Determination of
Radionuclide Sorption
for Assessment of
High-Level Waste Isolation

Technical Position
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Geochemistry Section - Geotechnical Branch
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1.0 INTRODUCTION

1.1 Purpose

This document presents a general approach for estimating radionuclide sorption on solids anticipated in a nuclear repository in support of high-level waste site characterization. It is not intended to prescribe specific methods for radionuclide sorption determinations. Instead, the information is provided to the Department of Energy (DOE) to be used as guidance for preparing detailed plans for radionuclide sorption determinations and submitting appropriate documentation early in the site characterization process.

1.2 Definitions of Radionuclide Sorption and Related Experimental Parameters

! This is a non-definition

Sorption - one or more physicochemical processes, excluding precipitation of stoichiometric (fixed composition) solid phases, in which the radionuclide is removed from a liquid phase by interaction with a solid phase or phases.

Sorption or Desorption Ratio, R_s - the amount of radionuclide on the solid versus the amount in the liquid

Distribution Coefficient, K_d - the sorption ratio determined under equilibrium conditions

Retardation Factor, R_f - the ratio of the velocity of the liquid to that of the radionuclide

Sorption Capacity - the maximum amount of radionuclide sorbed on a unit mass of solid. pH dependent! Cation exchange capacity would be a better Term

1.3 Use of Sorption

When a ~~liquid~~ ground water ~~is~~ flow~~ing~~s through ~~permeable solid media~~ rocks and soils, sorption processes act to retard the migration of the solute relative to the water ~~liquid flow~~. ~~The mobility of radionuclides depends, in part, on whether they are strongly sorbed~~. Radionuclide sorption experiment~~ation~~s can be used to estimate ~~this~~ retardation and~~, thus,~~ to quantify two aspects of repository performance. First, sorption experiments can be used to help screen for "key" radionuclides which are defined here as those radionuclides with significant activities that are both highly toxic and mobile, ~~and which occur in significant quantities in the nuclear waste repository~~. Second, sorption studies can ~~also~~ be used to determine the relative abilit~~y~~ies of the ~~subsurface~~ proposed repository sites ~~(the engineered system and the host rock out to the accessible environment~~) to isolate radionuclides. Parameters such as sorption or desorption ratios, sorption capacities, and retardation factors derived from these studies can be used to quantify the ability of the subsurface repository to retard radionuclide migration.

(redundant)

already defined in 10CFR60

redundant

1.4 Regulatory Framework

Three Federal agencies have major roles in the national program for disposal of high-level radioactive wastes. The EPA has developed a generally applicable environmental standard which serves as the overall performance objective for releases from high-level waste disposal. The NRC will develop and issue regulations which cover all aspects of high-level waste disposal, and which will implement the EPA standard. The DOE has the lead responsibility for formulating national policy for disposal of HLW, and has determined that national policy should focus on disposal of HLW in mined geologic repositories. Further, DOE is responsible for constructing and operating a waste disposal facility in accordance with NRC regulations. The NRC will consider DOE license applications for HLW disposal to determine whether the proposals will conform to the regulation.

Wrong - Congress has determined national policy in the Nuclear Waste Policy Act!

2.0 BACKGROUND

As defined by 10CFR60 A geologic repository controls the rate of radionuclide release to the accessible environment by means of two ~~major~~ subsystems: (1) the geologic setting; and (2) the engineered system. The geologic setting (site) is selected for its geologic, hydrologic and geochemical attributes that enhance wrong - The present sites were chosen primarily for political reasons. ~~B~~ WIPP, NTS and Hanford are on Federal land designated for nuclear activities.

radionuclide isolation. It is the responsibility of the DOE to decide when and how much credit ~~will be~~ to taken for sorption to meet NRC and EPA criteria for radionuclide release. ~~Sorption need not be considered if other features of the repository are adequate to meet the criteria.~~ The EPA and NRC criteria do not mandate consideration of sorption. If, on the other hand, sorption is to be considered, input of radionuclide sorption parameters to performance assessment models is necessary. Radionuclide sorption parameters applicable to the nuclear waste repository system are difficult to determine precisely because future geochemical conditions cannot be known with complete certainty. This However, by determining sorption parameters experimentally using site-specific phases and conditions, and applying a safety factor, if necessary, it should be possible to make reasonable estimates of sorption in the subsurface repository.

no meaning

Poor Policy

No such thing exists, 10 CFR 60 does not have a definition of "safety factor"

2.1 Experimental Approaches for Sorption Determination

Determining sorption will be very difficult if the groundwater flow path is unknown! as for BWIP at present

In general, sorption experiments can be subdivided into two types: 1) closed or static systems; and 2) open or dynamic systems (NEA, 1983; McKinley and Hadermann, 1984). Both approaches have been used to describe repository performance. For characterizing sorption phenomena, closed-system experiments, such as batch sorption tests, involve contacting radionuclide-free (or deficient) solids with a radionuclide-bearing solution for the duration of the experiment followed by analytical determination of the sorption ratio, R_s. Batch desorption experiments, on the other hand, involve contacting radionuclide-free (or deficient) liquid with radionuclide-bearing solids, followed by measurement of the quantity of radionuclide leached. Open-system experiments, such as flow-through column tests, involve the introduction of fluid at one end of a reaction vessel containing solid and the removal of the fluid at the other end. The solid material sorbs solute and, as a result, retards the migration of the solute relative to that of the liquid, expressed as a retardation factor, R_f.

not necessarily

no one could run the experiment with competing radionuclides already adsorbed onto the sediment

There are advantages to both experimental approaches. For example, the advantages of the closed-system experiments are that they are relatively simple & cheap to carry out and the residence time of the solution in contact with the solid can be greater than in a open-system experiment. The longer residence times may more closely simulate those in a nuclear waste repository. On the other hand, the advantages of the open-system experiments are that they may better

Not very accurate

A column test can be run for months. Either way it could be done. And the flow rate could approach zero

Rock-water ratios are very different in batch tests than in nature model radionuclide migration in flowing systems by revealing the presence of multiple speciation, mass action competition, colloids or particulates that the closed-system experiment (batch test) might miss. (Kelmers, 1984).

NO! There are ways that sorption can be evaluated in the field. The NRC should not preclude Field methods or natural analogues.

3.0 STATEMENT OF POSITION

It is the position of the NRC that sorption parameters used in performance assessment calculations should be derived experimentally. The DOE sites should:

1) Develop a tentative matrix of experiments that involves radionuclides and starting materials based on the anticipated range of proportions and compositions of phases under the various physicochemical conditions expected in the subsurface repository;

2) Characterize solid and liquid reactants and products;

3) For closed-system experiments, determine sorption isotherms by varying radionuclide concentrations up to an apparent concentration limit;

4) Determine the applicability of sorption parameters to repository performance by using various experimental approaches including both open and closed laboratory systems, in-situ field tests, and natural analogues; and

5) Document the magnitudes of experimental and conceptual uncertainties from all anticipated sources.

4.0 DISCUSSION

Details on the individual points from the Statement of Position, along with a discussion on why the NRC staff thinks these points are important, are given below. It is the responsibility of the DOE to demonstrate that when these parameters ~~(plus some safety factor, if necessary)~~ are used in performance assessment calculations, the radionuclide migration is not underestimated.

!! undefined & undeterminable!

Failure to consider and ~~evaluate~~ implement the points in the Statement of Position could make it difficult for the DOE to demonstrate ~~provide reasonable assurance~~ that the sorption parameters are appropriate for characterizing the site.

4.1 Matrix Development

A matrix of experiments should be developed as a planning tool for characterizing the sorption properties of a subsurface repository. Variables such as solid composition and texture, liquid composition, proportion of phases, temperature, pressure, particle size, flow rate and regime (porous and fractured media), time, and ionizing radiation should be considered in the matrix.

Initially, radionuclide studies can be prioritized by comparing EPA and NRC criteria to radionuclide inventories in the repository. Some radionuclides may occur in low enough quantities that, if they meet NRC release rate requirements, they will not contribute significantly to exceeding the EPA standards. These may be assigned a lower priority than those radionuclides whose cumulative releases over 10,000 years are likely to exceed the EPA standards in the absence of sorption effects.

It is recommended that the matrix include scoping experiments, performed early in the experimental program, which involve relatively simple systems (few components). These simple system experiments might be useful in determining the effects of various physicochemical conditions on sorption. Following the scoping experiments, the matrix development should reflect combinations of the above parameters that simulate physicochemical conditions and phase assemblages likely to release radionuclides to the accessible environment. Consequently, the size of the matrix would be greatly reduced by first considering the dependence or interrelation of phases and conditions upon each other and deleting incompatible combinations.

The NRC staff considers it important that the DOE develop a matrix for planning sorption experiments. The DOE can then effectively demonstrate its rationale for choosing some combinations of parameters for study and eliminating other combinations as inappropriate. Without a matrix, some crucial experiments that

The DOE does not demonstrate with reasonable assurance. The licensing process establishes reasonable assurance.

characterize radionuclide migration may be overlooked. As a result, the DOE might not be able to demonstrate ~~with reasonable assurance~~ that the derived sorption parameters are appropriate.

Apparently, the NRC will be using the U.S.G.S spelling ground water or ground-water depending on use.

4.2 Characterization of Reactants and Products

The DOE should provide proof that crushed solids are relevant to repository conditions should they chose to use crushed rock in sorption experiments

In choosing appropriate solid reactants for sorption studies, emphasis should be placed on the identification and characterization of solids including waste form, canister, backfill, seals, packing, and host rock primary and secondary phases occurring along paths the radionuclide-bearing groundwater will take as it flows away from the waste. These are the solids most likely to react with groundwater and thereby affect radionuclide concentrations and release rates. Characterization of the solids should include chemical, mineralogical, textural, and particle size determinations. The applicability of crushed solids in sorption experiments to repository conditions should be addressed. It is possible that the surfaces of crushed material are significantly different from the surfaces of intact material, both porous and fractured. Grinding may expose the surfaces of solid phases different from those which groundwater would contact in a repository and/or may change the reactivity of the same mineral surfaces with dissolved radionuclides.

Stronger wording needed see above

poor word choice

Similarly, the range of groundwater compositions expected in a repository system should be considered in selecting liquid reactants. Generally, in the rock-dominated environments of a high-level waste repository, groundwater compositions ~~can~~ will be affected by reactions with the rock. ~~solids at various temperatures and pressures.~~ Consideration of the range ~~in~~ of water compositions used in experimentation should be based on the range of compositions of analyzed groundwaters at ambient conditions, the range of compositions calculated from solid assemblages assumed to have equilibrated with the groundwater, and the range of groundwater compositions experimentally determined at elevated temperatures and pressures.

Question is vague

The applicability of synthetic starting materials to the conceptual model employed in developing the matrix should be addressed. Failure to do so might result in experiments that do not adequately simulate repository conditions. For example, the preparation of radionuclide-bearing groundwater commonly involves the addition of a small amount of acidified tracer to a synthetic solution simulating the natural groundwater. The resulting solution may neither be representative of repository conditions nor be stable. Kelmers et al., (1985) found that in a sorption experiment in which a synthetic groundwater was tagged with an acidified uranium solution, more uranium was subtracted from the control(liquid only) than from the test (liquid + solid) resulting in a negative sorption ratio. This indicates that the synthetic groundwater was unstable and inappropriate for modeling repository conditions.

In addition to characterizing the reactants, it is also important to characterize the experimental products. Following the experiment, analysis of the liquid products should include the determination of major, minor, and trace element concentrations, along with pH and redox conditions.

The extent of sorption of some dissolved radionuclides on engineered barrier materials and host rock can be strongly dependent on the redox potential (Eh) and acidity (pH) of the groundwater. For example, Benjamin and Leckie (1981) show that the sorption of Cd, Cu, Zn, and Pb on amorphous iron oxyhydroxide is strongly dependent on pH. The percentage of cation sorbed varies from approximately zero to one hundred with a change in pH of two units. Likewise, Kelmers et al., (1984) have shown that sorption ratios for neptunium and technetium are dependent on the redox condition of the system.

The characterization of solid products from sorption experiments is important because, for example, under the same physicochemical conditions, different solid phases can have drastically different sorptive capacities for the same radionuclide. Characterization of the solids is important in determining which reactions took place and how these reactions depend on experimental technique. In addition to determination of the composition of individual solid phases, characterization should include surface area and/or particle size measurements. Because sorption is predominantly a surface phenomenon, the surface area of the solid may strongly affect the experimentally determined sorption parameters. For example, neptunium sorption ratios increased two orders of magnitude as particle diameter decreased from 200 to 2 μm(Kelmers et al., 1984).

4.3 Isotherm Development for Closed-System Experimentation

Probable release scenarios call for radionuclide concentration gradients in the repository system. Under equilibrium conditions, the concentrations of radionuclides in the repository can range from zero to an apparent concentration limit. Under equilibrium conditions, the apparent concentration limit is the greatest radionuclide concentration the liquid can maintain when the temperature, pressure, and moles of all other components in the liquid, n_j, are held constant. The apparent concentration limit is controlled by the solubility of some stoichiometric radionuclide-bearing solid phase. Figure 1, a generic sorption isotherm, illustrates the relationship between concentration on the solid versus concentration in the liquid when all other parameters are held constant. Analysis of the liquid product can assure the constancy of the other parameters. Although this figure shows a linear sorption region, many sorbed species, including radionuclides, show nonlinear relationships between the quantity sorbed and the solution concentration. Thus, sorption ratios are dependent on solute concentration (Serne and Relyea, 1982).

Because radionuclide concentrations are expected to vary in the repository and sorption parameters are concentration dependent, the ~~NRC staff considers it~~ necessary ~~reasonable to design~~ experiments should be designed to determine the effect of concentration on sorption ratios. Sorption isotherms should be determined up to an apparent concentration limit. Experimentally, it should be possible to determine an apparent concentration limit of a radionuclide in liquid in contact with solid. For example, at the same temperature, pressure, and n_j in the liquid, two sorption experiments with different concentrations of the same radionuclide in the liquid starting material should yield the same radionuclide concentration in the liquid products at the apparent concentration limit.

4.4 Determination of Sorption Parameters by Multiple Experimental Approaches

True QA would make it necessary

DELETE

~~If a sorption experiment could be designed that simulated all anticipated repository conditions, it would not be necessary to use multiple experimental approaches to determine sorption parameters. However, simulation of all anticipated repository conditions in sorption experimentation would be difficult and/or impractical. The fact that some parameters or conditions cannot be bounded requires the extrapolation of these conditions to those expected in the repository. This extrapolation introduces uncertainty into the~~

How about a scientific mechanistic approach to sorption? Then sorption would be predictable with changes in chemistry!

This section is not insignificant it is simply out of place

DELETE

modeling of sorption parameters. Therefore, multiple approaches can lend support to, and reduce the uncertainties of, experimental results from studies in which some parameters are not site specific. Some experimental parameters can be varied over a large enough range as to bound the conditions anticipated in the repository. These parameters include surface area/volume ratio, (SA/V), temperature, pressure, composition, and flow rate. Other parameters that often are not duplicated in the laboratory are scale, residence time, water/rock ratio, and flow characteristics, which can include saturated versus unsaturated flow and porous media versus fractured flow.

Laundry list — move it back into the discussion

Experiments are (should be) designed so that measureable effects of physicochemical reactions can be monitored in a reasonable time. At the relatively low temperatures anticipated in the repository, chemical reactions involving geologic materials can be extremely slow. In order to accelerate these reactions so that changes are measureable in experimental time, conditions other than those anticipated in the nuclear waste repository are sometimes imposed on the experimental system. For example, experiments have employed crushed solid material, high concentrations of solutes, agitation, catalysts, rapid flow rates, and elevated temperatures. — Some theory is needed to relate those conditions to repository conditions

In addition to accelerating reaction rates, laboratory experiments are designed so that the amount of material can be handled reasonably. By scaling down systems of interest (repository size) to laboratory size, certain physical conditions must be altered. For example, the water/rock ratio in most repository systems is significantly less than one. However, in order to obtain enough water for analysis in laboratory experiments, the W/R ratio is ordinarily increased significantly. This technique makes the bulk chemistry of the experimental system different from that in the repository. The proportions of phases in experiments has been shown to affect radionuclide sorption parameters (Meyer, 1983; Rafferty et al., 1981; Meier et al., 1982). Thus, the effect of this technique on sorption parameters should be considered. One can argue that in a fractured media, with little porosity, most of the rock will not be in contact with the groundwater. Consequently, water/rock ratios used in experimentation should be higher than those that take into account all the rock in a repository system. If this argument is used, however, it follows that the solid reactants should be predominantly fracture material and not bulk

Rewrite to move back with emphasis on water/rock ratio

What is the point of this discussion? No guidance here folks.

START SECTION HERE

rock. Sorption experiments involving crushed bulk rock might have little applicability to sorption phenomena in fractured media.

To verify the ~~applicability~~ quality of experimentally determined sorption data ~~parameters to a repository system,~~ the ~~site~~ applicant should use multiple experimental approaches. [This approach was a recommendation of the WRIT Program (Serne and Relyea, 1982).] Using this approach, sorption parameters can be analyzed and compared. For example, the sorption ratio, R_s, obtained from batch experiments has often been used to calculate a retardation factor, R_f. The relationship between R_s and R_f is taken to be

multiple experimental approaches will not insure that experiments are relevant only that the data are consistent

Delete:- This is a distractor

$$R_f = 1 + \rho R_s(1 - \phi_e)/\phi_e$$

where ρ is the bulk density of the rock, and ϕ_e is the effective porosity. This relationship is based on ion exchange theory as applied to porous media flow. However, due to the variety of processes that contribute to sorption, the calculated R_f value may not equal the measured R_f value determined from a flow-through column experiment.

Comparison of the sorption and desorption parameters obtained from closed-system and open-system experiments is recommended. Generally, the sorption parameters derived from closed-system experiments are equal to or greater than those derived from open-system tests using the same solid material (NEA Workshop, 1983). As a result, closed-system tests may overestimate the effectiveness of a repository system to isolate radionuclides (Relyea, et al., 1980). The difference in sorption ratios may be due to particle abrasion in stirred closed-system experiments or the relatively short residence times in open-system experiments (NEA Sorption Workshop, 1983). Other factors that can cause a discrepancy between the sorption parameters from open and closed systems are the presence in the liquid of multiple radionuclide species, colloids, and particulates. Changes in physicochemical parameters such as temperature, fluid velocity, radionuclide concentration, and fluid composition may shed some light on the causes of the discrepancy between the two types of systems.

Sorption experiments on crushed porous material may be inadequate because mineral surfaces may have been altered mechanically.

Extrapolation of sorption parameters from laboratory experiments to a large-scale, long-term repository system can be highly uncertain. The flow characteristics of the groundwater can have a drastic effect on the applicability of laboratory-derived sorption parameters to repository performance. Most experiments use crushed material as a solid medium because it is easy to handle and characterize, and accelerates solute-solid reactions. ~~The application of crushed material~~ to intact porous media may not ~~be adequate but not so when the natural system is a~~ especially for fractured rocks. Sinnock et al., (1984) and Nuttall and Ray, (1981) have calculated that rates of radionuclide migration via fracture flow can be two orders of magnitude greater than that via porous media flow. Thus, for performance assessment calculations, consideration of flow regime can be of the utmost importance.

Crushed rock experiments are very dissimilar to the fracture flow environment.

If groundwater flux in a repository is predominantly via fracture flow, sorption tests in the laboratory may not adequately simulate repository conditions. One method of further reducing the uncertainty caused by the inadequate simulation of various flow characteristics could be to perform in situ tests on site-specific solid material (Serne and Relyea, 1982; Abelin et al., 1984; Neretnieks et al., 1982). The scale of these tests can be larger than that of the experimental tests but smaller than that of the repository. Furthermore, the in situ solid materials would probably not have suffered the effects of handling (grinding, sieving, washing) required in laboratory tests. Time constraints~~, however, would~~ still apply in these experiments but time constraints can be evaluated by assessing sorption and desorption of natural uranium series radionuclides. Comparison between the laboratory and field results can illustrate the usefulness of the different approaches. However, the physicochemical conditions must be carefully controlled in the in situ tests to ensure a parallelism in the approaches. Therefore, DOE should ~~consider~~ perform~~ing~~ in situ tests ~~for comparison with laboratory tests, in order~~ to reduce the uncertainties ~~of application to more realistic repository~~ systems. in predicting repository performance.

Although field tests can expand spatial scale over that which is normally handled in a laboratory, the time scale is still several orders of magnitude less than that of a repository. ~~The study of~~ Natural analogues ~~is encouraged, since such~~ studies are needed to understand the effects of time ~~possibly can shed some light~~ on the migration of radionuclides in the repository ~~systems~~ that have existed for long periods of time. The natural analogue should be demonstrably equivalent to some particular process anticipated

~~present~~ in the repository and have well-defined boundary conditions. ~~Common~~ Examples of systems used as natural analogues are ore bodies such as Oklo (Brookins, 1978) and the uranium deposits in the Northern Territories of Australia (Airey, 1983). Igneous intrusives have also been studied for they simulate anticipated thermal histories and alteration patterns. [illegible] and Lamb (1983) has demonstrated that information on sorption kinetics can be measured directly at a proposed repository site by means of measuring natural thorium, uranium and radium isotopic activities in solution and on solid phases.

4.5 Documentation of Uncertainties

~~Sources of~~ Uncertainty stems from failure to duplicate anticipated repository conditions and incorrect experimental procedures and results. The failure to duplicate repository conditions can be caused by an incorrect understanding of the conditions, an inability to duplicate the conditions or an ~~inadvertent~~ improper experimental design. Incorrect experimental results can result from imprecise data or misinterpretations of the data. The uncertainties of sorption studies can be minimized by using multiple techniques to determine repository conditions, analyses to bound adverse impacts, and multiple experimental methods.

Misstated - Incorrect experimental procedure causes incorrect experimental results

Incorrect interpretations result from missing data -

The NRC staff recommends that the DOE consider the impact of the error and uncertainty on the sorption? result, and the effect of errors and uncertainties in sorption ~~result~~ on ~~the~~ repository performance. Subsequently, the DOE should focus on the uncertainties which have the most impact on repository performance.

which? sorption or performance assessment?

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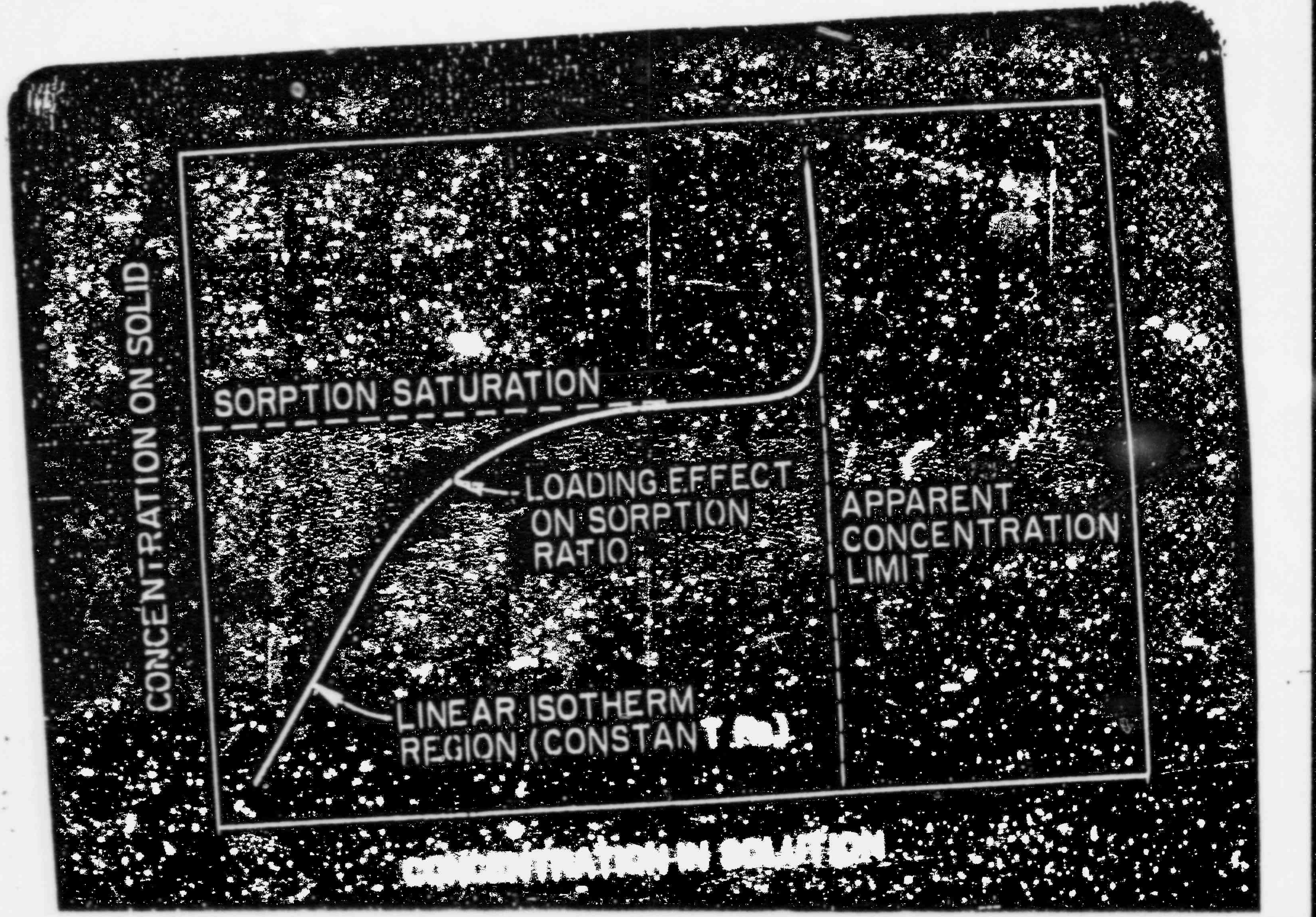
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