2. ☐ Analysis   Check all that apply

Type of Analysis	☐ Engineering
	☐ Performance Assessment
	☐ Scientific

Intended Use of Analysis	☐ Input to Calculation
	☐ Input to another Analysis or Model
	☐ Input to Technical Document
	☐ Input to other Technical Products

Describe use:

3. ☒ Model   Check all that apply

Type of Model	☐ Conceptual Model	☒ Abstraction Model
	☐ Mathematical Model	☐ System Model
	☐ Process Model	

Intended Use of Model	☒ Input to Calculation
	☒ Input to another Model or Analysis
	☒ Input to Technical Document
	☒ Input to other Technical Products

Describe use:

Abstraction of EBS colloid process models for use in TSPA Calculations.

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10.	Lead/Supervisor	Robert J. MacKinnon	Robert MacKinnon	4/14/00
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FIGURES

TABLES

ACRONYMS AND ABBREVIATIONS

AMR	Analysis/Model Report
ANL	Argonne National Laboratory
CLST	Container Life and Source Term
CRWMS	Civilian Radioactive Waste Management System
CSNF	commercial spent nuclear fuel
DHLW	defense high-level radioactive waste
DOE	U.S. Department of Energy
DSNF	DOE-owned spent nuclear fuel
EBS	engineered barrier system
eq/L	equivalence (equivalents/liter)
EPA	Environmental Protection Agency
HLW	high-level radioactive waste
IRSR	Issue Resolution Status Report
KTI	Key Technical Issues
LANL	Los Alamos National Laboratory
QAP	Quality Assurance Plan
M	molarity (moles/liter)
m	molality (moles/kilogram)
MIC	microbially influenced corrosion
M&O	Management and Operations
NRC	Nuclear Regulatory Commission
REE	rare earth element
RN	radionuclide
SA	surface area
SNF	spent nuclear fuel
SRL	Savannah River Laboratory
TSPAI	Total System Performance Assessment and Integration
TSPA-SR	Total System Performance Assessment-Site Recommendation
TSPA-VA	Total System Performance Assessment-Viability Assessment
VA	Viability Assessment
YMP	Yucca Mountain Project
ZPC	zero point of charge

1. PURPOSE

As directed by a written development plan (CRWMS M&O 1999a), specifically Task 7, an abstraction for colloids in the engineered barrier system (EBS) is to be developed. The purpose of this abstraction is to assist Performance Assessment Operations (PAO) and its Engineered Barrier Performance Department in modeling occurrence and behavior of colloids in the geochemical environment within a repository drift, thus allowing PAO to provide a more detailed and complete in-drift geochemical model abstraction and to address the key technical issues (KTI) raised in the general acceptance criteria set out in the U.S. Nuclear Regulatory Commission (NRC) Total System Performance Assessment and Integration (TSPAI) Issue Resolution Status Report (IRSR) (NRC 1998a), as well as the NRC IRSR for the Evolution of the Near-Field Environment (NFE) Revision 2 (NRC 1999a) and Container Lifetime and Source Term (CLST) Revision 2 (NRC 1999b).

The scope of this Analysis/Model Report (AMR) is to provide the conceptual framework for the abstraction of models developed to describe the occurrence and behavior of colloids in the waste package and EBS/drift environments. The models have been derived from data developed primarily by Argonne National Laboratory (ANL) and Los Alamos National Laboratory (LANL) for the Total System Performance-Site Recommendation (TSPA-SR) calculations performed with the computer code GoldSim (Golder 2000). The models describe the calculated type and concentration of colloids generated in the waste package from degradation of the waste ("waste form colloids") and corrosion of the waste package ("corrosion" or "iron (hydr)oxide colloids"), and associated radionuclides; the types and potential role of colloids produced from the introduction of man-made materials during repository construction and operations; and the observed types and concentrations of colloids present in groundwater ("groundwater colloids") in the vicinity of Yucca Mountain. In addition, attachment/detachment and transport characteristics and mechanisms of colloids anticipated in the repository are discussed, including waste form colloids, colloids formed from man-made materials introduced into the repository, and groundwater colloids. The abstraction of the process models is intended to capture the most important characteristics of radionuclide-colloid behavior for use in predicting the potential impact on repository performance of colloid-facilitated radionuclide transport.

Quantities of colloids could be mobilized primarily as a result of alteration of the defense high-level radioactive waste (DHLW) and to a lesser extent the spent nuclear fuel (SNF) waste forms. The abundance of colloids produced within the waste package and introduced into the EBS/drift, the colloids entering the drift with ground water, and colloids resulting from introduction and/or degradation of introduced materials, as well as their stability over some period of time, will depend on several factors. First is the extent of waste form alteration and the alteration products formed. A second category includes such environmental factors as ionic strength, pH, cation concentrations, colloid concentration in groundwater, colloid content of groundwater entering the waste package from the drift, the presence of fulvic and humic acids, and microbes or microbe fragments. Suspended colloids may subsequently flocculate, be chemically or mechanically filtered, or dissolved. If the environmental factors change, colloid-sized particles may be peptized. In addition, colloids may sorb readily at the interfaces between air and water in rocks and engineered barriers and, depending upon the degree of saturation and the configuration of

the porous medium, may be retarded, even immobilized, or transported. These issues are relevant to colloid transport, which is of primary concern within the engineered barrier system in the drift and in the near- and far-field.

In this report three sources of colloids in the EBS/drift are considered: (1) the "colloid source term," defined as the sum of radionuclides associated with colloids that are released from a failed waste package; (2) groundwater colloids; and (3) colloids generated as a result of the introduction and degradation of construction materials and repository structures. The modeling and abstraction are confined to these three sources.

Colloidal systems are defined here as those in which the component phases are dispersed in an aqueous medium (groundwater, leachate), ranging in size from 1 nm to 1 um, approximately. Much has been learned about the environmental behavior of colloids over the last few decades, but due largely to their small size and unique surface-dominated chemical and physical characteristics, they have proven to be difficult to study. Therefore the approach taken in this analysis was to bound the probable behavior of colloid-associated radionuclides. The lack of certain data can be mitigated as well by comparing these bounding predictions with relevant laboratory and field studies and with "analog sites," where environmental conditions are analogous in critical respects to conditions used in experiments and anticipated in the repository. These approaches have been used to the extent possible to reduce the uncertainties associated with predictions of colloid-associated radionuclide concentrations, stability, and transport characteristics.

2. QUALITY ASSURANCE

The Quality Assurance (QA) program applies to this AMR. The Performance Assessment Operations responsible manager has evaluated the technical document activity in accordance with QAP-2-0 *Conduct of Activities*. The QAP-2-0 activity evaluation, *Conduct of Performance Assessment* (CRWMS M&O 1999b), determined that this activity is subject to the *Quality Assurance Requirements and Description* (QARD) DOE/RW-0333P (DOE 2000) requirements. Preparation of this analysis did not require the classification of items in accordance with QAP-2-3, *Classification of Permanent Items*. This activity is not a field activity. Therefore, an evaluation in accordance with NLP-2-0, *Determination of Importance Evaluations* was not required. This AMR was developed using AP-3.10Q, *Analyses and Models*.

3. COMPUTER SOFTWARE AND MODEL USAGE

No computer software was used for calculations and/or modeling in support of this AMR. No codes or routines were developed.

Reference to parameters used in calculations by GoldSim is made in this AMR. The parameters are provided for use by the TSPA-SR team in GoldSim, but no GoldSim calculations were performed for this AMR.

Graphs and figures were prepared with the following commercially available software: Microsoft Excel 97, Microsoft PowerPoint 97, and Adobe Illustrator 8.0.1, using only the built in standard deviation functions.

4. INPUTS

The rationale for the approach used in the EBS colloids and concentration abstraction for TSPA-SR is based largely on the observation and characterization of radionuclide-bearing colloids formed during waste form degradation tests conducted at ANL, as well as on observations of colloids in groundwater and consideration of the potential contributions of introduced materials to colloid populations in the drift. Data, data interpretation, and conceptual and process models used in the analysis were extended and developed from those used in TSPA Viability Assessment (VA). Additional data and improved process models from ongoing and recently completed laboratory and field testing programs at ANL and LANL are incorporated into this AMR. Results of these activities enabled reduction in uncertainties through the refinement of parameter ranges and a better understanding of fundamental mechanisms of colloid formation, stability, and behavior and the association of radionuclides with colloids.

4.1 DATA AND PARAMETERS

Technical product inputs and sources are listed in Table 1 and on the Document Input Reference System. Primary technical inputs have been taken from CRWMS M&O (2000a, b) input transmittal source DTNs.

Table 1. Summary of Parameters Used in Abstraction

Data Tracking Number (DTN)	Parameter Name	Parameter Description	Units[a]	Basis for Value/ Source	Value and Distribution
Waste Form Colloids					
LL991109851021.095	$C_{RNcoll,wf,irrev,max}$	Highest observed or expected concentration of Pu associated with both DHLW and SNF waste-form colloids	mol/L	CRWMS M&O (2000b)	6e-8 mol/L[b]
MO0003SPALOW12.001	$C_{RNcoll,wf,irrev,min}$	Lowest observed or expected concentration of Pu associated with both DHLW and SNF waste-form colloids	mol/L	CRWMS M&O (2000b)	1e-11 mol/L[b]
LL991109851021.095	$M_{coll,wf,both,max}$	Highest observed or expected mass of both DHLW and SNF waste-form colloids per unit volume or mass of water	mg/L	CRWMS M&O (2000b)	5 mg/L[c]
N/A, intermediate parameter resulting from development of TSPA in this AMR	$C_{RNcoll,wf,irrev}$	Concentration of irreversibly attached (or engulfed as a solid inclusion) radionuclide element RN associated with waste-form colloids	mol/L	Determined in PA calculation	N/A

Data Tracking Number (DTN)	Parameter Name	Parameter Description	Units[a]	Basis for Value/ Source	Value and Distribution
N/A, intermediate parameter resulting from development of TSPA in this AMR	$C_{RNcoll,wf,rev}$	Concentration of reversibly attached radionuclide element RN associated with waste-form colloids	mol/L	Determined in PA calculation	N/A
N/A, intermediate parameter resulting from development of TSPA in this AMR	$M_{coll,wf,both}$	Mass of waste-form colloids with reversibly sorbed or irreversibly attached (or engulfed) radionuclide element RN per unit volume or mass of water	mol/L	Determined in PA calculation	N/A
MO0003SPAION02.003	$I_{lo-thresh,coll,wf}$	Ionic strength below which waste-form colloids are stable	mol/L	CRWMS M&O (2000b)	0.01 mol/L
MO0003SPAION02.003	$I_{hi-thresh,coll,wf}$	Ionic strength above which waste-form colloids are unstable	mol/L	CRWMS M&O (2000b)	0.05 mol/L
MO0004SPAKDS42.005	$K_{d,RNcoll,wf,rev}$	Distribution coefficients for reversible sorption of radionuclide element RN onto waste-form colloidss	mL/g colloid	Data developed from LANL data; CRWMS M&O (2000c)	Geometric mean values: Pu = 1e+4 Am = 1e+5 log-normal distributions; geometric std. dev. = 10
N/A, intermediate function resulting from development of TSPA in this AMR	$ST_{coll,wf,is}$	Function relating stability of waste-form colloids to ionic strength, based on results of DHLW glass degradation experiments (based on Pu concentration)	mol/L	CRWMS M&O (2000b)	[Pu colloid] = -1.25e-6 × I +7.25e-8
N/A, intermediate function resulting from development of TSPA in this AMR	$ST_{coll,wf,pH}$	Function relating stability of waste-form colloids to ionic strength and pH, over limited pH range, based on results of montmorillonite stability experiments	mol/L	CRWMS M&O (2000b)	I = pH/200
Iron-(hydr)oxide Colloids					
N/A, intermediate parameter resulting from development of TSPA in this AMR	$C_{RNcoll,FeOx}$	Concentration of reversibly attached radionuclide element RN associated with iron-(hydr)oxide colloids	mol/L	Determined in PA calculation	N/A
N/A, intermediate parameter resulting from development of TSPA in this AMR	$M_{coll,FeOx}$	Mass of iron-(hydr)oxide colloids per unit volume or mass of water	mg/L	Determined in PA calculation	N/A
MO0003SPAHIG12.002	$M_{coll,FeOx,max}$	Highest observed or expected mass of iron-(hydr)oxide colloids per unit volume or mass of water	mg/L	Reasonable conservative estimate	1e0 mg/L
MO0003SPAHIG12.002	$M_{coll,FeOx,min}$	Lowest observed or expected mass of iron-(hydr)oxide colloids per unit volume or mass of water	mg/L	Reasonable conservative estimate	1e-3 mg/L
MO0003SPAION02.003	$I_{lo-thresh,coll,FeOx}$	Ionic strength below which iron-(hydr)oxide colloids are stable	mol/L	CRWMS M&O (2000b)	0.01 mol/L
MO0003SPAION02.003	$I_{hi-thresh,coll,FeOx}$	Ionic strength above which iron-(hydr)oxide colloids are unstable	mol/L	CRWMS M&O (2000b)	0.05 mol/L

Data Tracking Number (DTN)	Parameter Name	Parameter Description	Units[a]	Basis for Value/ Source	Value and Distribution
MO0004SPAKDS42.005	$K_{d,RNcoll,FeOx}$	Distribution coefficients for reversible sorption of radionuclide element RN onto iron (hydr)oxide colloids	mL/g colloid	Data developed from LANL data; CRWMS M&O (2000c)	Geometric mean values: Pu = 1e+4 Am = 1e+4 log-normal distributions; geometric std. dev. = 10
N/A, intermediate function resulting from development of TSPA in this AMR	$ST_{coll,FeOx,pHlo}$	Function relating stability of iron-(hydr)oxide colloids to pH and ionic strength at relatively low pH values	mol/L	CRWMS M&O (2000b)	$I = -0.02 \times pH +0.17$
N/A, intermediate function resulting from development of TSPA in this AMR	$ST_{coll,FeOx,pHhi}$	Function relating stability of iron-(hydr)oxide colloids to pH and ionic strength at relatively low pH values	mol/L	CRWMS M&O (2000b)	$I = +0.02 \times pH - 0.17$
N/A, intermediate function resulting from development of TSPA in this AMR	$ST_{coll,FeOx,pHint}$	Function relating stability of iron-(hydr)oxide colloids to pH and ionic strength at intermediate pH values	Boolean	CRWMS M&O (2000b)	Boolean (pH ≥ 8 AND pH ≤ 9)
Groundwater Colloids					
N/A, intermediate parameter resulting from development of TSPA in this AMR	$C_{RNcoll,gw}$	Concentration of reversibly attached radionuclide element RN associated with groundwater colloids	mol/L	Determined in PA calculation	N/A
N/A, intermediate parameter resulting from development of TSPA in this AMR	$M_{coll,gw}$	Mass of groundwater colloids per unit volume or mass of water	mg/L	Determined in PA calculation	N/A
MO0003SPAHLO12.004	$M_{coll,gw,max}$	Highest observed or expected mass of groundwater colloids per unit volume or mass of water	mg/L	CRWMS M&O (2000b)	3e-2 mg/L
MO0003SPAHLO12.004	$M_{coll,gw,min}$	Lowest observed or expected mass of groundwater colloids per unit volume or mass of water	mg/L	CRWMS M&O (2000b)	3e-6 mg/L
MO0004SPAKDS42.005	$K_{d,RNcoll,gw}$	Distribution coefficients for reversible sorption of radionuclide element RN onto groundwater colloids	mL/g colloid	LANL data; CRWMS M&O (2000c)	Geometric mean value: Pu, Am = 1e+3 log-normal distribution; geometric std. dev. = 10
MO0003SPAION02.003	$I_{lo-thresh,coll,gw}$	Ionic strength below which groundwater colloids are stable	mol/L	CRWMS M&O (2000b)	0.01 mol/L
MO0003SPAION02.003	$I_{hi-thresh,coll,gw}$	Ionic strength above which groundwater colloids are unstable	mol/L	CRWMS M&O (2000b)	0.05 mol/L
N/A, intermediate function resulting from development of TSPA in this AMR	$ST_{coll,gw}$	Function relating stability of groundwater colloids to ionic strength at intermediate ionic strengths	mg/L	CRWMS M&O (2000b)	see below
Other output					

Data Tracking Number (DTN)	Parameter Name	Parameter Description	Units[a]	Basis for Value/ Source	Value and Distribution
N/A, intermediate parameter resulting from development of TSPA in this AMR	C_{RNcoll}	Concentration of mobile colloidal radionuclide element RN per unit volume or mass of water	mol/L	Determined in PA calculation	N/A
N/A, intermediate parameter resulting from development of TSPA in this AMR	C_{RNdiss}	Concentration of dissolved radionuclide element RN per unit volume or mass of water	mol/L	Input parameter and determined in PA calculation	N/A
N/A, intermediate parameter resulting from development of TSPA in this AMR	M_{coll}	Total mass of all mobile colloids per unit volume or mass of water	mg/L	Determined in PA calculation	N/A
N/A, intermediate parameter resulting from development of TSPA in this AMR	I_{invert}	Ionic strength of water resulting from mixing of fluid from a breached waste package with groundwater in the invert.	mol/L	Determined in PA calculation	N/A
N/A, intermediate parameter resulting from development of TSPA in this AMR	pH_{invert}	pH of water resulting Ionic strength of water resulting from mixing of fluid from a breached waste package with groundwater in the invert.	mol/L	Determined in PA calculation	N/A
Input parameters					
N/A, parameter provided by GoldSim calculation	I_{wp}	Ionic strength of fluid from breached waste package	mol/L	Input parameter	N/A
N/A, parameter provided by GoldSim calculation	pH	pH of fluid from breached waste package	pH units	Input parameter	N/A
N/A, parameter provided by GoldSim calculation	C_{RNdiss}	Concentration of dissolved radionuclide element RN per unit volume or mass of water	mol/L	Input parameter and determined in PA calculation	N/A
N/A, parameter provided by GoldSim calculation	I_{gw}	Ionic strength of groundwater entering drift	mol/L	Input parameter	N/A
N/A, parameter provided by GoldSim calculation	pH_{gw}	pH of groundwater entering drift	pH units	Input parameter	N/A
Other data					
LA0003NL831352.002 LA0003NL831352.003	[referred to in AMR]	Distribution coefficients and forward and reverse adsorption/desorption rates for element RN onto waste-form colloids—source data	various	LANL laboratory data	various

NOTE: [a] mol/L is actually moles per liter of dispersion and is not molarity in the strict sense

[b] uncertainty is linked to uncertainty for ionic strength, which is an input parameter; no additional uncertainty is added

[c] mass is correlated with irreversible radionuclide concentration

4.2 CRITERIA

The NRC Total System Performance Assessment and Integration (TSPAI) IRSR (NRC 1998a) establishes generic technical acceptance criteria considered by the NRC staff to be essential to a defensible, transparent, and comprehensive assessment methodology for the repository system. These regulatory acceptance criteria address five fundamental elements of the DOE TSPA model for the Yucca Mountain site, namely:

1. Data and model justification (focusing on sufficiency of data to support the conceptual basis of the process model and abstractions)

2. Data uncertainty and verification (focusing on technical basis for bounding assumptions and statistical representations of uncertainties and parameter variabilities)

3. Model uncertainty (focusing on alternative conceptual models consistent with available site data)

4. Model verification (focusing on testing of model abstractions using detailed process-level models and empirical observations)

5. Integration (focusing on appropriate and consistent coupling of model abstractions).

Relevant to the topic of this AMR, elements (1) through (4) of the acceptance criteria are addressed herein and/or in the supporting Calculation document(s). Element (5) of the NRC acceptance criteria, which strictly applies to the completed synthesis of process-level models and abstractions, will be addressed separately in the TSPA-SR. As well, this AMR addresses relevant KTIs raised in the NFE IRSR Revision 2 (NRC 1999a) and CLST IRSRs Revisions 1 (NRC 1998b) and 2 (NRC 1999b).

4.3 CODES AND STANDARDS

This AMR was prepared to comply with the above NRC TSPAI acceptance criteria, as well as the DOE interim guidance (Dyer 1999) which directs the use of relevant subparts/sections of the proposed NRC high-level waste rule, 10 CFR Part 63 (64 FR 8640). Subparts of this proposed rule that are particularly applicable to data include Subpart B, Section 15 (Site Characterization) and Subpart E, Section 114 (Requirements for Performance Assessment). Subparts applicable to models are outlined in Subpart E, Sections 114 (Requirements for Performance Assessment) and 115 (Characteristics of the Reference Biosphere and Critical Group).

5. ASSUMPTIONS

Assumptions made as a part of the analysis and model development are presented here in Table 1 and incorporated into the discussion in Section 6, Abstraction and Implementation in GoldSim.

Table 2.   Assumptions Used in the Analysis and Model Development

Category	Assumption	Rationale	Section Used
Colloid Formation	The colloids produced from corrosion of DHLW glass and SNF are predominantly smectite; a good approximation for abstraction is that all waste form colloids are smectite, or are irreversibly attached to smectite.	ANL experimental data indicate that preponderance of DHLW glass-derived colloids is smectite, and that degradation of SNF produces smectite as well (CRWMS M&O 2000a).	6.3
	Metal (hydr)oxide colloids, large particles, and scale may form from degradation of the waste container and its internal components as well as certain EBS components; a good approximation for abstraction is that all metal (hydr)oxides are iron-(hydr)oxides.	The preponderance of metal in the waste container and EBS steels is iron.	6.3
	SNF produces colloids less readily under experimental conditions approximating repository conditions for reasons related to the mechanisms of alteration of the SNF.	Based on observations and inferences in connection with ANL experimental program (CRWMS M&O 2000a)	6.3
	Defense SNF (DSNF) and commercial SNF (CSNF) are considered together as "generic" SNF and are not treated separately in the abstraction	The preponderance of SNF is CSNF. Metallic fuels may produce fine particulates as they degrade, but it is currently considered that DSNF will not adversely affect repository performance (CRWMS M&O 2000b)	6.3
	The waste form degrades according to in-package geochemistry process models and abstractions and are represented by a single GoldSim mixing cell	TSPA methodology (in progress)	6.3
Colloid Stability and Concentration	The stability of smectite colloids will depend to varying degrees on their concentration and the composition, ionic strength, and pH of the solution	Generally acknowledged relationship (e.g., Tombacz et al. 1990)	6.3
	The stability of smectite is less sensitive to pH (is increased) at higher values of pH, more sensitive to pH (is decreased) at lower values	Observed in experiments (Tombacz et al. 1990)	6.3
	Am is assumed to behave as does Pu in terms of mobilization from waste degradation and formation of irreversible association with smectite colloids and clay layers on DHLW	Reasonable assumption based on chemical characteristics of Pu and Am	6.3
	The properties of colloidal smectite and iron-(hydr)oxides are well known and applicable to radionuclide-bearing colloids	Smectite and iron-(hydr)oxide colloid behavior (Tombacz et al. 1990; Liang and Morgan 1990)	6.3
	Iron-(hydr)oxide colloid stability and concentration depend upon both the pH and ionic strength that may be encountered in the repository	Generally acknowledged relationship (Liang and Morgan 1990)	6.3
	Waste form colloids produced at 90°C are stable at 25°C in the drift	Stability is considered to increase as T drops (CRWMS M&O 2000b)	6.3

Category	Assumption	Rationale	Section Used
Radionuclide Attachment to Colloids	Some radionuclides released from degraded SNF occur as aqueous (dissolved) species and are available to sorb to smectite and iron-(hydr)oxide colloids, forming pseudocolloids	Experimental results and solubility determinations indicate that radionuclides may dissolve, and colloids are available for sorption of radionuclides (CRWMS M&O 2000a)	6.3
	Smectite colloids contain entrained radionuclide-bearing phases; further, all discrete radionuclide-bearing colloid-sized phases (besides smectite) forming at/near DHLW glass are entrained in smectite colloids	ANL experimental data indicate that smectite colloids contain entrained or embedded RN-bearing phases; conservative assumption made because reversibility of attachment was not analyzed (CRWMS M&O 2000a).	6.3
	Smectite colloids with or without entrained radionuclide-bearing phases may have adsorbed dissolved radionuclides	Smectite colloids are considered to have the capability of sorbing dissolved radionuclides to form pseudocolloids (e.g., Kim 1994).	6.3
	Pu, Am, Th, Pa, Cs, Np, U, and Sr are assumed to be the most significant radionuclides available for colloid association; data indicate smectite colloids may contain Pu, Am, Th, U, Cm, Np, and rare earth elements (REEs), and that Pu generally behaves similarly to Th, Am and REEs	These are RNs of greatest interest for dose calculations. Smectite colloid RN compositions observed in ANL experiments (CRWMS M&O 2000a).	6.3
	Radionuclide association with colloids is irreversible within the waste container and EBS environments, except for dissolved radionuclides available for pseudocolloid formation.	Reasonable conservative assumption; transport times for colloids leaving the container and traversing the invert are considered to be short in comparison with sorption–desorption times (DTNs: LA0003NL831352.002 and LA0003NL831352.003).	6.3
	Adsorption/desorption of radionuclides on colloids to form pseudocolloids is defined according to experimentally determined rates of adsorption and desorption of Pu, Am, and other radionuclides on several types of colloids, and consequently to an effective K_d incorporating both the forward and reverse attachment rates	Based on results of experiments performed at LANL (DTNs: LA0003NL831352.002 and LA0003NL831352.003)	6.3
Physical Filtration of Colloids	Physical filtration of colloids may occur within EBS/drift but is not considered in the abstraction	All stable colloids are considered to leave the EBS/drift (invert) and enter the UZ, which over-estimates the consequences and is considered bounding	6.3
Colloid Sorption at the Air-Water Interface	Colloid sorption at the air-water interface may occur within the EBS/drift, waste package, but is not considered in the abstraction	This over-estimates the consequences and is considered bounding, as colloid sorption to stationary substrates at air-water interfaces would retard colloid transport	6.3

Category	Assumption	Rationale	Section Used
Gravitational Settling of Colloids	Gravitational settling of non-agglomerated colloids may occur within the EBS/drift, but is not considered in the abstraction.	This over-estimates the consequences and is considered bounding, as all stable colloids are considered not to settle but to leave the EBS/drift and enter the UZ	6.3
Microbes and Colloidal Organic Components	Processes involving interactions between contaminants and microbes and organic components (such as humic and fulvic acids) are not considered in this abstraction	This is considered to over-estimate the consequences and is considered bounding, as biological influences have been known to enhance colloid agglomeration (CRWMS M&O 2000b)	6.3
Colloid Transport	All stable colloids are assumed to leave the EBS/drift and enter the UZ	This over-estimates the consequences and is considered bounding; processes such as sorption to EBS components, gravitational settling, and filtration would tend to retain colloids in the drift.	6.3

6. ANALYSIS/MODEL

6.1 INTRODUCTION

Colloids may affect repository performance if they are first generated in significant quantities, are stable within the waste package, EBS, and far-field environments, carry a significant radionuclide load, and transport readily. Processes relevant to the waste package and EBS environment are discussed in this section in the context of conceptual models, process models, and approach to abstraction used in this effort.

6.2 COLLOID CHARACTERISTICS

6.2.1 Definitions

Several types of colloids are expected to be generated within the Yucca Mountain repository system. Colloids will be present at the waste form and in the drift and the unsaturated and saturated zones. At the waste form, colloids are produced during the degradation of the waste form and waste package. In the repository, additional colloids may be produced from the degradation of steel and concrete components of the EBS. Several types of colloids (mineral and organic colloids) are present in the unsaturated zone, as well as the saturated zone, and may be introduced into the repository. Mineral colloids include clays, iron (hydr)oxides (goethite, hematite), and silica; organic colloids include microbes and microbe fragments as well as humic and fulvic acids. Formation and introduction of colloids to the EBS/drift environment are discussed below in the context of the models considered in the abstraction.

(a) (b) (c)
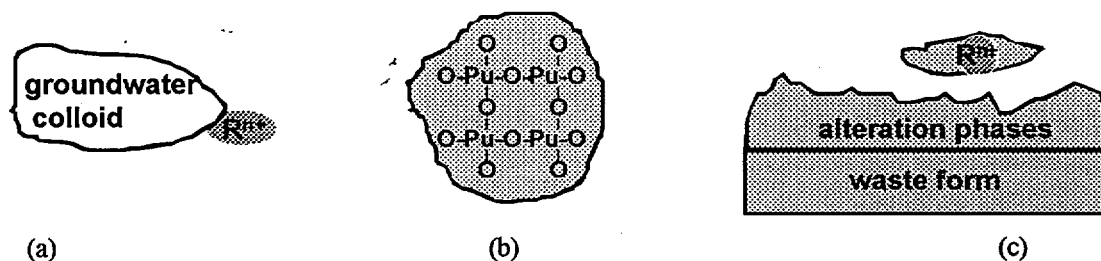
NOTE: Several types of radionuclide-bearing colloids are depicted: (a) pseudo-colloids, (b) real colloids, and (c) waste form colloids. The radionuclide (R^{n+}) associated with the pseudo and waste form colloids can be an ionic species, a real colloid, or a discrete radionuclide-bearing phase. The size range for the colloids is approximately 1 to 1000 nm. (CRWMS M&O 2000a, Figure 1)

Figure 1. Several Types of Radionuclide-Bearing Colloids

Real colloids are formed from polymerization of hydrolyzed radionuclide ions (Kim 1994). (Real colloids may be considered a subset of *intrinsic colloids*, in which radionuclides form a part of the chemical compound comprising a colloidal particle.) For real colloids to form it is necessary that the fluid be saturated with respect to a solid phase, sometimes amorphous; thus the formation of real colloids is solubility limited. No real colloids were detected in the experimental investigations at ANL (CRWMS M&O 2000a). Further, the dilute (low ionic strength) nature of J-13 groundwater suggests that real colloids are unlikely to form in the drift.

Pseudocolloids may be formed by the incorporation, adsorption, and ion exchange of dissolved radionuclides onto existing colloids, either formed from waste form degradation, from introduced man-made materials, or introduced with groundwater (Kim, 1994). Thus radionuclides which might otherwise sorb to minerals in the rock, or other immobile substrates, instead attach to mobile colloidal particles and added to the dissolved radionuclides increase the mobile radionuclide mass.

Waste form colloids, or *primary colloids* (also a subset of *intrinsic colloids*), may provide the most significant contribution to colloid-facilitated radionuclide transport. These colloids form from nucleation of colloids from waste form dissolution and spallation of colloid-sized waste form alteration products which contain radionuclides and have the potential for increasing mobile concentrations higher than achievable with real colloids or pseudocolloids (CRWMS M&O 2000a).

6.2.2 Colloid Formation and Occurrence

6.2.2.1 Colloids from Waste Form Degradation

Three broad categories of waste form are to be placed in the repository. The most abundant waste type is commercial spent nuclear fuel (CSNF) from commercially owned and operated electric power reactors. Defense spent nuclear fuel (DSNF) is a diverse collection of waste from reactors at U.S. Department of Energy (USDOE) nuclear complex sites. Defense high-level radioactive waste (DHLW) is a borosilicate glass-based waste form containing radionuclides. In the following descriptions, CSNF and DSNF are treated together.

<u>Corrosion of DHLW</u>. Testing on corrosion of DHLW has gone on for over ten years in the case of static tests, in which glass samples are immersed in fluid, and for over four years in the case of drip tests, in which fluid is dripped at specified rates onto the glass sample. In the static tests, geometric surface area (SA) of the sample was varied relative to the volume (V) of fluid used; this proportion is referred to as SA/V. Glass waste forms tested include Savannah River Laboratory (SRL) and West Valley Demonstration Project (WVDP) glasses. Fluids used were equilibrated J-13 water (EJ-13) and deionized water. Methods used to characterize colloids produced during the tests include transmission electron microscopy and filtration (analysis of filtrates with inductively coupled plasma-mass spectrometry or alpha spectroscopy). Descriptions of the tests may be found in CRWMS M&O (2000a) and Ebert (1995).

Colloids produced from both types of glass waste testing were primarily smectite clay containing discrete radionuclide-bearing phases which are incorporated ("entrained") in the clay (CRWMS M&O 2000a). Iron silicate colloids were also observed. The entrained phases were identified as primarily brockite (thorium calcium orthophosphate) and an amorphous thorium-titanium-iron silicate, similar to thorutite. The phases also contained other actinides and rare earth elements (REEs). Uranium was detected within the clays and iron silicates in some samples.

Currently no data are available on the chemical and physical properties of the entrained phases, so that their solubility and disassociation/association properties with the clay cannot be derived and used in the model. The colloidal properties of the entrained radionuclide-bearing phases therefore are assumed to be governed by the properties of the smectite, and the stability (and therefore mobility) of smectite colloids will control the mobility of the entrained phases. (Testing is currently underway at LANL to determine the attachment/detachment properties of the entrained radionuclide-bearing phases in smectite colloids produced from the ANL glass tests.) Accordingly, the radionuclides are assumed to be irreversibly attached to the smectite colloids.
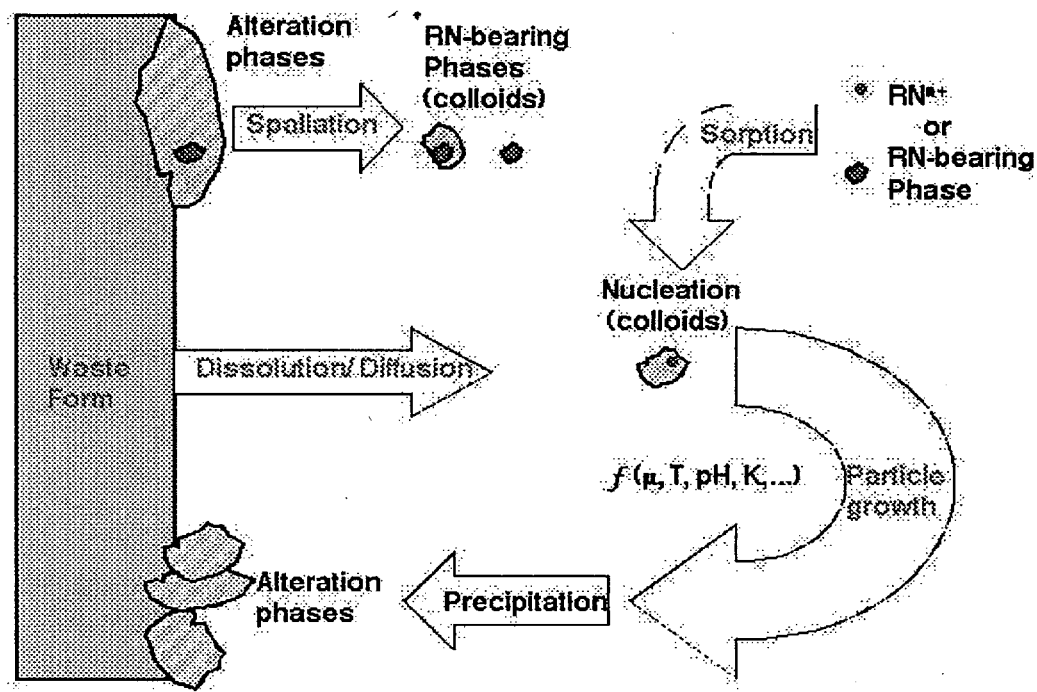
Figure 2. Schematic of colloid formation from waste form corrosion whereby several processes are represented. First, spallation of radionuclide (RN) bearing alteration phases from the waste form that are within the colloidal size range, and second, nucleation of colloids from dissolution of waste form and sorption of ionic RN species or RN-bearing phases. Particle growth is controlled by factors such as ionic strength, temperature, pH, solubility, and others. When particle diameters exceed 1 micron or solution chemistry destabilizes the colloids, precipitation of RN-bearing alteration phases occurs. (CRWMS M&O 2000a, Figure 2)

As the glass corrodes, the clay alteration product forms, grows, and evolves as a layer on the surface of the glass. The clay layer grows as the glass beneath the clay alters, and as the clay-forming elements saturate in the solution and precipitate on the clay surface. Through these mechanisms colloid-sized clay that does not attach to the clay layer also precipitates from the solution (CRWMS M&O 2000a).

It was observed in the static tests that colloids developed and increased in concentration with time, up to a point where the colloid concentration reached a maximum and then the dispersion became unstable. This was attributed by the investigators primarily to the accompanying increase in ionic strength to a threshold above which the colloids flocculated and settled by gravity (CRWMS M&O 2000a).

Corrosion of SNF. Testing on the corrosion of SNF has been conducted for over four years. Two types of spent fuel from pressurized water reactors with different burn-ups were used. EJ-13 water was introduced in two ways, at the bottom of the test vessel to maintain 100% relative

humidity, and by dripping onto the top of the spent fuel fragments. Generated colloids were characterized by using the same analytical techniques as were used in the glass testing. Descriptions of the SNF tests may be found in CRWMS M&O (2000a) and Finn et al. (1994).

In comparison to what was observed in the glass waste form tests, very little radionuclide-bearing colloidal and suspended particle material was detected in the fluid samples. (It is possible that this was a result of the test vessel configuration [CRWMS M&O 2000a].) In one sample, colloids characterized were primarily a hydrous uranium oxide (schoepite) and a partially crystalline uranium silicate (soddyite). The colloids contained the rare earth elements (REEs) La, Ce, Pr, Nd, Sm, and Eu. Plutonium was not detected in these colloids; however, alteration on the corroded surface of the spent fuel contained significant concentrations of Pu, along with Zr, Ru, REEs.

In another sample, smectite clay colloids were formed that were similar structurally and compositionally to the smectites produced in the glass tests, although they contained Mn, Ni, Fe, and Cr.

Further details on degradation of CSNF may be found in CRWMS M&O (2000b), Section 6.1.1.1.

6.2.2.2   Natural Colloids in Groundwater

Three types of groundwater colloids may be present at Yucca Mountain:

(1) Mineral fragments are generally hydrophobic, hard particles that are kinetically stabilized or destabilized by electrostatic forces and may consist of crystalline or amorphous solids (Kim 1994). Steric coatings may enhance the stability of mineral fragments by preventing close contact. Mineral fragments may act as substrates for sorption of radionuclides (they may also consist of precipitated or coprecipitated actinide solids).

(2) Humic substances are generally hydrophilic particles that are stabilized by solvation forces (Minai et al. 1992). They can be powerful substrates for uptake of metal cations and are relatively small (less than 100,000 atomic mass units).

(3) Microbes are relatively large colloidal particles that are stabilized by hydrophilic coatings on their surfaces, which behave as steric stabilizing compounds. They may act as substrates for extracellular actinide sorption or they may actively bioaccumulate radionuclides intracellularly.

Clays, silica, hematite, and goethite colloids occur in groundwater in the vicinity of Yucca Mountain, and it is assumed that small quantities of these colloids will enter a failed waste package and the invert and be available to interact with released radionuclides. The presence and potential influence of natural colloids on formation of radionuclide-bearing colloids in the invert (as well as in the waste package) are considered by assuming that groundwater colloids are smectite and iron (hydr)oxide.

An assessment of the concentration of humic substances in groundwaters collected near the Yucca Mountain was conducted by Minai et al. (1992). In that study, humic substances were

extracted from several thousand gallons of J-13 well water through a series of steps including acidification, purifying with ion exchange columns, and freeze drying. The humic substances that were collected were characterized with NMR and other spectroscopic techniques. Experiments were conducted to characterize sitebinding densities and complexation with Am. Results and supporting calculations indicated that the presence of humic substances in J-13 well water could affect oxidation speciation by reducing some radionuclide species. Considering the presence of calcium in J-13 well water, however, the authors estimated that the effective complexation capacity of humic substances is about 2.3×10^{-10} eq/L (equivalents per liter) at pH 6.9 and 2.7×10^{-11} eq/L at pH 8.2 (page 199). Considering the very low complexation capacity of humic substances in this system, they are not considered as groundwater colloids.

Because of the relatively large sizes of microbes, they are very susceptible to filtration in geologic media. Consequently, microbe-facilitated contaminant transport is not considered in this abstraction.

6.2.2.3 Colloids from Degradation of Introduced Materials

"Introduced materials," materials brought into the repository excavation during construction and waste storage, may behave as a source of colloids. Due to the large quantities of these materials distributed over several hundred miles of repository drift, it has been said that the contributions of some of these materials could be significant (Meike and Wittwer 1993).

Colloids may form from: (a) materials that are emplaced as, or originally contain, colloids; and (b) materials that degrade to form colloids. Meike and Wittwer (1993) list several types of materials likely to be used during repository construction, construction, during the service lifetime of the facility, or as permanent fixtures:

- *Insoluble metals* such as rockbolts and measurement devices.
- *Insoluble organics* such as cementitious materials and backfill materials.
- *Insoluble organic solids* such as neoprene, epoxy resin, plastic, rubber, and exhaust deposits.
- *Inorganic liquids* such as water for drilling, dust control, and cement emplacement.
- *Miscible organic liquids* such as fluorescein dye and antifreeze.
- *Immiscible organic liquids* such as hydraulic fluids, lubricants, oils, paints, greases, and fuels.
- *Gases* such as H_2, N_2, CO, CO_2, and C_2H_2.

Colloids may possibly form as a result of release from bulk material (dissolution of matrix, mechanical stress, etc.), aggregation, microbial activity, or introduction. (Hydr)oxides, primarily of iron, may form as colloids from corrosion of ferrous drift components; corroding steels may also provide Co, Cr, Cu, and Ni. Hydrated cement and concrete may release colloids, and studies have suggested that sorption of metals and actinides onto cement particles appears to be independent of particle size, and results from batch experiments are similar to results from field studies (Meike and Wittwer 1993).

Analyses of EBS processes and potential colloid phenomena are summarized in CRWMS M&O (2000d). In general, it appears likely that the most important processes contributing to colloid-induced radionuclide mobility are waste form degradation and corrosion of ferrous repository components. Cementitious components may produce some clay colloids; such potential contributions are effectively included in the clay waste form and groundwater colloid analyses. The titanium drip shield and fuel cladding not likely to produce colloids. Crushed rock material may contain colloid-sized particles, but they are likely to be clay and iron (hydr)oxide, which are already included in waste form and corrosion colloid analyses.

6.2.3 Colloid Stability and Concentration

For a detailed discussion of colloid stability and concentration, the reader is referred to CRWMS M&O (2000b), Section 6.1.2.

6.2.4 Colloid Transport Properties and Processes

For a detailed discussion of colloid transport properties and processes, the reader is referred to CRWMS M&O (2000b), Sections 6.1.3 through 6.1.9. The discussion includes radionuclide attachment to colloids, physical filtration of colloids, colloid sorption at the air-water interface, gravitational settling of colloids, colloid diffusion, and microbes and organic components. In this section, the development of K_d values for uptake of strontium (Sr), cesium (Cs), thorium (Th), uranium (U), protactinium (Pa), neptunium (Np), plutonium (Pu), and americium (Am) on colloids is described, along with caveats. These radionuclide elements were chosen for analysis because, based on a screening exercise, they are considered by TSPA to be important to dose; high uncertainty surrounds their potential behavior as pseudocolloids; and daughter products of uranium may remain embedded in uranium-bearing colloids.

6.3 ABSTRACTION AND IMPLEMENTATION IN GoldSim

6.3.1 Abstraction Summary

Clays, silica, hematite, and goethite colloids occur as mineral colloids in groundwater in the vicinity of Yucca Mountain, with clays and silica the most abundant. It is assumed for simplicity and conservatism that groundwater colloids are clays: (1) iron (hydr)oxide colloids are present in very small amounts, and (2) Pu and Am appear to sorb more strongly to, and desorb generally more slowly from, clays than silica colloids (LA0003NL831352.002 and LA0003NL831352.003), and, therefore, substituting clay for silica colloids is conservative. It is assumed that these colloids will enter a failed waste package along with groundwater and be available to interact with released radionuclides. Further, it is assumed that groundwater entering the drift and invert from the surrounding unsaturated zone will mix under certain circumstances with releases from a failed waste package and the groundwater colloids will likewise be available to interact with released radionuclides.

Two scenarios are analyzed in TSPA. In the first scenario, the drip shield is intact and incoming groundwater from above flows around the drip shield and migrates downward through the invert

(and backfill, if used) and into the unsaturated zone. No waste package is breached, and therefore no release from a waste package occurs.

In the second scenario, the drip shield has failed, and incoming groundwater from above flows downward through the breach in the drip shield, onto and around the waste package, and downward into the invert. Releases from a breached waste package migrate downward through the invert and mix with the groundwater in the invert. The mixed fluid migrates downward through the invert into the unsaturated zone.

The groundwater is assumed to contain smectite colloids whose stability and concentration are determined by the ionic strength and pH of the groundwater. The waste package release is assumed to be a fluid containing colloids and dissolved radionuclides resulting from the reaction of waste with groundwater that has entered the waste package (calculated by GoldSim).

There are three types of colloids in the release: (1) waste form colloids, assumed to be smectite; (2) waste package corrosion colloids, assumed to be iron (hydr)oxide; and (3) groundwater colloids, assumed to be smectite. Some of these colloids have associated radionuclides as they leave the waste package. The waste form colloids may have irreversibly attached (embedded) and/or reversibly attached (sorbed) radionuclides. The corrosion and groundwater colloids may have reversibly attached radionuclides. (It should be stated that the terms "reversible" and "irreversible" as used here imply mechanism of attachment. Because of the strong and rapid sorption of actinides to iron (hydr)oxide colloids and their very limited, slow desorption (DTNs: LA0003NL831352.002 and LA0003NL831352.003), this *sensu strictu* "reversible" attachment may be considered effectively "irreversible" over the colloid migration travel times anticipated within the waste package and the EBS.)

The mass of radionuclides irreversibly attached to the waste form colloids is determined from reaction within the waste package (CRWMS M&O 2000b). The mass of radionuclides reversibly attached to all three types of colloids is determined by the product of three parameters: (1) the mass concentration of dissolved (aqueous) radionuclide in the fluid, (2) the mass concentration of colloid material in the fluid, and (3) the k_d of a specific radionuclide on a specific colloid mineralogical type.

6.3.2 Inputs to GoldSim

The abstraction uses a set of input parameters for the TSPA code GoldSim; these parameters are listed and described in Table 1. Most of the input parameters are taken from CRWMS M&O 2000a, CRWMS M&O 2000b, CRWMS M&O 2000c, LA0003NL831352.002, and LA0003NL831352.003; certain others have been extracted from available Project documents and the scientific literature.

6.3.3 Calculated GoldSim Parameters Needed for Abstraction

Parameters needed for the GoldSim calculation of invert processes include outputs from the waste package to the invert (radionuclide and colloid concentrations, ionic strength, and pH), and

input from the unsaturated zone to the drift (ionic strength and pH). Inputs are described in Table 1 and Figures 3 through 13.

6.3.4 Calculation of Mass Concentrations of Colloid-Associated Radionuclides and Colloids

This section describes the logic presented in Figures 3, 6, 8, and 10 through 13. Parameters used in the abstraction are presented in Table 1.

The mass concentration of radionuclides leaving the waste package is calculated by GoldSim for each time step. The mass concentration of colloids is affected by the ionic strength and pH of the fluid and is determined by GoldSim for each time step. The k_ds for the various radionuclides on the two mineralogical colloid types have been determined in the laboratory and taken from LA0003NL831352.002, LA0003NL831352.003 and CRWMS M&O (2000c).

In the first scenario, the groundwater and waste package release both migrate downward but separately into the unsaturated zone, and the chemistry of both the groundwater and waste package release, including the calculated masses of radionuclides and colloids, remain unchanged during migration through the invert and backfill to the unsaturated zone threshold.

In the second scenario, the groundwater and waste package release mix within the invert, a fluid of combined chemistry is produced, and new mass concentrations of dissolved radionuclides and colloids, as well as masses of radionuclides sorbed onto colloids, are calculated by GoldSim.

6.3.4.1 Inputs to the Drift

Outputs from the waste package, which are also inputs to the drift, are colloid-associated radionuclide concentrations (C_{RNcoll}), colloid masses (M_{coll}), and the water chemical parameters I_{wp} (ionic strength) and pH_{wp}. (Figure 3).

The total C_{RNcoll} is made up of individual radionuclide mass concentrations irreversibly and reversibly attached to waste form (smectite) colloids ($C_{RNcoll,wf,irrev}$ and $C_{RN,wf,rev}$), reversibly attached to corrosion iron-(hydr)oxide colloids ($C_{RNcoll,FeOx}$), and reversibly attached to groundwater (smectite) colloids ($C_{RNcoll,gw}$).

The total M_{coll} is the sum of the mass concentrations of the waste form colloids ($M_{coll,wf,both}$; "both" signifies that all smectite waste form colloids irreversibly or irreversibly associated with radionuclides, or both, are included); the iron-(hydr)oxide corrosion colloids; and the smectite groundwater colloids.

Groundwater chemical parameters I_{gw} (ionic strength) and pH_{gw} are also inputs to the invert.
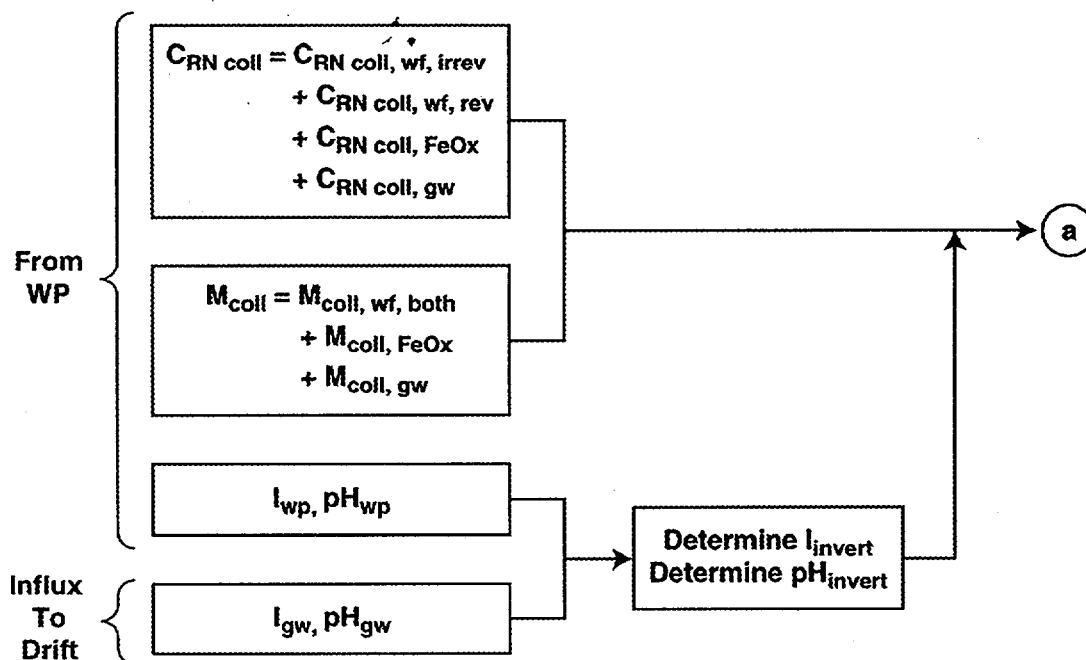
Figure 3. Inputs To Invert from Waste Package and Water Mixing In Invert

6.3.4.2 Water mixing in the drift

In the groundwater-waste package release mixing scenario, the water chemical parameters I_{invert} (ionic strength) and pH_{invert} are determined from the resultant chemical composition of the mixture. This is schematically illustrated in Figure 3.

6.3.4.3 Determination of stability and mass of waste form colloids as a function of I and pH

The effect of pH and ionic strength on the stability of waste-form colloids (based on stability behavior of montmorillonite colloids) has been abstracted from CRWMS M&O (2000b) (Figure 12) as shown in Figure 4. The estimation of waste form colloid mass has been abstracted from CRWMS M&O (2000b) (Figure 13) and is shown in Figure 5. The colloid mass calculation is illustrated by the flow chart and is performed according to the logic statements in Figure 6.
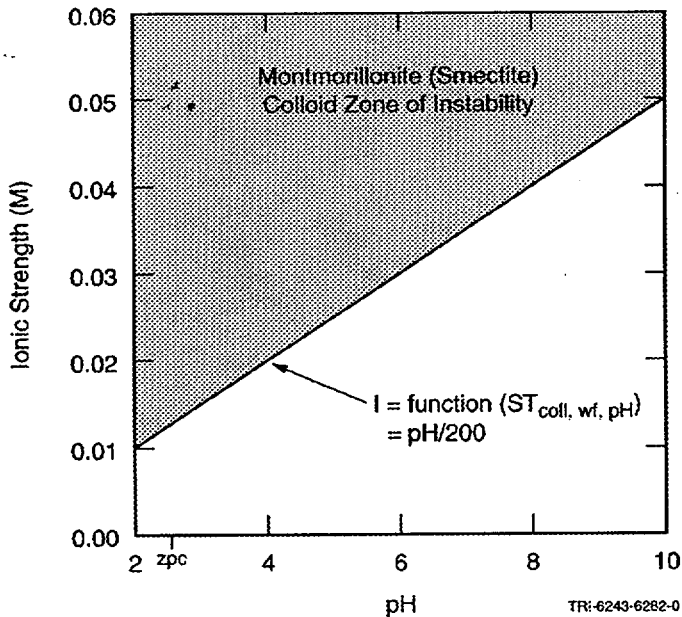
NOTE: Schematic representation (used in abstraction) of smectite stability as a function of pH and ionic strength. Figure 12 from CRWMS M&O (2000b).

Figure 4.   Schematic Representation of Smectite Stability as a Function of pH and Ionic Strength
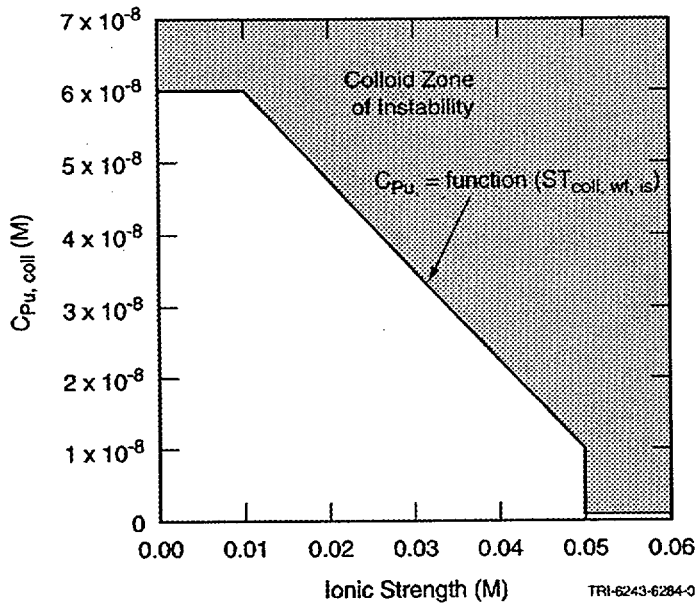
NOTE: Schematic relationship (used in abstraction) between RN-bearing colloid concentration and ionic strength. The function represents the bound of the DHLW corrosion experimental data (see CRWMS M&O (2000b) Figure 7); the maximum value represented, 6×10^{-8}, is the maximum concentration of colloids observed in the DHLW experiments. (Figure 13 from CRWMS M&O 2000b).

Figure 5.   Schematic Relationship Between Radionuclide-Bearing Colloid Concentration and Ionic Strength

Figure 6. Stability, Mass of WF Colloids - f (I, pH)

Logic statements for Figure 6:

Input I_{invert} and pH_{invert}

IF $((I >= I_{hi-thresh,coll,wf})$ OR $(I >= function(ST_{coll,wf,pH}))$

$\quad$ THEN $C_{RNcoll,wf,irrev} = C_{RNcoll,wf,irrev,min}$

function $(ST_{coll,wf,pH})$: $pH/200$

The mobile mass of waste form colloids is determined as follows:

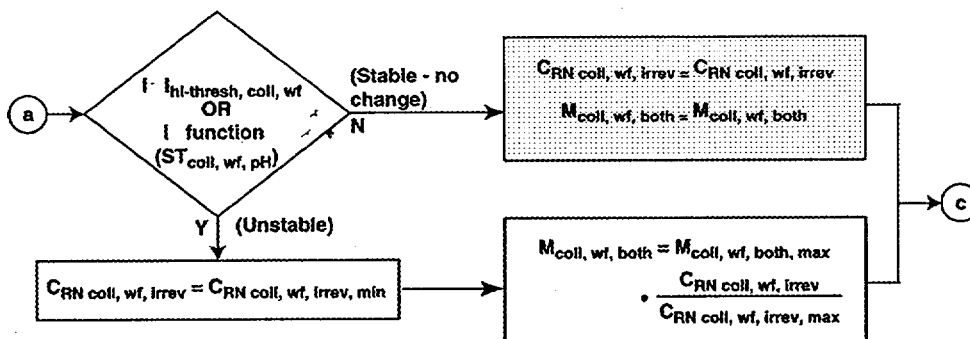$M_{coll,wf,both} = M_{coll,wf,both,max}$ x $C_{RNcoll,wf,irrev}$ / $C_{RNcoll,wf,irrev,max}$

6.3.4.4 Determination of stability and mass of iron (hydr)oxide colloids as a function of I and pH

The effect of pH and ionic strength on the stability of iron (hydr)oxide colloids, as well as determination of colloid mass, has been abstracted from CRWMS M&O (2000b) (Figure 11) and presented in Figure 7. The calculation of colloid stability and mass is illustrated by the flow chart and calculated according to the logic statements in Figure 8.
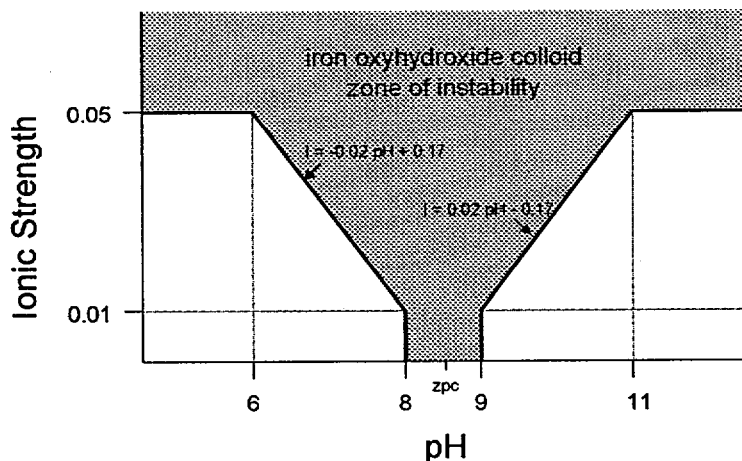
NOTE: Schematic representation (used in abstraction) of iron-(hydr)oxide colloid stability as a function of ionic strength and pH. At and near the ZPC colloids are unstable, even at low ionic strengths. At higher ionic strengths the pH range at which colloids are unstable is greater. Above ionic strength 0.05 colloids are assumed to be unstable for all pH (Abstracted from Liang and Morgan 1990, Figure 1). Figure 11 from CRWMS M&O (2000b)

Figure 7. Schematic Representation of Iron-(Hydr)oxide Colloid Stability as a Function of pH and Ionic Strength
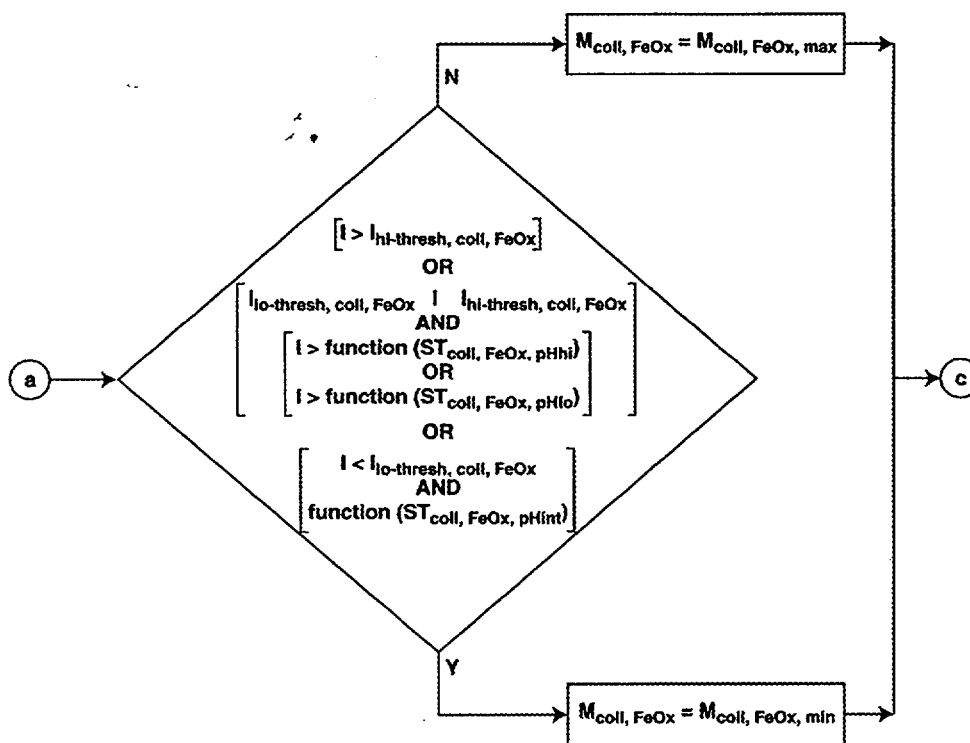
Figure 8. Stability, Mass of FeOx Colloids - f (I, ph)

<u>Logic statements for Figure 8</u>:

IF

    BEGIN

        $(I > I_{hi\text{-}thresh,coll,FeOx})$

        OR $((I >= I_{lo\text{-}thresh,coll,FeOx})$

            AND $(I > function\ (ST_{coll,FeOx,pHhi}))$ OR $(I > function\ (ST_{coll,FeOx,pHlo})))$

        OR $((I < I_{lo\text{-}thresh,coll,FeOx})$ AND $(function\ (ST_{coll,FeOx,pHint})))$

    END

    THEN

        $M_{coll,FeOx} = M_{coll,FeOx,min}$

ELSE

    $M_{coll,FeOx} = M_{coll,FeOx,max}$

function$(ST_{coll,FeOx,pHhi})$: -0.02 x pH + 0.17
function$(ST_{coll,FeOx,pHlo})$: +0.02 x pH - 0.17
function$(ST_{coll,FeOx,pHint})$ (boolean): ((pH >=8) AND (pH <=9))

6.3.4.5. Effect of ionic strength on mobile mass of groundwater colloids

The effect of ionic strength on the mobile mass of groundwater colloids has been abstracted from CRWMS M&O (2000b) (Figure 14) and is presented in Figure 9. The calculation of groundwater colloid mass is illustrated by the flow chart and calculated according to the logic statements in Figure 10.
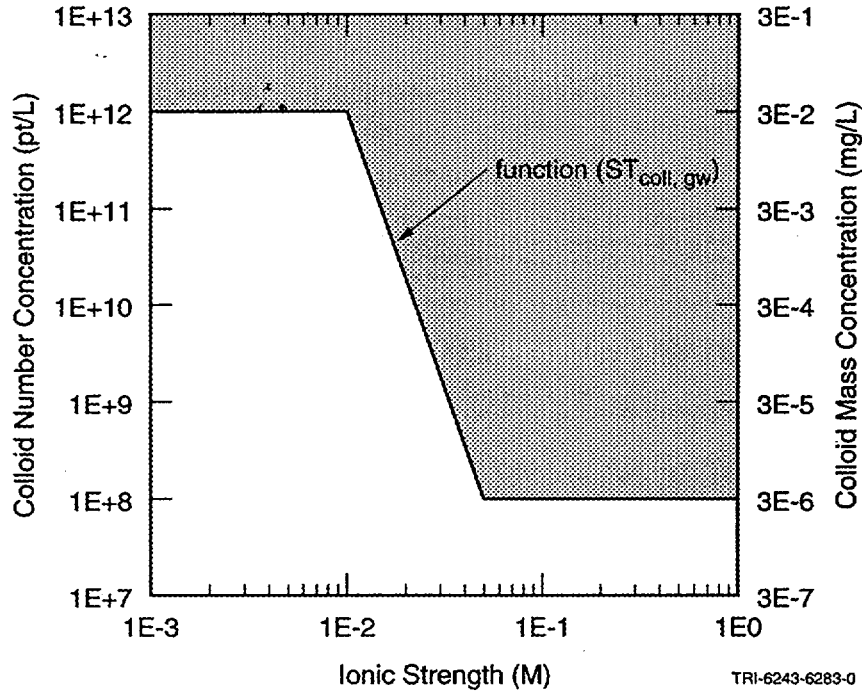
NOTE: Schematic relationship (used in abstraction) between groundwater colloid concentration and ionic strength. Colloid masses were calculated using number populations from Figure 9 extrapolated to include colloid sizes between 10 nm and 1 micrometer. A colloid mineral density of 2.5 g/cm3 was assumed. Figure 14 from CRWMS M&O (2000b).

Figure 9.   Schematic Relationship between Groundwater Colloid Concentration and Ionic Strength
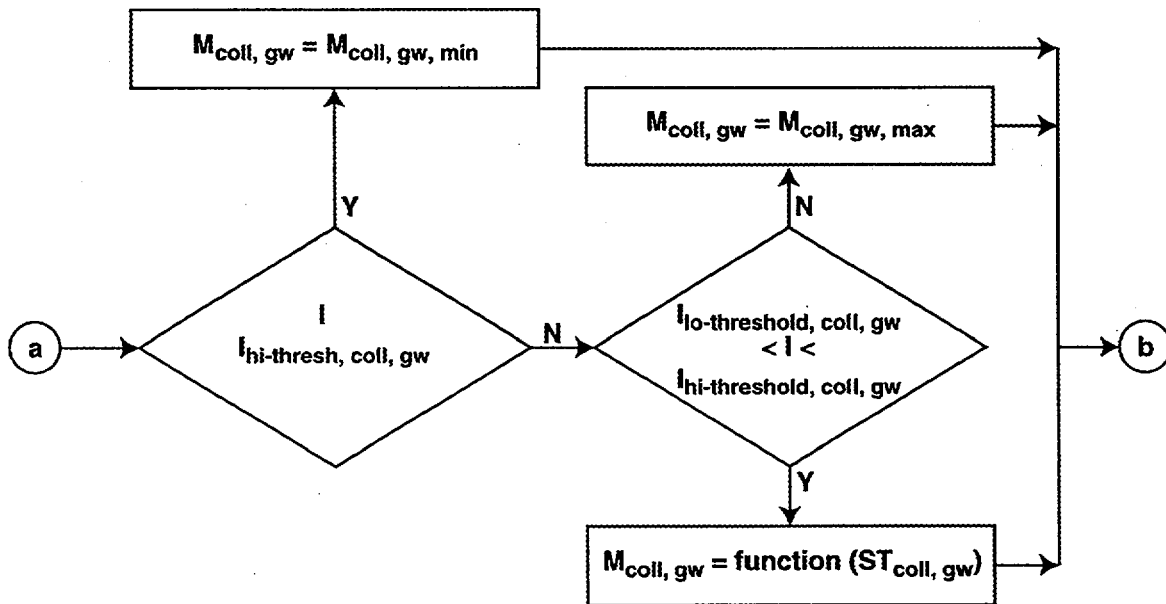
Figure 10. Groundwater Colloids - Effect of Ionic Strength on Mobile Mass

Logic statements for Figure 10:

IF I $>=$ $I_{hi\text{-}thresh,coll,gw}$

$\qquad$ THEN $M_{coll,gw} = M_{coll,gw,min}$

$\qquad$ ELSE IF $(I > I_{lo\text{-}thresh,coll,gw})$ AND $(I < I_{hi\text{-}thresh,coll,gw})$

$\qquad\qquad$ THEN $M_{coll,gw} = $ function$(S_{coll,gw})$

$\qquad$ ELSE $M_{coll,gw} + M_{coll,gw,max}$

function$(S_{coll,gw})$: $10^{\wedge}[A \times \log(I) + B]$

$\qquad$ where:

$A = [\log(M_{coll,gw,max}) - \log(M_{coll,gw,min})] / [\log(I_{lo\text{-}thresh,coll,gw}) - \log(I_{hi\text{-}thresh,coll,gw})]$

$B = \{[\log(M_{coll,gw,min}) \times \log(I_{lo\text{-}thresh,coll,gw})] - [\log(M_{coll,gw,max}) \times \log(I_{hi\text{-}thresh,coll,gw})]\} / [\log(I_{lo\text{-}thresh,coll,gw}) - \log(I_{hi\text{-}thresh,coll,gw})]$

6.3.4.6 Determination of stability and mass of groundwater colloids (based on stability behavior of montmorillonite colloids) as a function of I and pH

The effect of pH and ionic strength on the stability and mass of groundwater colloids (based on stability behavior of montmorillonite colloids) is illustrated by the flow chart and calculated according to the logic statements in Figure 11.

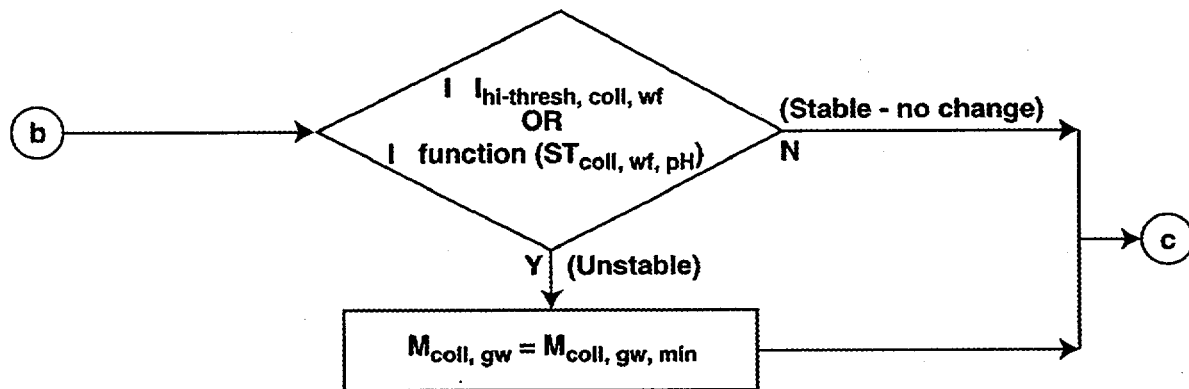Figure 11. Groundwater Colloids - Effect of pH on Stability

Logic statements for Figure 11:

IF $(I >= I_{hi\text{-}thresh,coll,wf})$ OR $(I >+ $ function$(ST_{coll,wf,pH}))$

$\qquad$ THEN $M_{RNcoll,gw} = M_{RNcoll,gw,min}$

function$(ST_{coll,wf,pH})$: pH/200

6.3.4.7 Redistribution of colloid-associated radionuclides in invert

The redistribution of radionuclides reversibly attached to colloids, and the determination of total dissolved radionuclides sorbed onto colloids, is illustrated by the flow chart in Figure 12 and by the logic statements below.

Reversibly Sorbed Colloidal RN and Dissolved RN

$$C_{RN\ diss} = C_{RN\ diss} + C_{RN\ coll,\ wf,\ rev} + C_{RN\ coll,\ FeOx} + C_{RN\ coll,\ gw}$$

$$C_{RN\ coll,\ wf,\ rev} = C_{RN\ diss} \cdot K_{d,\ RNcoll,\ wf,\ rev} \cdot M_{coll,\ wf,\ both}$$
$$C_{RN\ coll,\ FeOx} = C_{RN\ diss} \cdot K_{d,\ RNcoll,\ FeOx} \cdot M_{coll,\ FeOx}$$
$$C_{RN\ coll,\ gw} = C_{RN\ diss} \cdot K_{d,\ RNcoll,\ gw} \cdot M_{coll,\ gw}$$

Irreversibly Attached Colloidal RN

$$C_{RN\ coll,\ wf,\ irrev}$$
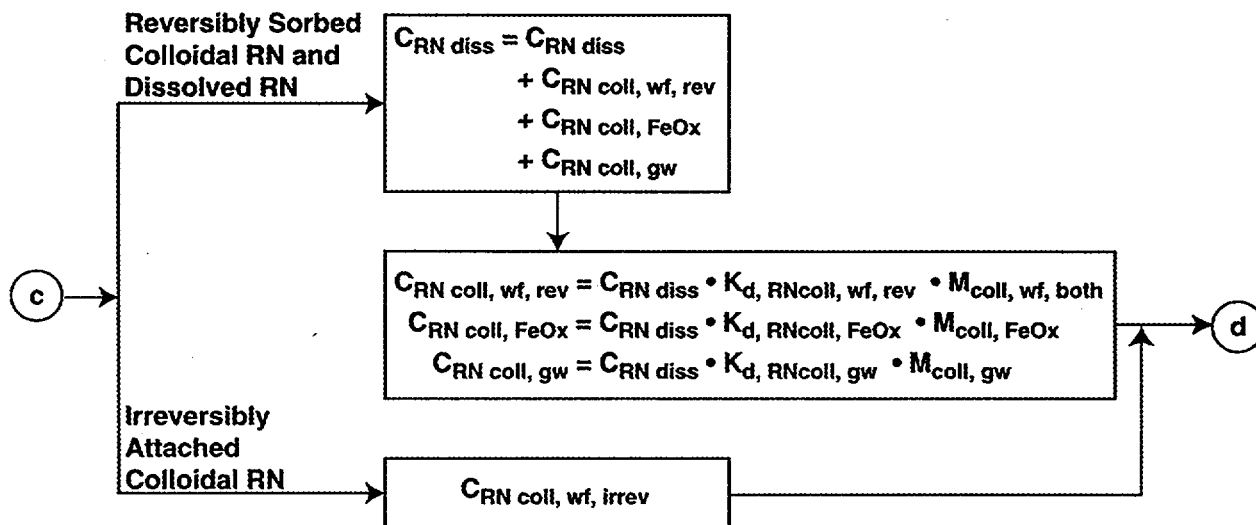
Figure 12. Redistribution of Colloid - Associated Radionuclides in Invert

Calculation of total radionuclide concentration available for redistribution:

$$C_{RNdiss} = C_{RNdiss} + C_{RN,coll,wf,rev} + C_{RN,coll,FeOx} + C_{RNcoll,gw}$$

Calculation of radionuclide sorption on colloids:

$$C_{RNcoll,wf,rev} = C_{RNdiss} \times K_{d,RN,wf} \times M_{coll,wf,both} \times 1E\text{-}6$$
$$C_{RNcoll,FeOx} = C_{RNdiss} \times K_{d,RN,FeOx} \times M_{coll,FeOx} \times 1E\text{-}6$$
$$C_{RNcoll,gw} = C_{RNdiss} \times K_{d,RN,gw} \times M_{coll,gw} \times 1E\text{-}6$$

6.3.4.8 Calculation of colloid source term for radionuclide element RN

The colloid source term is calculated by summing the determined concentrations of waste-form, iron (hydr)oxide, and groundwater colloids and the concentrations of the radionuclides associated with the colloids, as shown in Figure 13.

Figure 13. Source-term Summation for RN in Invert

Logic statements for Figure 13:

$C_{RNcoll} = C_{RNcoll,wf,irrev} + C_{RNcoll,wf,rev} + C_{RNcoll,FeOx} + C_{RNcoll,gw}$

$M_{coll} = M_{coll,wf,both} + M_{coll,FeOx} + M_{coll,gw}$

6.4 ASSESSMENT AND VALIDATION OF ABSTRACTION

The model is considered valid for its intended purpose based on several considerations. The assumptions used are considered conservative and/or bounding, depending upon the assumption and the basis for the assumption.
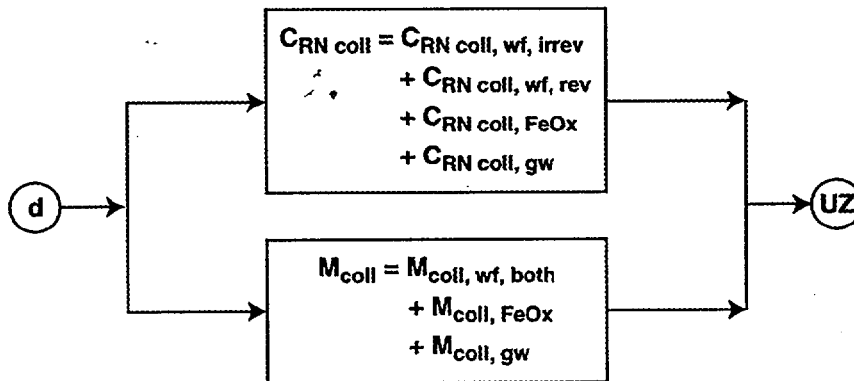
The types and characteristics (including stability and concentration) of colloids formed from the degradation of the waste forms and likely to occur in the EBS/drift as used in the abstraction are based on extensive observations of colloids from testing programs and natural groundwaters. Because of the minute dimensions and unique properties of colloids, their study presents formidable difficulties; the body of work used for the abstraction developed in this AMR represents a significant fraction of the state-of-the-art colloid research performed to date.

Certain processes were not incorporated into the abstraction, including filtration of colloids in the EBS/drift, sorption to the air-water interfaces in unsaturated environments, and gravitational settling. It was considered realistic to conservative to do so, as it was assumed that all of the stable colloids generated within a breached waste package left the waste package and entered the invert in the drift (CRWMS M&O 2000b). Further, it was assumed that all stable colloids entering the invert enter the UZ.

Attachment mechanisms of radionuclides to colloids was also based on laboratory tests and field observations. The assumption that all radionuclides associated with colloids produced from corroded DHLW glass were irreversibly attached is conservative and is based on numerous direct observations. (CRWMS M&O 2000b). The assumption that radionuclide attachment to colloids produced from degradation of CSNF is reversible is also based on numerous direct observations, in which no embedded phases of the sort observed in the DHLW glass-derived colloids were detected (CRWMS M&O 2000b).

Test runs of the abstraction using GoldSim are planned in order to help assess code verification, model sensitivity, and confidence in model results. The test run results may be presented in a subsequent revision to this AMR or other vehicle.

It is requested that the mass balance used in the abstraction be an output value during the execution of the colloid source term abstraction in TSPA. The mass balance value will be useful in assessing alternative models for colloid transport.

NRC IRSR issues and comments, along with resolution status, are listed in Table 3.

Table 3.   NRC IRSR Issues

Source	Issue	Resolution
NRC (1998b, p. 49)	DOE should specifically determine colloid contribution to actinide release from four types of colloid formation processes: condensation, detachment of secondary phases, sorption onto existing colloids, detachment of WF fragments.	This abstraction addresses the lack of condensation (true) colloids which may be expected in a failed waste canister, as well as sorption onto existing colloids. Detachment of secondary phases and waste form fragments is addressed in the alternative abstraction, which is impractical at this stage of TSPA development.
NRC (1998b, p. 50)	DOE should demonstrate the validity of its empirical correlation for colloid stability versus pH and ionic strength over a wide range of evolving environmental conditions on the bases of mechanistic interpretations.	This has been accomplished in the abstraction by taking existing data on smectite concentration and smectite and iron-(hydr)oxide stability as a function of ionic strength and pH, bounding the data, and developing simplified relationships useful in the abstraction.
NRC (1998b, p. 50)	The influence of corrosion products generated by steel components on the formation of colloids needs to be examined by DOE.	This has been incorporated as an assumption. Quantitative estimates of corrosion products and colloids are not currently available.
NRC (1998b, p. 53)	DOE should develop appropriate scenarios and estimations of colloid transport through package perforations.	In this abstraction all colloids are assumed to exit the failed canister by whatever perforation is present.
NRC (1998b, p. 54)	DOE should consider higher RN releases from glass due to colloids and pulse-release of colloidal RN's from hydrated glass.	Detachment of secondary phases and waste form fragments is addressed in the alternative abstraction, which is impractical at this stage of TSPA development.
NRC (1998b, p. 56)	DOE should account for large excursions in glass corrosion behavior due to spallation of colloids.	Detachment of secondary phases and waste form fragments is addressed in the alternative abstraction, which is impractical at this stage of TSPA development.
NRC (1999b, p. 23)	DOE has identified and considered likely processes for SNF degradation... [including] dissolution of the irradiated UO_2 matrix, with...formation of secondary minerals and colloids.	Colloid formation from degradation of SNF was considered by evaluating the test data from Argonne National Laboratory and incorporating the results into the abstraction.
NRC (1999b, p. 25)	DOE has identified and considered likely processes for degradation of HLW glass... [including] dissolution of the primary phase [and] formation of secondary minerals and colloids.	Colloid formation from degradation of DHLW glass was considered by evaluating the test data from Argonne National Laboratory and incorporating the results into the abstraction.
NRC (1999b, p. 46)	...resolution [of Subissue 3]...is achieved through examination of [among other things] the consistency of the information with evidence generated by studies in other countries.	Studies from other countries were not explicitly considered in the abstraction.

Source	Issue	Resolution
NRC (1999b, p. 55)	...it has been recommended that DOE determine colloid contribution to actinide release [from SNF]...in PA calculations.	Colloid contribution to actinide release was considered by evaluating the test data from Argonne National Laboratory and incorporating the results in the abstraction.
NRC (1999b, p. 59)	DOE has not conducted any relevant scale-down test to evaluate the location, morphology, and characteristics of corrosion penetrations.	Corrosion penetrations were not addressed in the abstraction.
NRC (1999b, p. 60)	DOE should provide confirmation that...radionuclides are not released [from HLW glass] at rates greater than SNF. Otherwise, DOE should consider the effects of [release from the hydrated layer] and...radionuclide release from HLW glass in PA	Release of colloid-associated radionuclides from DHLW glass was incorporated into the abstraction, based on test results from Argonne National Laboratory.
NRC (1999b, p. 62)	It is important that the long-term radionuclide release rate in the corrosion models used by DOE include the influence of stage III behavior [in which reprecipitation of secondary phases occurs, as well as cracking and exfoliation of the altered surface layer].	Release of colloid-associated radionuclides from DHLW glass due to such processes was incorporated into the abstraction, based on test results from Argonne National Laboratory.
NRC (1999b, p. 63)	Recent research...ha[s] shown formation of colloids in the alteration phases...[and] transport of 100 percent of the actinides as colloids to the environment.	Transport of actinides has been treated in the abstraction by considering colloids with irreversibly attached actinides and dissolved actinides available for reversible attachment.

7. CONCLUSIONS

The following conclusions may be drawn regarding this development of an abstraction for the calculation of the in-drift colloids and concentrations in TSPA:

- A useful model has been developed for GoldSim to calculate colloid concentration as a function of ionic strength, as well as for determining the stability of smectite and iron-(hydr)oxide colloids as a function of both ionic strength and pH.

- The abstraction employs bounding relationships that are closely tied to the colloid generation and characterization experimental programs conducted at ANL and LANL and to documented colloid characteristics of a variety of groundwaters.

- The abstraction is conservative, relies on bounding estimates, and incorporates as much realism as is considered defensible.

- The abstraction addresses many stakeholder comments and concerns, such as those presented in the NRC IRSRs.

There are several significant sources of uncertainty attached to this abstraction, some relating to assumptions regarding colloid generation from degradation. The potential formation of colloids from degradation of N-Reactor fuel, and its potential contribution to repository performance, must be investigated. At this time the data are preliminary; however, the program is ongoing, and more data are anticipated. For now, it is assumed in the abstraction that, due to the small quantity of N-reactor fuel, any colloids generated from degradation of the fuel will have little or no effect on repository performance. A plan for verifying this assumption and concomitantly reducing the associated uncertainty, is described in Section 6.1.1.1 of CRWMS M&O (2000b). If this assumption, after examination, proves untrue, use of the assumption could result in

underestimation of the contribution of colloids derived from degradation of N-reactor fuel to repository performance.

Another, but less significant, uncertainty is the assumed concentrations of colloids in groundwater. Data from a range of groundwaters were used in an attempt to establish a fundamental relationship between colloid concentration and ionic strength. The large amount of scatter in the data were accommodated by bounding the data. As a result, colloid concentrations in some circumstances may be overestimated. This, however, is conservative.

The abstraction is considered valid and usable in TSPA calculations for any time after the temperature in the repository has decreased to well below boiling after the thermal pulse. Many of the waste degradation tests were performed at 90°C but mostly sampled at near room temperature. Therefore the test results may be applied to drift processes during the post-thermal period. The range of ionic strength and pH for which colloid masses and stability are calculated in the abstraction are within the ranges anticipated from in-drift chemistry calculations and abstraction.

In general, the bounding relationships employed in the abstraction incorporate uncertainty present in the data used. Additional uncertainty may result from GoldSim and the ways in which GoldSim performs calculations. For example, the choices of distributions and the method of sampling a particular distribution may result in uncertainties in determination of colloid concentrations, ionic strength, pH, and radionuclide concentrations.

Data used directly as input were taken from Liang and Morgan (1990) and Tombacz et al. (1990) and are currently TBV, awaiting approval as accepted data.

This document may be affected by technical product input information that requires confirmation. Any changes to the document that may occur as a result of completing the confirmation activities will be reflected in subsequent revisions. The status of the input information quality may be confirmed by review of the Document Input Reference System database.

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