the 99.99 iodine units that enter the dissolver or 95.99 iodine units as shown in the upper results box. The remaining iodine and other volatile radionuclides progress to the Fuel to SX results block. This will be discussed in Sect. 2.8.

		Volatilized in Dissolver	
		Iodine	91.19
		Complex Iodine	4.80
		Kr	949.05
		C-14	940.80
		Tritium	9.99
Fuel to Dissolver		2. Dissolver Split to Off-Gas*	
Iodine	99.99	Iodine (%)	96.00%
		I converted & volatilized (%)	5.00%
Kr	950	Kr (%)	99.90%
C-14	980	C-14 (%)	96.00%
Tritium	999	Tritium (%)	1.00%

Fig. 2-5. Dissolver Split to Off-gas portion of DF Calculator.

2.5 Dissolver Off-Gas Split to the Cell Off-Gas

This block is shown in Fig. 2-6. The number “3” in the blue title block associates this block with the dissolver off-gas split that is labeled “3” in Fig. 2-1. In this block, the user specifies the percentage of the species volatilized in the dissolver that leaks from the dissolver into the dissolver cell. This leakage is assumed to be through seals, etc., and may be different for each species. In this example, 0.07% of the iodine and 2% of the Kr, ¹⁴C and tritium will be “lost” to the dissolver cell. This stream is shown coming out of the right side of the Dissolver Off-Gas split to the Cell Off-Gas block. The balance of the stream volatilized in the dissolver is shown in the Dissolver Off-gas results block.

Emissions from Treated Dissolver Off-gas			
Iodine	0.009112705		
Complex Iodine	0.00239808		
Kr	930.069		
C-14	92.1984		
Tritium	9.7902		
B. Dissolver Off-gas DF		Dissolver Off-gas	
Iodine (DF)	10000	Iodine	91.13
Complex Iodine (DF)	2000	Complex Iodine	4.80
Kr (DF)	1	Kr	930.07
C-14 (DF)	10	C-14	921.98
Tritium (DF)	1	Tritium	9.79
Volatilized in Dissolver		3. Dissolver Off-gas Split to Cell Off-Gas**	
Iodine	91.19	Iodine (%)	0.07%
Complex Iodine	4.80		
Kr	949.05	Kr (%)	2.00%
C-14	940.80	C-14 (%)	2.00%
Tritium	9.99	Tritium (%)	2.00%

Fig. 2-6. Dissolver Split to Off-gas and DOG abatement portion of DF Calculator.

2.6 Dissolver Off-Gas DF

This block is shown in Fig. 2-6. The letter “B” in the blue title block associates this block with the location of a DOG abatement system on the DOG line from the Dissolver Off-gas stream block also labeled “B” in Fig. 2-1. In this Input block, the individual DFs for each species of interest are entered. The results block above the Dissolver Off-Gas DF shows the impact of the abatement system on the stream entering (the results block shown to the right of the Dissolver Off-gas DF input block). In this example, no abatement is assumed (that is, a DF of 1) for Kr and tritium in the DOG abatement system, and the Dissolver Off-Gas in and out values are the same for these species. A DF of 10 is shown for ¹⁴C. An iodine DF of 10,000 is shown for iodine and 2,000 for complex iodine species. The ability to have different DFs for the two different forms of iodine allows the user to explore the impacts of more penetrating iodine species. The results of the DFs applied to the Dissolver Off-gas stream are shown in the results block labeled “Emissions from treated Dissolver Off-gas” using the same unit as in the Feed

block for each of the species. This results block will be summed in the Treated Plant Off-gas block as part of the Overall Plant DF calculations discussed in Sect. 2.12.

2.7 Dissolver Cell Off-Gas DF

This block is shown in Fig. 2-7. The letter “C” in the blue title block associates this block with the location of a potential abatement system on the Dissolver Cell off-gas line from the split from the dissolver cell block also labeled “C” in Fig. 2-1. In this input block, as with previous DF input blocks, the individual DFs for each species of interest are entered. The results block above the Dissolver Off-gas Cell Off-gas DF shows the impact of the abatement system on the stream entering, i.e., the Dissolver Cell Off-gas results block below the input block. In this example no abatement is assumed (that is, a DF of 1) on the dissolver cell off-gas and the in and out values are the same. This results block will be summed in the Treated Plant Off-gas block as part of the Overall Plant DF calculations discussed in Sect. 2.12.

2.8 Split in Solvent Extraction and Other Processes to the Vessel Off-Gas

This block is shown in Fig. 2-8. The number “4” in the blue title block associates this block with a split to the off-gas stream arising from the solvent extraction systems and vessels downstream of the dissolver also labeled “4” in Fig. 2-1. This block is also assumed to include the off-gas from the solvent treatment portion of the reprocessing plant. In this block, the user specifies the percentage of the stream entering the block (the Fuel to SX stream from the Dissolver Block, see Sect. 2.5) that is volatilized in the process vessels and tanks. These splits are entered as the percentage input to this block that is volatilized. In this example, 50% of the iodine and 99.99% of the Kr will be volatilized along with 10% of the ¹⁴C and 1% of the tritium.

In the process vessels and tanks, a portion of the iodine may react to form more complex iodine species that may be more difficult to trap. To address this case, the user can specify the percentage of the volatilized iodine that is “converted” to this alternate form. In this example, the sum of the two iodine forms is 50% of the 4.00 iodine units that enter the solvent extraction/tankage portion of the plant or 1.00 units of iodine in an easy to trap form and 1.00 iodine units as a complex form shown in the upper results box labeled “Vessel Off-Gas.” The remaining iodine and other “volatile” radionuclides progress to the Waste Systems. This will be discussed in Sect. 2.10.

		Emissions from Treated Dissolver Cell Off-gas	
		Iodine	0.063834
		Complex Iodine	0.00336
		Kr	18.981
		C-14	18.816
		Tritium	0.1998
Dissolver Off-gas		C. Dissolver Cell Off-gas DF	
Iodine	91.13	Iodine (DF)	1
Complex Iodine	4.80	Complex Iodine (DF)	1
Kr	930.07	Kr (DF)	1
C-14	921.98	C-14 (DF)	1
Tritium	9.79	Tritium (DF)	1
3. Dissolver Off-gas Split to Cell Off-Gas**		Dissolver Cell Off-gas	
Iodine (%)	0.07%	Iodine	0.06
		Complex Iodine	0.00
Kr (%)	2.00%	Kr	18.98
C-14 (%)	2.00%	C-14	18.82
Tritium (%)	2.00%	Tritium	0.20

Fig. 2-7. Dissolver Cell Off-Gas abatement portion of DF Calculator.

2.9 Vessel Off-Gas DF

This block is shown in Fig. 2-9. The letter “D” in the blue title block associates this block with the location of a potential abatement system on the VOG line from separation process labeled “D” in Fig. 2-1. In this input block, as with previous DF input blocks, the individual DFs for each species of interest are entered. The results block above the Vessel Off-gas DF shows the impact of the abatement system on the stream entering (the results block below). In this example, no abatement is assumed (that is, a DF of 1) for Kr and tritium in the DOG abatement system, and the Dissolver Off-Gas in and out values are the same for these species. A DF of 10 is shown for ¹⁴C. An iodine DF of 100 is shown for iodine and 20 for complex iodine species. The results of the DFs applied to the Vessel Off-gas stream are shown results block labeled “Emissions from treated Vessel Off-gas” using the same units as in the Feed block for each of the species. This results block will be summed in the Treated Plant Off-gas block as part of the Overall Plant DF calculations discussed in Sect. 2.12.

		Vessel Off-gas			
		Iodine	1.00		
		Complex Iodine	1.00		
		Kr	0.95		
		C-14	3.92		
		Tritium	9.89		
Fuel to SX		4. SX Split to Off-Gas		To Waste Systems	
Iodine	4.00	Iodine (%)	50.00%	Iodine	2.00
		I converted & volatilized (%)	50.00%		
Kr	0.95	Kr (%)	99.99%	Kr	9.5E-05
C-14	39.2	C-14 (%)	10.00%	C-14	35.28
Tritium	989.01	Tritium (%)	1.00%	Tritium	979.1199

Fig. 2-8. Split to the Vessel Off-Gas portion of DF Calculator.

2.10 Split in Waste System to the Waste System Off-Gas

This block is shown in Fig. 2-10. The number 5 in the blue title block associates this block with a split to the WOG stream arising from the waste treatment systems downstream of the main processes also labeled 5 in Fig. 2-1. In this block, the user specifies the percentage of the stream entering the block (To Waste System stream from the SX split to Off-gas Block, see Sect. 2.8) that is volatilized during waste processing operations. These splits are entered as the percentage of the species input to this block that are volatilized. In this example, 80% of the iodine and 100% of the Kr, ¹⁴C, and tritium will be volatilized.

During the waste treatment processing, a portion of the iodine may react to form more complex iodine species that may be more difficult to trap. To address this case, the user can specify a percentage of the volatilized iodine that is “converted” to this alternate form. In this example, 10% of the total iodine volatilized is in a complex form. In this example, the sum of the two iodine forms is 80% of the 2.00 iodine units that enter the waste treatment portion of the plant, or 1.44 units of iodine will be found as an easy-to-trap form and 0.16 iodine units as a complex form. This is shown in the upper results box labeled Waste System Off-Gas. The remaining iodine and other “volatile” radionuclides are assumed to be retained in the Waste as shown in the “retained in Waste” results box. The portion retained in the waste does not enter into the denominator of the calculation of the overall plant DF. This will be discussed further in Sect. 2.12.

Emissions from Treated Vessel Off-gas	
Iodine	0.009999
Complex Iodine	0.049995
Kr	0.949905
C-14	0.392
Tritium	9.8901
D. Vessel Off-gas DF	
Iodine (DF)	100
Complex Iodine (DF)	20
Kr (DF)	1
C-14 (DF)	10
Tritium (DF)	1
Vessel Off-gas	
Iodine	1.00
Complex Iodine	1.00
Kr	0.95
C-14	3.92
Tritium	9.89

Fig. 2-9. Vessel Cell Off-Gas abatement portion of DF Calculator.

2.11 Waste System Off-Gas DF

This block is shown in Fig. 2-11. The letter “E” in the blue title block associates this block with the location of a potential abatement system on the Waste System off-gas line from the split from the Waste systems block also labeled “E” in Fig. 2-1. In this Input block, as with previous DF input blocks, the individual DFs for each species of interest are entered. The results block above the Waste Systems Off-gas DF shows the impact of the abatement system on the input stream (the results block below). In this example, no abatement is assumed (that is, a DF of 1) for Kr and tritium in the DOG abatement system, and the Dissolver Off-Gas in and out values are the same for these species. A DF of 10 is shown for ¹⁴C. An iodine DF of 100 is shown for iodine and 20 for complex iodine species. The results of the DFs applied to the Waste System Off-gas stream are shown in the results block labeled “Emissions from treated Waste System Off-gas” using the same unit as in the initial Fuel block for each of the species.

This results block will be summed in the Treated Plant Off-gas block as part of the Overall Plant DF calculations discussed in Sect. 2.12.

		Waste System Off-gas			
		Iodine	1.44		
		Complex Iodine	0.16		
		Kr	0.00		
		C-14	35.28		
		Tritium	979.12		
To Waste Systems		5. Waste Systems Split to Off-Gas		Retained in Waste	
Iodine	2.00	Iodine (%)	80.00%	Iodine	0.40
		I converted & volatilized (%)	10.00%		
Kr	9.5E-05	Kr (%)	100.00%	Kr	0
C-14	35.28	C-14 (%)	100.00%	C-14	0
Tritium	979.1199	Tritium (%)	100.00%	Tritium	0

Fig. 2-10. Split to the Waste System Off-Gas portion of DF Calculator.

2.12 Overall Plant Off-Gas DF

This block is shown in Fig. 2-12. The letter “F” in the green title block associates this block with the Reprocessing Plant Stack also labeled “F” in Fig. 2-1. This portion of the DF Calculator is composed of two results blocks.

The first results block is the Treated Plant Off-gas block which sums the five previously discussed effluent streams:

- 1) Emissions from treated Head-end Cell Off-gas (Sect. 2.3),
- 2) Emissions from treated Dissolver Off-gas (Sect. 2.5),
- 3) Emissions from treated Dissolver Cell Off-gas (Sect. 2.3),
- 4) Emissions from treated Vessel Off-gas (Sect. 2.3), and
- 5) Emissions from treated Waste treatment Off-gas (Sect. 2.11).

The amount remaining in the off-gas at the stack is reported in the same units for each species as was used to describe the feed to the plant. A graphical representation of the contribution to the total iodine released at the stack is provided for iodine in two charts that are found on the “Iodine Plots” tab. The first chart shows the percent contribution to the total iodine released at the stack from each of the five streams (Fig. 2-13). Each stream is a sum of the iodine and complex iodine values for that individual stream. The second chart shows the distribution of the complex iodine among the five streams (Fig. 2-14.) Charts for tritium, ¹⁴C and Kr are also provided on their respective tabs. For these three species, only one chart is provided since only one species for each radionuclide is tracked.

The second results block is the Overall Plant Off-gas DF block that calculates, as the name implies, the DF for the plant off-gas as a ratio of the quantity of each volatile radionuclide species entering the plant

divided by the quantity exiting from the stack. The quantities retained in the five potential abatement systems and the quantity retained in the waste are assumed to be held in such a manner that they would not be subject to subsequent release from the stack. In this example, the total iodine DF is 585. The Kr and tritium DFs are both 1 as would be expected because no abatement was applied to either radionuclide and none was retained in the waste. The plant DF for ¹⁴C was ~7, with the bulk of the carbon abatement occurring in the DOG abatement system.

Emissions from Treated Waste Off-gas	
Iodine	0.014399
Complex Iodine	0.007999
Kr	9.5E-05
C-14	3.528
Tritium	979.1199
E. Waste Systems Off-gas DF	
Iodine (DF)	100
Complex Iodine (DF)	20
Kr (DF)	1
C-14 (DF)	10
Tritium (DF)	1
Waste System Off-gas	
Iodine	1.44
Complex Iodine	0.16
Kr	0.00
C-14	35.28
Tritium	979.12

Fig. 2-11. Waste System Off-Gas abatement portion of DF Calculator.

Treated Plant Off-gas		F. Overall Plant Off-gas DF	
Iodine	0.107344	Iodine	584.4678
Complex Iodine	0.063752		
Kr	1000	Kr	1
C-14	134.9344	C-14	7.411009
Tritium	1000	Tritium	1

Fig. 2-12. Vessel Cell Off-Gas abatement portion of DF Calculator.

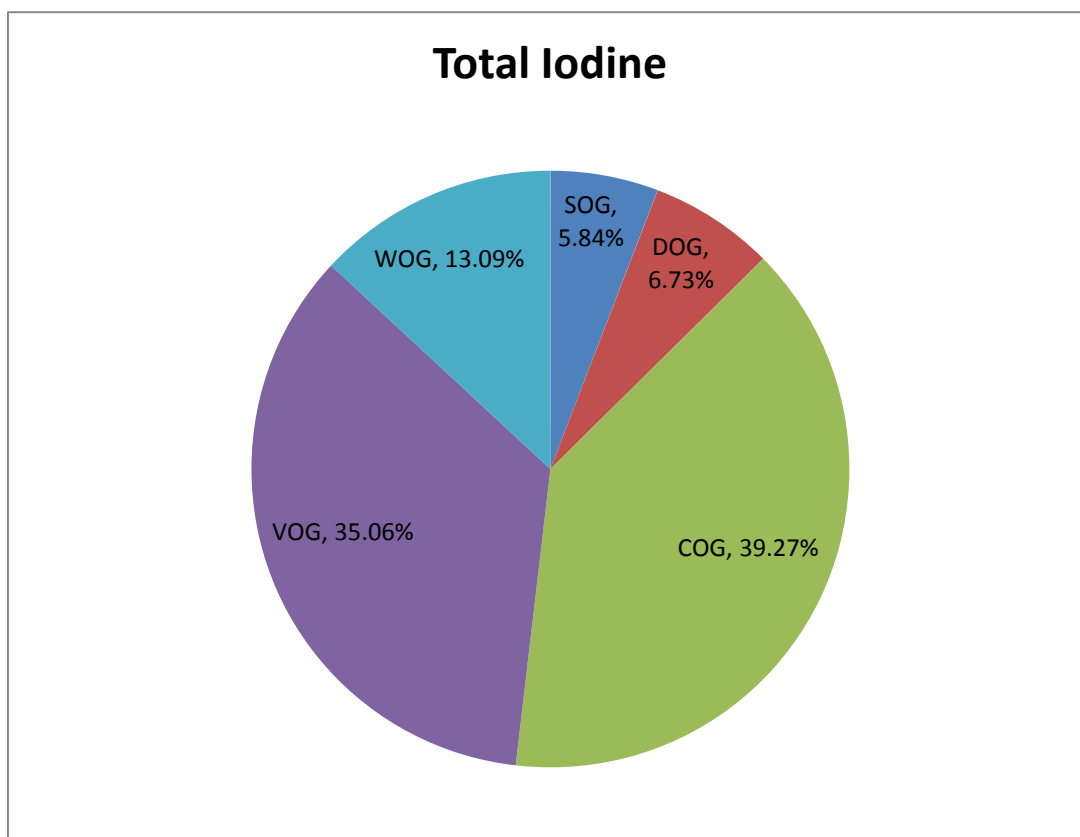


Fig. 2-13. Distribution of iodine sources by major process stream.

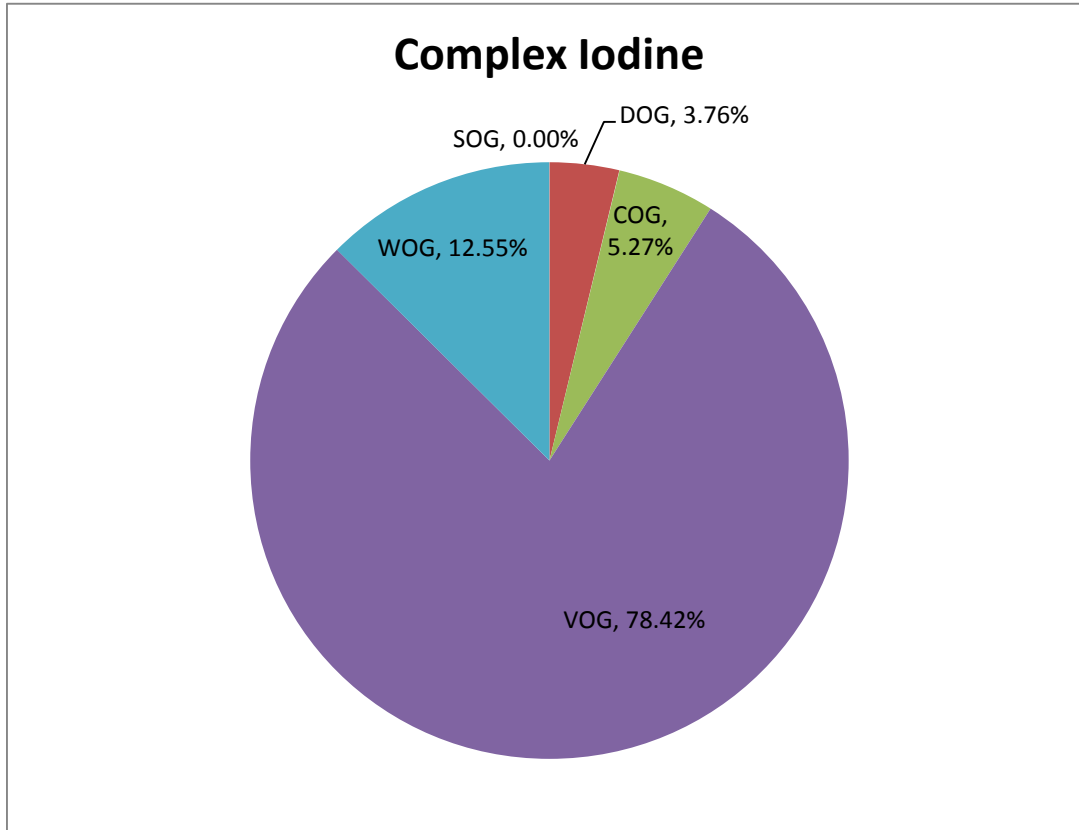


Fig. 2-14. Distribution of complex iodine sources by major process stream.

3. CONCLUSIONS

To understand the importance of capturing penetrating forms of iodine as well as the other volatile radionuclides, a calculation tool was developed in the form of an Excel[®] spreadsheet to estimate the overall plant decontamination factors (DFs). The tool requires the user to estimate splits of the volatile radionuclides within the major portions of the reprocessing plant, speciation of iodine and individual DFs for each off-gas stream within the Used Nuclear Fuel reprocessing plant. The impact to the overall plant DF for each volatile radionuclide is then calculated by the tool based on the specific user choices. The Excel[®] spreadsheet tracks both elemental and penetrating forms of iodine separately and allows changes in the speciation of iodine at each processing step. It also tracks ³H, ¹⁴C and ⁸⁵Kr. Figure 3-1 shows the overall layout of this tool. This document provides a basic users guide to the manipulation of this tool.

The DF Calculator uses two types of building blocks. The first is input blocks in which the user can insert information about the feed to the system, splits in various unit operations, and capture efficiencies for each stream of interest. These blocks are indicated by the blue title block (see Fig. 3-1). The second class of blocks comprises the stream composition blocks or results blocks. These are designated by the green title block. Based on the user input, the quantity of each species reaching the stack is calculated, and an effective plant DF commuted. Graphic output is also provided for each species regarding the contribution of each of the five off-gas streams that contribute to the overall plant gaseous emissions.



Fig. 3-1. Overall layout of the DF Calculator Tool.

4. BIBLIOGRAPHY

Bruffey, S.H., B.B. Spencer, D. M. Strachan, R. T. Jubin, N. Soelberg, and B. J. Riley. 2015. *A Literature Survey to Identify Potentially Problematic Volatile Iodine-Bearing Species Present in Off-gas Streams*. Report No. FCRD-MRWFD-2015-000421, ORNL-SPR-2015/290, INL/EXT-15-35609, UT-Battelle, LLC, Oak Ridge National Laboratory.

Jubin, R., N. Soelberg, D. Strachan, and G. Ilas. 2011. *Assessments and Options for Removal and Immobilization of Volatile Radionuclides from the Processing of Used Nuclear Fuel*. Report No. FCR&D-SWF-2011-000305, UT-Battelle, LLC, Oak Ridge National Laboratory.

Jubin, R., N. Soelberg, D. Strachan, and G. Ilas. 2012. *Fuel Age Impacts on Gaseous Fission Product Capture During Separations*. Report No. FCRD-SWF-2012-000089, PNNL-22550, UT-Battelle, LLC, Oak Ridge National Laboratory.

Jubin, R., N. Soelberg, and D. Strachan. 2013. *Iodine Pathways and Off-Gas Stream Characteristics for Aqueous Reprocessing Plants – A Literature Survey and Assessment*. Report No. FCRD-SWF-2013-000308 (ORNL/LTR-2013/383, INL/EXT-13-30119, PNNL-22885), UT-Battelle, LLC, Oak Ridge National Laboratory.

Jubin, R. T., S. H. Bruffey, and B. B. Spencer. 2015. "Performance of Silver Exchanged Mordenite for Iodine Capture Under Vessel Off-Gas Conditions." Presented at Global 2015, Paris, France.