METHOD FOR MAKING A URANIUM CHLORIDE SALT PRODUCT

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Abstract

The subject apparatus provides a means to produce UC\textsubscript{3} in large quantities without incurring corrosion of the containment vessel or associated apparatus. Gaseous Cl\textsubscript{2} is injected into a lower layer of Cd where CdCl\textsubscript{2} is formed. Due to its lower density, the CdCl\textsubscript{2} rises through the Cd layer into a layer of molten LiCl-KCl salt where a rotatable basket containing uranium ingots is suspended. The CdCl\textsubscript{2} reacts with the uranium to form UC\textsubscript{3} and Cd. Due to density differences, the Cd sinks down to the liquid Cd layer and is reused. The UC\textsubscript{3} combines with the molten salt. During production the temperature is maintained at about 600°C. While after the uranium has been depleted the salt temperature is lowered, the molten salt is pressure siphoned from the vessel, and the salt product LiCl-KCl-30 mol % UC\textsubscript{3} is solidified.
CHLORINATOR SCHEMATIC
FOR MAKING LiCl–KCl–30 m/o UCl₃

250 Kg UCl₃

Cl₂ FEED 1093 g mol
2.36 L/min (STP)
173 HRS

POUROUS BASKET AGITATOR LOAD
173 Kg URANIUM

QUARTZ TUBE

TEMP. –600ºC
HEAT GENERATION
RATE –1 KW
DURING CHLORINATION

CdCl₂ FORMATION RATE – 0.29 L/HR

FIG. 1
METHOD FOR MAKING A URANIUM CHLORIDE SALT PRODUCT

CONTRACTUAL ORIGIN OF THE INVENTION

The United States Government has rights in this invention pursuant to Contract No. W-31-109-ENG-38 between the U.S. Department of Energy and the University of Chicago.

FIELD OF THE INVENTION

This invention is a method for making uranium chloride, UC13-, by combining C12 with Cd to form CdCl2 which reacts with Uranium to form UC13.

BACKGROUND OF THE INVENTION

There is an ongoing problem concerning the development of a method of producing large quantities of uranium chloride (UC13) which is not corrosive to the containment vessel. The use of gaseous HCl as the chlorinating agent results in extensive levels of corrosion in ferrous metal containers and any associated ferrous components. To counter this, the use of non-corrosive reactants was explored. It is known that CdCl2 is not a corrosive chlorinating agent and can be combined with uranium to form UC13. However, if CdCl2 is purchased in its commercial form and then purified, the process becomes too expensive and also creates Cd metal waste. An alternate method involves the use of pyrophoric HCL with Uranium to form UC13; however, to make large batches of UC13, it is further into the liquid Cd while the opposing end is connected to a C12 feed. The second tube is used to transport a stream of gaseous C13 into the liquid Cd where it combines with the liquid Cd to form CdCl2. The solubility of Cd in CdCl2 at 600° C is 15 mol %. This allows the material at the C12 nozzle or orifice to have reactant Cd present; thus, the CdCl2 pool which forms around the nozzle should never be inert. Since the density, at 600° C, of CdCl2 is 3.37 while that of Cd is 7.82 and since the solubility of CdCl2 in Cd is near zero, the CdCl2 should feed into the overlying salt layer. The baffles serve to direct the flow of the CdCl2 towards the center of the containment vessel. A porous basket containing uranium, U, ingots is submerged in the liquid salt. The uranium reacts with the CdCl2 in the following manner:

3CdCl2 + 2U = 2UC13 + 3Cd

(LiCl—KCl)(LiCl—KCl—UC13)

Since both of the reactions, to produce CdCl2 and 2UC13 + 3Cd, are exothermic, the rate of C12 addition is controlled to produce heat at the 1 KW rate. The cooling rate of the vessel is designed to match this heat production rate in order to maintain the temperature of the interior of the vessel and its contents at approximately 600° C.

The porous basket containing uranium ingots is rotated in the salt causing mixing which is necessary to scrub off the UC13 and Cd, reaction products, from the uranium surface. The UC13 dissolves in the LiCl—KCl salt converting it to LiCl—KCl—UC13 as shown above. The cadmium metal is essentially insoluble in the salt and is heavier than the salt which results in the Cd sinking and recombining with the Cd layer under the molten salt layer. The C12 feed is halted prior to consumption of all of the uranium metal in the basket in order to guarantee a low level of CdCl2 in the product salt, LiCl—KCl—UC13. The residual concentration of Cd in the product salt should be <100 ppm. The equilibrium level of CdCl2 in the salt when the uranium metal is present is 5x10^-10 mol fraction.

After the reaction is complete, the temperature can be lowered to approximately 450°-500° C for pressure siphoning of the product salt from the vessel. As salt heet together with the Cd phase will be left in the vessel. The next batch of LiCl—KCl is then added.

BRIEF DESCRIPTION OF THE DRAWINGS

The present invention is illustrated in the accompanying drawing where:

FIG. 1 is a diagram of the chlorinator apparatus.

DETAILED DESCRIPTION OF THE INVENTION

FIG. 1 depicts a schematic of the apparatus for the chlorinator employed to formulate the UC13. As is shown in FIG. 1, the chlorination process is housed in a containment vessel. A layer of cadmium, Cd, is the base layer within the vessel. The upper layer is initially a CdCl2 free salt such as LiCl—KCl. The chlorinator is kept at an operational temperature of approximately 600° C which results in both layers being liquids. A hollow tube, preferably of a ferrous composition, penetrates the liquid salt and terminates within the liquid Cd while the opposing end remains above the surface of the liquid salt. In the preferred embodiment, the end of the tube forms a baffle. A corresponding baffle can be built into the container on the side opposing the tube. The baffles are arranged so that they slope upward to approximately the center of the vessel and terminate so as to create a fixed opening within the liquid Cd. A secondary hollow tube, preferably made of quartz, is threaded through tube. The lead end of the second tube extends past the end of the first tube such that it is further into the liquid Cd while the opposing end is connected to a C12 feed. The second tube is used to transport a stream of gaseous C13 into the liquid Cd where it combines with the liquid Cd to form CdCl2. The solubility of Cd in CdCl2 at 600° C is 15 mol %. This allows the material at the C12 nozzle or orifice to have reactant Cd present; thus, the CdCl2 pool which forms around the nozzle should never be inert. Since the density, at 600° C, of CdCl2 is 3.37 while that of Cd is 7.82 and since the solubility of CdCl2 in Cd is near zero, the CdCl2 should feed into the overlying salt layer. The baffles serve to direct the flow of the CdCl2 towards the center of the containment vessel. A porous basket containing uranium, U, ingots is submerged in the liquid salt. The uranium reacts with the CdCl2 in the following manner:

3CdCl2 + 2U = 2UC13 + 3Cd

(LiCl—KCl)(LiCl—KCl—UC13)

Since both of the reactions, to produce CdCl2 and 2UC13 + 3Cd, are exothermic, the rate of C12 addition is controlled to produce heat at the 1 KW rate. The cooling rate of the vessel is designed to match this heat production rate in order to maintain the temperature of the interior of the vessel and its contents at approximately 600° C.

The porous basket containing uranium ingots is rotated in the salt causing mixing which is necessary to scrub off the UC13 and Cd, reaction products, from the uranium surface. The UC13 dissolves in the LiCl—KCl salt converting it to LiCl—KCl—UC13 as shown above. The cadmium metal is essentially insoluble in the salt and is heavier than the salt which results in the Cd sinking and recombining with the Cd layer under the molten salt layer. The C12 feed is halted prior to consumption of all of the uranium metal in the basket in order to guarantee a low level of CdCl2 in the product salt, LiCl—KCl—UC13. The residual concentration of Cd in the product salt should be <100 ppm. The equilibrium level of CdCl2 in the salt when the uranium metal is present is 5x10^-10 mol fraction.

After the reaction is complete, the temperature can be lowered to approximately 450°-500° C for pressure siphoning of the product salt from the vessel. As salt heet together with the Cd phase will be left in the vessel. The next batch of LiCl—KCl is then added.
The foregoing description of a preferred embodiment of the invention has been presented for purposes of illustration and description. It is not intended to be exhaustive or to limit the invention to the precise form disclosed, and obviously many modifications and variations are possible in light of the above teaching. The embodiments described explain the principles of the invention and practical applications and should enable others skilled in the art to utilize the invention in various embodiments and with various modifications as are suited to the particular use contemplated. It is intended that the scope of the invention be defined by the claims appended hereto.

The embodiment of this invention in which an exclusive property or privilege is claimed is defined as follows:

1. A method for the formation of UCl$_3$, including:
   - heating a containment vessel to approximately 600°C;
   - establishing a base layer of cadmium in a base portion of the containment vessel where said cadmium layer is molten at 600°C;
   - establishing a second initial layer of salt above the layer of cadmium where said salt layer is molten at 600°C;
   - feeding chlorine gas through a hollow tube into the cadmium layer to form CdCl$_2$ which is less dense than the cadmium and thus, rises into the molten salt layer;
   - suspending a basket containing a plurality of uranium ingots in the molten salt layer in such a manner that the uranium ingots react with the CdCl$_2$ to form UCl$_3$ which is soluble in the molten salt;
   - providing sufficient reaction time to allow most of the uranium to react with the CdCl$_2$;
   - controlling a flow rate of Cl$_2$ so that heat generated by a pair of exothermic reactions, Cd+Cl$_2$→CdCl$_2$ and 3CdCl$_2$+2U→2UCl$_3$+3Cd which occur in the containment vessel is balanced by a heat removal rate from the containment vessel to maintain the temperature within the containment vessel at approximately 600°C;
   - removing the basket from the salt layer;
   - cooling the containment vessel to between 450°C and 500°C and pressure siphoning a product salt containing the UCl$_3$ from the containment vessel leaving a small heel of salt and the cadmium.

2. The method of claim 1 wherein the initial salt layer is LiCl—KCl.

3. The method of claim 1 wherein the basket is rotated during the reaction process to clean off the cadmium and the UCl$_3$ from the surface of the uranium.

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