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Novel Sorbent Development and Evaluation for the Capture of Krypton and Xenon from Nuclear Fuel Reprocessing Off-Gas Streams

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ABSTRACT

The release of volatile radionuclides generated during Used Nuclear Fuel reprocessing in the US will most certainly need to be controlled to meet US regulatory emission limits.

A US DOE sponsored Off-Gas Sigma Team has been tasked with a multi-lab collaborative research and development effort to investigate and evaluate emissions and immobilization control technologies for the volatile radioactive species generated from commercial Used Nuclear Fuel (UNF) Reprocessing.

Physical Adsorption technology is a simpler and potential economical alternative to cryogenic distillation processes that can be used for the capture of krypton and xenon and has resulted in a novel composite sorbent development procedure using synthesized mordenite as the active material.

Utilizing the sorbent development procedure, INL sigma team members have developed two composite sorbents that have been evaluated for krypton and xenon capacities at ambient and 191 K temperature using numerous test gas compositions. Adsorption isotherms have been generated to predict equilibration and maximum capacities enabling modeling to support process equipment scale-up.

I. INTRODUCTION

Commercial Used Nuclear Fuel (UNF) reprocessing results in the release of volatile radioactive gases that evolve into facility off-gas systems. These gases are typically composed of primary fission product isotopes including krypton (^{85}Kr), iodine (^{129}I), tritium (^3H), carbon (^{14}C) and multiple xenon (Xe) isotopes. Although Xe isotopes have very short half-lives (~35 days) the capture of Xe generated from fuel irradiation may have commercial economic value. The removal of Xe upstream Kr also provides the added benefit of increasing Kr capacity due to competition between Xe and Kr for sorbent capture sites. The resultant radioactivity emissions released in off-gas streams are regulated by the U.S. Environmental Protection Agency. These regulations address the Fuel Cycle (40 CFR 190.10), NRC Licensees (10 CFR 20.1101, .1301, .1302, App. B) and DOE Facilities (40 CFR 61.92) by establishing dose limits resulting from nuclear fuel cycle activities in the

commercial sector (Ref. 1-3). ^{85}Kr will most likely require capture prior to off-gas release in order to satisfy these requirements.

An Off-Gas Sigma Team was formed in late 2009 with a purpose to integrate and coordinate research and development activities associated with the capture and immobilization of the volatile fission gases produced from nuclear fuel reprocessing activities. The Sigma Team concept was to assemble a multi-disciplinary team composed of members from various national laboratories in the DOE complex. Current Off-Gas Sigma Team laboratory members are: Argonne National Laboratory (ANL), Idaho National Laboratory (INL), Oak Ridge National Laboratory (ORNL), Pacific Northwest National Laboratory (PNNL) and Sandia National Laboratory (SNL) (Ref. 4).

INL Sigma Team members are investigating physical adsorption techniques for use in potential Kr capture processes. The purpose of this paper is to describe the development and evaluation of hydrogen and silver mordenite composite sorbents for the capture of Kr and Xe from various simulated nuclear fuel cycle process off-gas compositions.

II. SORBENT DISCUSSION AND DEVELOPMENT

Physical adsorption and cryogenic distillation are two candidate processes that can be used to capture volatile components from UNF reprocessing off-gas streams. Cryogenic distillation has been used at plant scale however, due to the associated high costs and process complexity, the physical adsorption process has been selected for further evaluation for the capture of Kr and Xe from off-gas streams. Physical adsorption processes can be operated in either temperature swing adsorption (TSA) or pressure swing adsorption (PSA) operations or a combination of both. However, isotherms generated from historical physical adsorption work suggest the sorption capacity for Kr is linearly dependant on pressure swing operations but exponentially dependant on temperature swing operations (Ref. 5). Hence investigations using thermal swing operation are being conducted.

The sorbent material selection for this work included an investigation into various synthetic zeolites reported to have proper cage structure and frameworks that result in surface area and pore size conducive for Kr capture. Activated charcoal is known to be a material that is capable of capturing Kr and Xe at reduced temperatures,

however reports of explosion incidents caused by the reaction of NO_x and charcoal at elevated desorption temperatures drove the investigation to alternative materials (Ref. 6). Hydrogen and silver mordenite cation forms were selected based on literature reviews (Ref. 7-14). However, it became evident that the ability to acquire hydrogen mordenite in an appropriately engineered form proved difficult. Silver mordenite (Ionex Ag900) was acquired commercially from Molecular Products, Inc., but the surface area was measured and found to be quite low (30-50 m^2/g). This low surface area was assumed to be a result of the pelletization manufacturing process. Synthesized cation form mordenite powders (Na, H and NH_3) are commercially available and were acquired from Zeolyst International. These powders were used for investigation into the possibility of preparing a composite engineered form material using a macroporous polymer to bind or encapsulate the mordenite powder crystal. A macroporous binder known for thermal and radiolytic stability was selected. Although mordenite powders exhibit high surface areas (500 m^2/g), in powder form their use in process columns would result in potential high differential operating pressure further supplementing the need for an engineered form.

Method development used for sorbent preparation included the use of a solvent in which the polymer binder is soluble. Selected mordenite powders were mixed with the solvent to form a slurry, then heated slightly, before adding the binder. The heated slurry was then thoroughly mixed with the binder to form a homogenous mixture. Using a specially designed glass apparatus, the mixture was dripped into a water reservoir where the liquidus drops formed insoluble solid sorbent particles. The final sorbent particles were then heated to remove residual water and analyzed for surface area and microporosity. A Micromeritics model 2020 ASAP surface area analyzer was used to measure surface area and microporosity of sorbents after preparation. A patent application for this sorbent development procedure has been filed (Ref. 15).

II. A. Experimental Apparatus and Capacity Measurement

In FY-10, a custom built cryostat system was procured and installed at the INL. The system enables sorbent capacity evaluations at temperatures from 150 to 500 K with sorbent bed masses ranging from 2 to 600 grams. The column effluent is monitored with an Agilent Technologies model 7890A gas chromatograph (GC) utilizing a Supelco carboxen 1010 plot fused silica capillary column with a thermal conductivity detector (TCD). Figure 1 includes a photograph of the experimental setup including the cryostat.

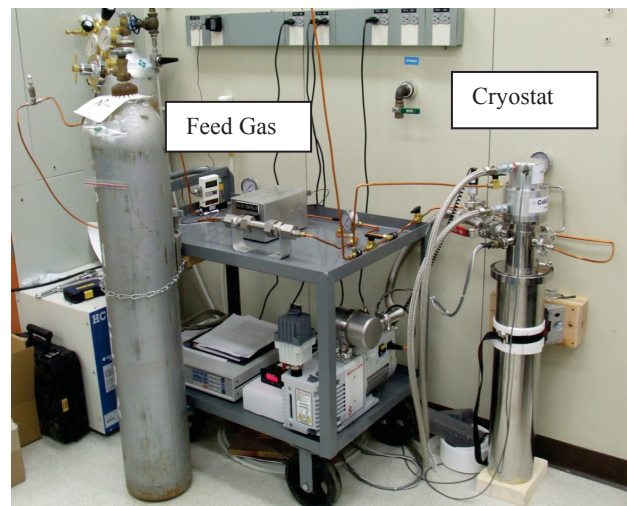


Fig. 1. Photograph of the Kr capture experimental setup.

Breakthrough curves were generated by plotting the concentration of the constituent of interest (C) in the column effluent as a function of the volume (V) of gas processed. The collected breakthrough curves were used to calculate the sorbent capacity (SC) for each constituent in the feed gas. The sorbent capacities were calculated from the following equation:

$$SC = \int_0^V \frac{(C_0 - C)}{M} dV \quad (1)$$

Where:

V = total volume processed at breakthrough, liters

C_0 = concentration in feed, mmol/L

C = concentration in effluent at V, mmol/L

M = sorbent mass, kilograms

Because C_0 and M are constant, Equation 1 can be rewritten as:

$$SC = \frac{1}{M} \left(C_0 V - \int_0^V C dV \right) \quad (2)$$

The statistical software TableCurve®, from Jandel Scientific, was used to find the best-fit for equations representing the breakthrough curves. The software was used to define the area below the breakthrough curve to obtain the value for the last term in Equation 2. This result was then subtracted from the total area $C_0 V$ to give the amount of Kr or Xe adsorbed on the material. The adsorbed amount was then divided by the sorbent bed mass to give a final sorbent capacity in mmol Kr or Xe per kilogram of sorbent.

II.B. Hydrogen Mordenite Evaluations

An engineered form composite sorbent incorporating hydrogen mordenite powder (HZ) was prepared using the procedure described previously. Typical wt% of the active powder in the finalized sorbent particles can range from 60-90% but the target was 80% loading of the hydrogen mordenite powder. Surface area analysis resulted in a Brunauer, Emmett and Teller (BET) surface area of 336 m²/g and a micropore area estimated by the Density Functional Theory (DFT) model of 270 m²/g. Scanning electron microscope photos taken of the HZ composite material included in Figure 2 show the uniformity of sorbent beads and the porosity of the interior of the beads.

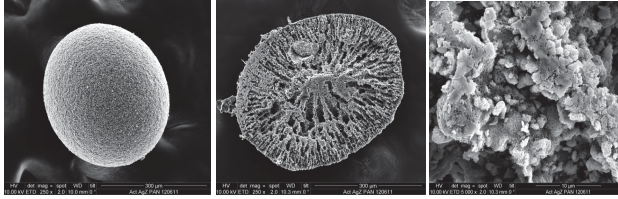


Fig. 2. SEM photos showing a sorbent bead, a bead cut in half and a closeup of the interior of the bead.

The HZ sorbent was evaluated with gases containing various compositions of Kr, Xe, N₂, air, and He at 190 K. The Kr concentration was 150 ppm and the Xe concentration was 1000 ppm with the N₂ at 79% and He as the volume balance for all gas compositions tested to correlate with implicit UNF reprocessing off-gas compositions. Capacities for Kr and Xe were calculated for all gas compositions tested at 190 K and are listed in Table I.

TABLE I. HZ sorbent capacities for selected test gas compositions at 190 K.

Feed gas mixture	Kr Capacity (mmol/kg)	Xe Capacity (mmol/kg)
Kr/He	100	N/A
Kr/N ₂ /He	0.569	N/A
Kr/Xe/He	74.0	1.09e3
Kr/Xe/N ₂ /He	8.28	6.35e2
Kr/Xe/Air	7.41	7.95e2

Kr capacity data calculated for the composite sorbent using the Kr/He gas mixture were slightly higher than the isotherm data capacities reported by Munakata (Ref. 13) for both hydrogen mordenite and activated charcoal. Munakata did perform tests at 195 K and the hydrogen

mordenite used was prepared from a cation mordenite ion-exchanged with hydrochloric acid.

II.B.1 HZ Thermal Cycling

Thermal cycling can have deleterious effects on sorbent performance. High operating temperature fluctuations for numerous sorption/desorption cycles can result in a slow loss of the crystalline structure of the mordenite and decrease the selectivity and capacity. A thermal cycling evaluation was also performed on the new sorbent by performing periodic baseline tests, at one set of operating conditions, sporadically among the 13 capacity and isotherm adsorption/desorption test cycles. The calculated Kr capacities for the baseline test conditions did not decrease suggesting the sorbent thermal stability and cycling assets were validated. At the completion of the capacity evaluations, the sorbent was visually inspected and weighed. The sorbent weight, surface area and microporosity remained unchanged which further validated the robustness of the material to thermal cycling. Table II presents the Kr capacities calculated for baseline sorbent cycling test conditions.

TABLE II. Kr capacities from baseline cycling tests.

Kr capacity (mmol/kg)	% Breakthrough
102	93.8
100	89.5
112	100
100	84.7

II.B.2 HZ Adsorption Isotherms

A final evaluation of the sorbent was to generate Kr adsorption isotherms at 190 K and 220 K. The Kr concentrations ranged from 75 to 2544 ppm. Using the Langmuir equilibrium model written as:

$$q = \frac{q_{\max} K_{\text{eq}} C}{1 + K_{\text{eq}} C} \quad (3)$$

values for maximum capacity at a specified temperature (q_{\max}) and the equilibrium constant (K_{eq}) were calculated from a Langmuir regression of the experimental data. Predicted values for q_{\max} and K_{eq} at 190 K were 0.94 mol/kg and 23.42 m³/mol, respectively. Values for q_{\max} and K_{eq} at 220 K were 0.2 mol/kg and 8.42 m³/mol, respectively. Figure 3 includes the graphed Kr isotherm data for 190 K and 220 K for the HZ sorbent material.

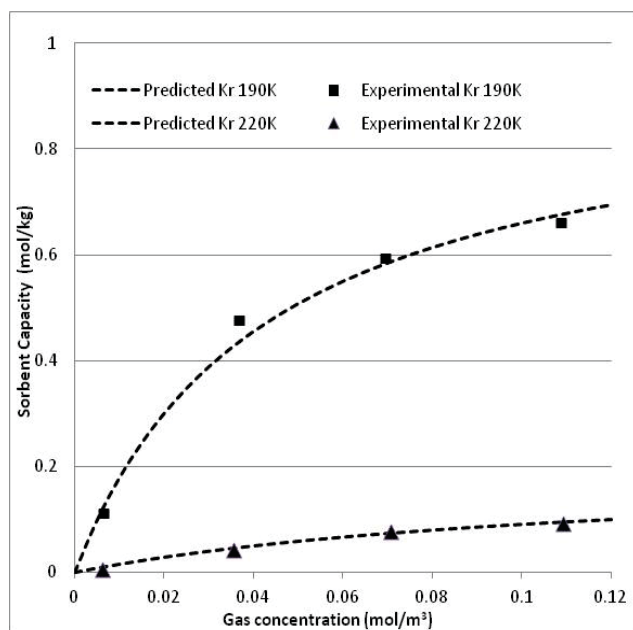


Fig. 3. Kr adsorption isotherms at 190 K and 220 K for HZ sorbent.

It can be seen from the graph that the Langmuir model predicts the adsorption data quite well. The Langmuir parameters can be utilized to develop models for the scale-up and process design calculations for implementation into a future fuel reprocessing off-gas treatment process.

II.C. Silver Mordenite Evaluations

Silver mordenite (AgZ) has been reported to exhibit strong adsorptive properties for Kr and Xe resulting in substantially higher adsorption capacities than other cation form mordenites (Ref. 14,16). An enhanced interaction between Xe and silver has also been discussed (Ref. 17,18). The presence of Xe in UNF off-gas streams is at concentration levels 10 times higher than Kr. The removal of the Xe prior to Kr capture would be beneficial both economically (Xe commercial value) and to enhance Kr capacities due to capture site competition with Xe.

The preparation of AgZ composite sorbent was more tedious than that for the HZ material. A silver mordenite powder form is not available and necessitated a silver exchange conversion procedure using the sodium cation form mordenite (NaZ) powder (Ref. 19). The resultant powder was analyzed for BET surface area using the Micromeritics ASAP 2020 surface area analyzer and the silver content was determined by dissolving a known mass of AgZ powder in a hydrofluoric/nitric acid mixture and analyzed by inductively-coupled plasma mass spectroscopy (ICP-MS) using a Perkin Elmer Elan DRCII Model 6000 instrument. The AgZ powder surface area analysis revealed 374 m²/g and the silver content resulting from ICP-MS analysis was 10.2 wt.%.

An engineered form composite using the AgZ powder was then prepared using the previously described procedure. The resultant surface area of the engineered form sorbent was only 81.9 m²/g. This lower than expected surface area indicated the AgZ final product was not useful for physisorption testing. A literature search revealed that there is a strong interaction of silver with sulfur which is contained in the solvent used in the binding process (Ref. 20,21). Two aprotic polar solvents, dimethylacetamide (DMAC) and 1-methyl-2-pyrillidinone (1M2P), were selected as candidate alternatives based on low toxicity and lack of sulfur in their composition. Two sorbents were prepared with the converted powder using each candidate solvent. The beads resulting from the prepared sorbents were characteristically more brittle than those made with the original solvent and would easily collapse into a powder by applying finger pressure. BET measurements revealed very low surface area for these sorbents signifying that the use of the alternative solvents would not achieve desired results, thus they were abandoned.

Following a thorough process of elimination involving investigating characteristics of the macroporous binder, silver converted powder, alternative solvents, and the overall preparatory process, the option to simply rearrange the sorbent preparation procedure was investigated. The rearranged procedure calls to first bind the NaZ powder in the binder followed by converting the NaZ composite with silver nitrate solution with very mild mixing by argon sparge at 95 °C for 2 days. A sample of the AgZ composite engineered form prepared in this manner was dissolved in a hydrofluoric/nitric acid solution and submitted for ICP-MS analysis. The analysis results indicated that this procedure resulted in a final silver concentration in the final material of 8.1 wt%, which supports a nearly complete conversion of all sodium to silver. The final surface area was measured to be 250 m²/g, which is acceptable for use in the planned experiments. The AgZ composite engineered form material was sieved to a particle diameter range between 0.425 and 1.40 mm and used for all testing at ambient and 191 K temperatures.

Prior to testing, the sorbent was activated at 125 °C for 16 hours with helium flow. The activation was performed to ensure the residual water vapor was removed from the pores, increasing availability of capture sites.

Following the initial activation, AgZ material evaluations at ambient temperatures were performed. Various feed gas compositions were tested to measure capacity for Xe and Kr. Results from the testing indicated no measurable Kr capacity, thus Kr results are not reported. Table III incorporates the gases tested with respective Xe measured capacities. All test gases used in

HZ testing were also used for AgZ testing. A gas flow of 50 sccm equating to a superficial velocity of 1.2 cm/s was used.

TABLE III. Measured AgZ sorbent capacities for various gas compositions at ambient temperature.

Feed gas mixture	Temp. (± 1 K)	Xe Capacity (mmol/kg)
Xe/He	296	31.5
Kr/Xe/He	295	31.6
Kr/Xe/N ₂ /He	297	8.53
Kr/Xe/Air	296	27.8

In general, the Xe capacity results at ambient temperature were very similar for all test gases used, except the Kr/Xe/N₂/He. It appears that the presence of oxygen with 79% nitrogen may influence the silver crystalline structure impacting i.e. increase, capacity when compared to gas compositions with nitrogen only. It is important to note that the addition of both nitrogen and oxygen did not significantly impact the Xe capacity of the AgZ at ambient temperature. These data suggest that AgZ can be used as a Xe polishing column at room temperature prior to Kr capture.

A series of tests was performed with the AgZ sorbent at 191 K. This test series utilized the same test gases used for ambient temperature testing with the addition of a Kr/He gas mixture. The results of this testing are included in Table IV below.

TABLE IV. Kr/Xe capacities for AgZ sorbent at 191 K.

Feed gas mixture	Kr Capacity (mmol/kg)	Xe Capacity (mmol/kg)
Kr/He	65.8	n/a
Xe/He	n/a	756
Kr/Xe/He	51.3	864
Kr/Xe/N ₂ /He	8.33	429
Kr/Xe/Air	5.25	468

As expected, the Xe capacities at 191 K were over an order of magnitude higher than capacities measured at ambient temperature (Ref. 5). As was found with the ambient temperatures, the addition of air did not have a significant impact on Xe capacities at 191 K. However, the addition of air did decrease the Kr capacity by an

order of magnitude. Also, when comparing the Kr capacity for the Kr/He gas composition to the Kr/Xe/He gas composition, a Kr capacity decrease of about 30% was measured when Xe is present. As found in the literature (Ref. 14), Xe capacity is an order of magnitude higher than Kr capacity when testing Xe/He and Kr/He gases and 50 to 80 times higher with gases where N₂ and O₂ are present.

II.C.1 AgZ selectivity of Xe relative to Kr at 191 K

The selectivity of Xe relative to Kr was calculated for tests where both Xe and Kr capacities were measured using the following equation:

$$\text{Sel}_{\text{Xe,Kr}} = \frac{X_{\text{Xe}}/Y_{\text{Xe}}}{X_{\text{Kr}}/Y_{\text{Kr}}} \quad (4)$$

Where X_{Xe} and X_{Kr} are mol fractions of Xe and Kr in the adsorbed phase, and Y_{Xe} and Y_{Kr} are mol fractions of Xe and Kr in the bulk phase. The Xe selectivities relative to Kr for selected test gases are shown in Table V. Selectivities could only be calculated for tests performed at 191 K, since there was no measurable Kr capacity at ambient temperature.

TABLE V. AgZ selectivity of Xe relative to Kr at 191 K.

Feed Gas composition	Selectivity Xe relative to Kr
Kr/Xe/Air	13.6
Kr/Xe/N ₂ /He	7.8
Kr/Xe/He	2.5

The selectivity of Xe relative to Kr with the feed gas composed mostly of air is over five times higher than selectivity of gas absent of N₂ and O₂. This would suggest the presence of air more impacts the Kr capacity than the Xe capacity. This would substantiate the possibility of using an ambient temperature polishing column containing AgZ sorbent upstream of a Kr specific sorbent such as HZ. However, more work should be completed in this area to determine the impacts of N₂ and O₂ on the Kr capacities in the final column and if the same selectivities could be achieved at higher test temperatures.

II.C.2 AgZ Thermal Cycling

Intermittently, baseline tests using the Kr/Xe/He feed gas were performed on the sorbent at ambient temperature. These tests were intended to ensure that the adsorption/desorption thermal cycles were not impacting the sorbent capacities for Xe. If the Xe calculated capacity remains relatively unchanged after undergoing multiple thermal cycles, the sorbent's thermal stability

and cycling assets are validated. The use of this particular feed gas at ambient temperature resulted in obtaining only Xe capacities since Kr capacities were not measureable. The ambient temperature condition was selected because the tests could be completed in a short time duration minimizing impacts to the desired testing schedule. Xe capacities of 31.6, 32.1 and 31.9 mmol/kg were measured, indicating that throughout the entire test regime of ambient and 191 K testing, the sorbent capacity for Xe remained unchanged, revealing the crystalline structure of the AgZ sorbent was not impacted by the 13 adsorption/desorption thermal test cycles.

The mass of the AgZ following removal from the cryostat after testing was measured at 5.368 g. This resulted in an insignificant 5 % weight loss assumed to be residual water vapor removed during desorption cycles. BET surface area analysis of the removed material was measured to be 244 m²/g, which is in good agreement with the surface area (250 m²/g) measured prior to testing. The unaffected surface area supports the fact that the material structure is thermally stable under numerous adsorption/desorption thermal cycles.

II.C.3 AgZ Adsorption Isotherm

A final evaluation of the AgZ sorbent was to generate a Xe adsorption isotherm at 220 K. The Xe concentrations ranged from 92 to 1040 ppm. The Langmuir equilibrium model was used for AgZ isotherm data. Predicted values for q_{\max} and K_{eq} at 220 K were 0.56 mol/kg and 76.6 m³/mol, respectively. Figure 4 includes the graphed Xe isotherm data at 220 K for the AgZ sorbent material.

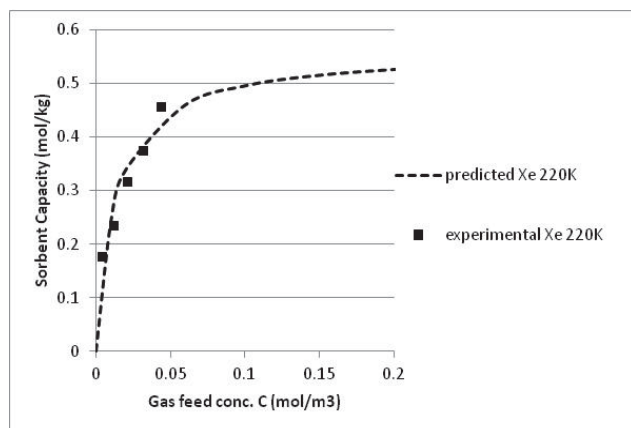


Fig. 4. Xe isotherm data at 220 K for AgZ sorbent.

It can be seen from the graph that the Langmuir model predicts the experimental adsorption data quite well. The Langmuir parameters can be utilized to develop models for the scale-up and process design calculations for implementation into a future UNF reprocessing off-gas treatment process.

III. CONCLUSIONS

A novel composite sorbent preparatory procedure has been developed at the INL. This procedure has been used to prepare two composite engineered form sorbents that have been tested with numerous test gases and evaluated for Kr and Xe capacities. Capacity results agree favorably with previously published results indicating the sorbents supplement a physical adsorption candidate technology decision for use in a UNF off-gas treatment facility. Further engineering evaluations are needed to study economical benefit comparisons between physisorption and cryogenic distillation technologies. In addition, sorbent cycling effects were also evaluated to observe thermal stability of the sorbents. Adsorption isotherms were generated for Kr and Xe using Langmuir regression with good agreement between experimental and predicted values.

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