

Uranium: Extending the Nuclear Fuel Cycle

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Abstract

This evaluation discusses the latest advances in the nuclear fuel industry's use of uranium, with a focus on recycling and health issues. It does this by stabilising the recombinant chemistry of uranium, which has increased interest in using compounds containing uranium in reactors as a class of materials that can produce safe and efficient nuclear fuel. Quantum Chemistry Experiments Employing Empty Law Force Constants Some uranium nitrides address the current topic regarding uranium triple bonding, i.e., the hypothesis that $N=U=O$, $N=U=NH$, $U=N$, and $N=U=N$ exhibit triple UN interactions, which can be verified and measured for the first time by local mode analysis. To investigate the chemical enrichment of uranium in full, we hope that this research will stimulate the uranium chemistry community and be a catalyst for fruitful theoretical and experimental partnerships.

Keywords: *Uranium, Recycling, Health, Chemistry, Reactors and Quantum*

Introduction

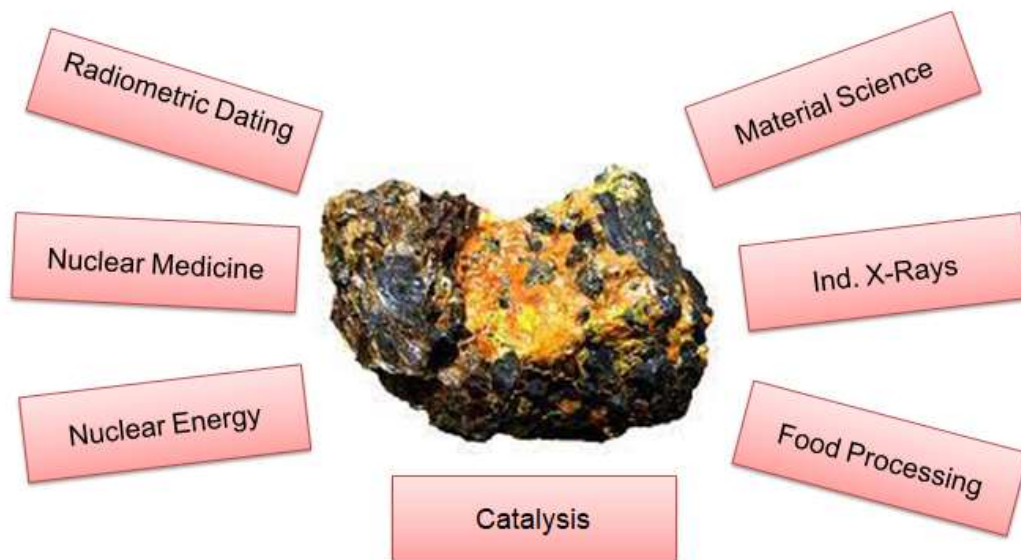
Uranium was discovered by the German scientist Martin Heinrich Klaproth in 1789. Uranium is the most common and recognised actinide element due to its use in nuclear fuel production. However, as Fig 1 [1] shows, the potential chemical applications of uranium are quite broad, including organometallic synthesis, catalysis, and other fields. Furthermore, uranium is one of the few naturally occurring actinides [2]. Other members—with the exception of thorium—are believed to have been formed by humans, although their signals are naturally due to the spontaneous fission of thorium and uranium. Three major isotopes of uranium are found in nature, with the non-fissionable ^{238}U being the most common and accounting for more than 99% of the recoverable uranium resources.

Uraninite or pitchblende, carnotite, and autunite are found in a wide variety of uranium minerals. Surface soil properties Uranium is separated from its ore using acid and solvent chemicals. Obtaining the required concentration of a given uranium isotope for energy production often requires the use of specialised isotope separation techniques. The most common isotope separation method is the gas extraction of UF_6 from uranium, followed by isotope separation brought on by mass differences. Organouranium complexes have uranium counterparts and are of great interest. Studies on the actinide cyclobutadienyl complexes and $(\eta^8\text{-C}_8\text{H}_8)_2\text{U}$ have been published, respectively [3].

The coordination chemistry of uranium has been extensively studied. Particular attention has been paid to potential applications in nuclear waste production. Reducing long-lived radioactive waste [4] requires efficient removal of uranium from spent nuclear fuel, or waste produced by the nuclear fuel cycle and recycled into the system. Although uranium oxides have long been used as nuclear fuel, they are difficult to handle in spent fuel reprocessing. Due to the stability,

flammability, and decomposition properties of water vapour, Uranium amides with N-substituents, such as uranyl nitrates utilising ligands based on picolinamide, complexes containing uranyl diamide and uranyl-Schiff base, are options for another suitable alternative to these amides. Another method of measuring chemical bond energies based on vibrational spectroscopy is used to bridge the gap in quantitative estimates of bond energies between the UO and the United Nations. Anxiety about future consequences still present today.

Fig 1. Instances of the wide variety of uses for uranium. [1]



Uranium from III to VI can adopt a variety of oxidation states, resulting in a wide range of covalency and 5f-/6d-orbital contributions to uranium-ligand (UL) bonding. Usually, U(III) is observed when donor-strong ligands exist and are ionic, such as lanthanide bonds. The topic of uranium polybonding has been explored, revealing new possibilities for small molecules. The relationship between uranium nitride and the United Nations' three standard networks is an interesting one. It has recently been suggested that uranium mononitride may be a desirable fuel for a variety of processes. Further research is needed to better understand the nature of the three UN networks. Computational chemistry is a powerful tool for mechanistic investigation, optimising uranium-based catalysts, studying electronic structure and uranium chemical bonding, and searching for new U-catalysed compounds.

Correlation effects must be carefully considered in theoretical descriptions of uranium complexes, as they play an important role in molecular properties, especially UL bonding. The complex Dirac equation, which yields accurate Dirac-wave functions as well as properties linked with the molecule, should be used rather than the neutral Schrödinger equation to capture these effects. Dirac's complete four-component (4c), including relativity in formalism, is the most rigorous approach to molecular structure calculations.

Effective special forces are discussed as a way of describing relativistic effects through nonrelativistic platforms; however, they are not feasible due to up to 20% impurities in uranium.

For even larger systems, the new NESC using the atomic approximation (NESCau) greatly improves relativistic calculations. The analysis is divided into areas such as the production of radioactive waste and uranium as nuclear fuel. Case studies are included in the analysis, addressing contextual issues as an alternative nuclear fuel. The article addresses the controversy surrounding U-N and U-O connection energies in the nitride of uranium and presents a new method for quantitatively assessing chemical bond energies by local vibration path analysis (LMA) results. The study concludes with a final analysis.

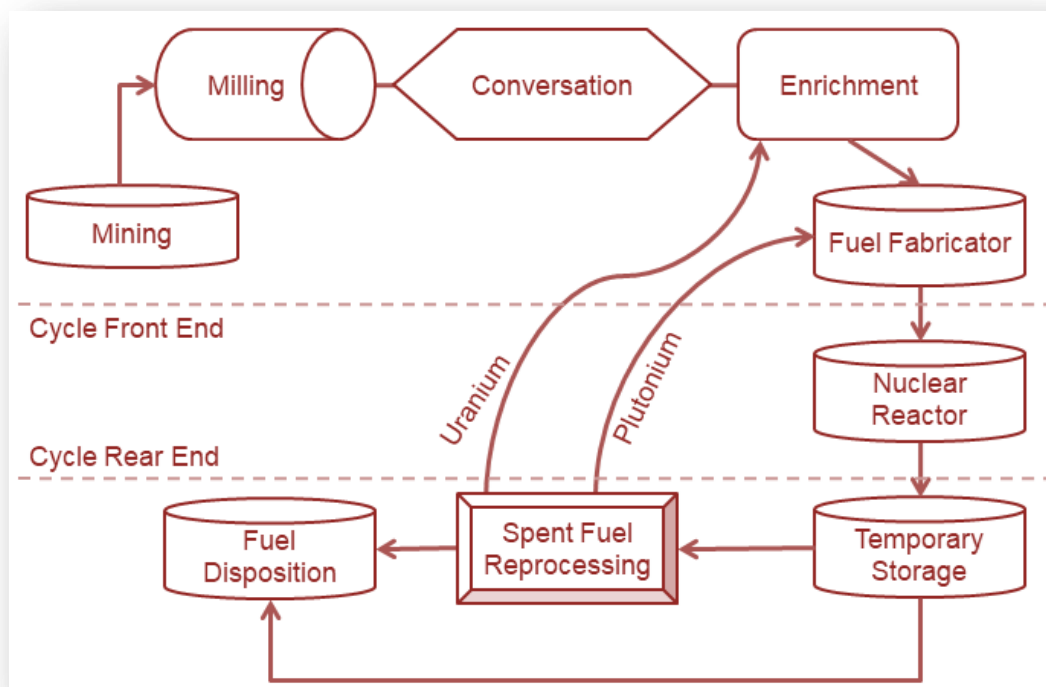
Objectives of the study

1. Verification of Uranium Bond Characteristics: Vibrational spectroscopy and nearby mode analysis have been used to verify the triple bonding nature of uranium compounds.
2. Quantitative Analysis of Bond Strength: The UN and UO bonds' covalent nature is evaluated by energy density measurements (H (rb)) and the local potential constant (BSO). strength assessment in quantitative terms.
3. Detailed understanding of uranium chemistry: study of oxidation states and bonding patterns of various uranium compounds to improve nuclear fuel efficiency and explore potential enrichment applications
4. To stimulate collaborative development in the field of uranium chemistry: encourage collaborative efforts involving theoretical and experimental methods to increase the depth of knowledge in the field of uranium chemistry, with the aim of enhancement and innovations in the discipline.

Uranium's Function in the Nuclear Fuel Cycle

The cycle of nuclear fuels in Fig 2 is an industrialized procedure that uses uranium to generate power through activities in a nuclear reactor plant. In other words, the cycle represents the flow of produced nuclear fuel up to the point where it is discarded. The most common form of uranium is UO₂. When uranium is depleted, fissile plutonium is produced. This explains why the next fuel cycle has a mixture of plutonium and uranium because the waste fuel can be reused and reprocessed [5,6]. Globally, uranium is a very common mine in many countries. To separate uranium from waste rock, the rock is first broken down in a mill and processed into tiny particles of dirt. This solution is then extracted with sulfur-containing acid. It is then processed into a solution to create a concentrate of uranium oxide, primarily U₃O₈, often called yellow-cake uranium because of its khaki colour. Usually, about 80% of the U₃O₈ concentrate is uranium. In contrast, starting stones could incorporate as little as 0.1% uranium. Yellow cakes generally have to be treated carefully because of impurities in the remaining minerals. This is an important process depending on the nature of the mineral. Other methods, such as supercritical carbon dioxide removal, have been proposed, especially for phosphate ores [7]. This is pure U₃O₈ offered for sale. A large nuclear power plant (1000 MW) needs about 200 tonnes to keep generating energy for a year.

Fig 2. Nuclear fuel cycle [8]



Uranium oxide is produced after uranium is smelted, but further processing is required before it can be used as nuclear reactor fuel. The fissionable fraction of naturally occurring uranium—that is, the fissionable nuclear reactor reaction that produces energy—is only 0.7%. The fissionable uranium (or isotope) is ^{235}U , and the rest is ^{238}U . It takes 3.5–5% of the fission isotope ^{235}U to keep the chain active. Consequently, to enrich (or "enhance") the ^{235}U element, a procedure known as enrichment in uranium must be used, which requires the presence of uranium in the gaseous state. Uranium dioxide, or UO_2 , is produced by producing U_3O_8 prior to use as fuel for reactors that do not require the enriched uranium they produce. UO_2 is converted to uranium hexafluoride, a very low-temperature gas, for reactors that require enriched uranium. First, UF_6 , UO_2 is transformed into solid UF_4 , which oxidises with F_2 to produce $\text{UF}_6(\text{g})$. Because this method requires less fluorine than direct conversion of the oxide to fluoride, it is generally preferred. Products from solid-state reactions should also be considered since they are frequent [9]. Such reactions have previously been studied theoretically for gas-phase processes [10].

U-enrichment procedures now employ centrifuges. However, there are additional methods, like radiation diffusion and laser-based materials, that were extensively used in the past and are now under investigation and then convert the uranium back to the oxide state. Several methods are available, such as an aqueous ammonia conversion system that liquefies UF_6 and then evaporates it. Dry conversion is a different process that is considered to be less harmful to the environment. The final product or reactor fuel, irrespective of the process, must have an appropriate oxygen-uranium ratio. Zirconium can be incorporated into nuclear fuel to improve strength and stability. It has also attracted interest for its likely uses in the reprocessing of used fuel [11].

The fission of uranium produces mostly radioactive. Therefore, nuclear waste protection issues exist. While reprocessing spent fuel is possible and is also used from nuclear waste because it still contains significant amounts of depleted uranium, fissile material generally must be stored with care because it is highly radioactive. Uranium oxide substitute: uranium nitride, is considered an alternative nuclear energy because of its elevated melting point [12]. $N\equiv U\equiv N$ was first synthesised by the synthesis of atomic uranium and molecular nitrogen in an argon matrix. It is clear that uranium in N_2 is capable of disrupting the triple bond. Further information on the surface and in its excited states comes from the recent investigation of two charged and uncharged states, $N\equiv U\equiv N$ [13], followed by the identification of uranium nitrides, with NUO including the corresponding, first-containing U in solid argon. The duplet spin state was anticipated based on calculations using density functional theory, which was found to be a consequence of the interaction between NO species. Zhou and Andrews also synthesized NUO in solid neon. The reaction of $NUNH$ with $N\equiv U\equiv N$ and hydrogen atoms [14] indicates the presence of two uranium-nitrogen triplet bonds.

Recycling of Used Fuel

Approximately, high-radioactive waste makes up 3%, plutonium makes up 1%, and uranium makes up 96% of spent fuel. Uranium and plutonium must be removed from nuclear waste in order to recycle spent fuel. In most cases, higher wastes are separated from waste fuels by treating them with strong nitric acid and then extracting them with tributyl phosphate (TBP) in kerosene. This gives fuel, which shrinks and turns into three dimensions. Other fission products are still present in water, and this solidifies uranium and plutonium [15]. The potential degradation of TBP may necessitate the elimination of possible products resulting from TBP degradation. Consequently, new solvents such as alamine 336 and aliquat 336 have been proposed and successfully used. Residual explosives can be removed by various methods. Ion exchange can be used to remove alkali-alkaline earth metals after pH changes. Due to their chemical similarities, lanthanides and actinides found in spent fuel are difficult to separate in the nuclear industry [16]. Different systems of coordination systems and ligands have been proposed for different approaches, which are complex. Chemical synthesis has often been used to effectively remove depleted uranium from wastewater for recycling. Moreover, their catalytic effect on uranium complexes has been demonstrated. Consequently, the synthesis of uranium chemistry will feature prominently in the analysis.

Uranium Coordination Chemistry

Interest in uranium bond chemistry has increased in recent years as stable foundational components for organometal catalysis and synthesis became available. The oxidation states of uranium atoms lead to a wide range of UL bonds of varying contributions from the 5f–6d orbitals and covalency. VI, or oxidation states, are present; U(IV) molecules are more difficult to synthesise under certain experimental conditions. U(V) compounds have been shown to enter the IV and VI oxidation states disproportionately; however, various stabilisation strategies have been proposed. Uranyl compounds are complexes of uranium containing a UO_2 group. According to high-level paired-group calculations, the end product of $UO + 2$ hydrolysis is $UO(OH) + 2$, where an intermediate valencer is formed in the process. Although the reaction barrier of the

corresponding U(VI) complex is marginally greater than the matching U(V) complex's, the synthesis is the subject of many studies.

For a time, uranium was thought to be the third least oxidizable element. However, $[\text{Cp}^0_3\text{U}]^-$, with Cp^0 respecting the $\text{C}_5\text{H}_3(\text{SiMe}_3)$ group, was the method used to synthesise the first U(II) complex in 2013 (see Fig. 3, complex 2a) followed by the more complex U(II) molecules, for example, by synthesising U(II) compounds and reacting with sodium as a reducing agent [17] tilt-structured U(III) and U(IV) metallocenes in addition to U(III). by potassium graphite The reduction resulted in the first stable U(II) $[\eta^5\text{-C}_5\text{Pr}_5)_2\text{U}]$ sandwich complex (see Fig. 3, complex 2b), which exhibits a unique linear Cp-U-Cp angle.

The primary cause of the complex's linear structure is the mixing of $6d_{z^2}$ and $7s$ orbitals, according to simulations of the electronic structure. Theoretical studies of $(\eta^5\text{-C}_5\text{Pr}_5)_2\text{U}$ -derived complexes are in progress, while NESCAU studies of sandwich complexes of actinides containing uranium have been completed. Numerous numerical studies have been carried out on negatively charged U(II) metallocenes, and because of the nature of electron withdrawal in the SiMe_3 group, it can act as a stabiliser for steady U(II) possessing an electron-donating ligand. Complexes have also been documented. The most stable state in these compounds is the high-spin quintet state. Uranyl nitrate, uranyl-diamide complexes, and base coordination complexes of uranyl-Schiff are N-substituted uranium amide complexes that have been described as promising possibilities for next-generation reactors. It is shown that a known uranyl cation is $[\text{UO}_2]^{2+}$. The isoelectric nitrogen counterpart of 2^+ $[\text{UN}_2]$ may play a role in nuclear reactor ceramic fuels in the future. Three U-N bonds and a central U (VI) $[\text{UN}_2]$ core are visible in the X-ray structures of triuranium complexes isolated in the argon matrix. According to photoelectric spectroscopy, electron entanglement and electronic excitation are predicted to bulk the $[\text{UN}_2]$ unit and stretch the U-N bond. It would be interesting to investigate the energies between a pentagonal bipyramidal $[\text{UN}_2\text{R}_2\text{R}'_3]$ core and two flanking uranium cations and ground They influence the strength of $\text{U}\equiv\text{N}$ triple bonds in excited states because excited-state uranium nitride can be abundant in nuclear reactors.

Human Health and Uranium

There have been accidents involving nuclear power plants, and safety precautions have caused serious problems. A catastrophic example is the accident at Chernobyl in Ukraine in 1986, when a fire exploded and released radioactive materials into the surroundings. Such events are characterised by dry lava, which disperses radioactive material and increases the likelihood of further explosions. High radiation levels in the surrounding areas increased the incidence of birth defects and cancer, especially leukemia. Japan's Fukushima Daiichi nuclear power plant suffered a similar disaster in 2011, causing power outages and toxic materials to leak into the sea. This catastrophe is similar to Chernobyl because it involves nuclear meltdown, gas, and the absorption of some contaminated salt water that is returned to the environment.

Many people have died and surrounding communities have been evacuated due to nuclear incidents in the Philippines. Discussion on nuclear power, in particular the long-term storage of radioactive waste, increased plant productivity while complying with safety regulations. The main health hazards posed by these accidents are depleted uranium (DU), resulting from the

enrichment and extraction of naturally occurring uranium. The main route of exposure to DU is aerosol inhalation, which can damage the kidneys, lungs, and other organs. But epidemiological studies have found no hard evidence of any adverse human effects from long-term exposure to DU-containing explosives. Major causes of uranium contamination pose health risks worldwide, and they include mining, phosphate coal, nuclear power plants, groundwater, and military operations. Carpet problems are failures and can be The World Health Organisation (WHO) established a limit of 30 mg/l air or food sources of uranium; however, data on other health problems such as cancer risk are not warranted. The radioactive isotope of uranium, which emits γ -rays, is highly carcinogenic to humans and animals, causing serious harm. Despite efforts to reduce the use of chemicals, the toxicity of these chemicals remains a concern.

Radionuclides are used in nuclear medicine for diagnosis and treatment. Uranium is used to produce radioisotopes such as $^{229}\text{Th}/^{225}\text{Ac}$ for targeted alpha therapy and $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ for imaging and research. Research reactors are the main source of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$; yields are lower than for cyclotrons and other methods. When the ^{233}U is depleted, ^{229}Th is created, yielding ^{225}Ac , which is used to treat prostate cancer. Radioactive contamination requires high levels of cleanup due to its potentially harmful nature. But cyclotrons aren't enough. Most $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ is produced in experimental reactors, while ^{229}Th is a decomposition product of ^{233}U , yielding ^{225}Ac .

Power of the UN Bond in Uranium Nitrides

This section presents an effective vibrational spectroscopy-based method for quantitatively assessing chemical bond strength or chemical bond weakness. This is used to check whether certain uranium nitrides, such as NUN, UN, $\text{NU}=\text{NH}$, and $\text{NU}=\text{O}$, display three UN connections or not. The idea of using normal-mode stretching force constants based on the well-known Badger law to describe chemical interactions originated in the 1920s and 1930s. However, its application to polyatomic compounds encountered significant challenges since specific vibrational modes tend to propagate in molecules as opposed to forming single bonds. M.A. This leads to a general application of Badger's rule based on local property energy constants and also serves as an alternative method for determining the energies of weak chemical bonds.

The local property constants (LMA) are sensitive to changes in electronic configuration and are not affected by the parameters used to characterise the molecule. They simply measure electrostatic properties based on atomic numbers. The seminal work of Zou and Kremer showed that the inherent energies of the binding or interaction of two atoms A and B, characterised by the coefficients q_n , are expressed in terms of local potential constants (AB); The Lishu Hale Extension represents a major advance in the quantitative interpretation of interactions in solids and glasses. The interaction energy scheme (BSO) is very useful for comparing large values of k .

$$BSO = u(ka)v$$

Additionally, the BSO value for a given zero force constant must be zero, and the u and v parameters are derived between two reference molecules whose k and BSO values are known; for example, bond orders of $n = 1$ for ethylene and $n = 1$ for ethane, respectively, are valid values for CC bonds. Meyer bonding orders [18] can be used as a guide in more intricate

bonding scenarios, like the bonding between metal and ligand. It should be remembered that the reference molecule and the group of molecules under study must be calculated using the same model chemistry.

Example molecules for single and double uranium-nitrogen bonds in this study are $\text{H}_2\text{N-UH}$ and HN=UH_2 , with k values of 2.253 and 4.865 mDyn/Å, respectively, in 2.166 and 1.872, respectively, which are in close agreement with previously published bond lengths. The corresponding Meyer bond orders for UN single and double bonds are 1.150 and 2.265, respectively. The bond order of the two UN bonds is 1.97, while the Meyer bond order of the one UN bond is 1.0. This makes the u and v coefficients used in this study 0.4894 and 0.8803, respectively.

Because of the single and double oxygen bonds in uranium, uranyl hydroxide ($\text{UO}_2(\text{OH})_2$) was used in the study as a model molecule for uranium oxygen bonds. UO bond lengths are 2.100 and 1.762 Å, while T_u UO single bond obtained k values of 2.791 and 7.114 mDyn/Å, respectively. The similar orders for Meyer bonds for individual and double UO bonds are 2.270 and 1.175, respectively. The corresponding bond order of the two UO bonds was 1.932, giving u and v values of 0.4855 and 0.7041, respectively. The Cremer-Kraka criterion for covalent bonding was used in conjunction with LMA to determine the covalent nature of UN and UO bonds using the Quantum Theory of Atoms in Molecules (QTAIM). Critically, it requires a bond distance, a bond critical point, and a sufficient condition if the energy density $H(\text{rb})$ at that point is less than zero is. The definition of $H(\mathbf{r})$ is:

$$H(\mathbf{r}) = G(\mathbf{r}) + V(\mathbf{r})$$

Tensile strength ($G(\mathbf{r})$) and potential energy ($V(\mathbf{r})$) were the main topics of analysis. It was found that a high $G(\mathbf{r})$ indicated a decrease in electron density, and a negative $V(\mathbf{r})$ with increasing electron density indicated stability. The control is denoted by the symbol $H(\text{rb})$; covalent bonds are denoted by $H(\text{rb}) < 0$ and $H(\text{rb}) > 0$. Relative effects were considered in calculations using the PBE0 density functional and the NESCau Hamiltonian in combination with the cc-pwCVTZ determined X2C basis for uranium and non-relativistic atoms for cc-pVTZ. Supporting previous hypotheses, the results (Table 1) verified the U-N interactions as triple interactions. The three bonds were identified using BSO values as $\text{U}\equiv\text{N}$ (2.976), $\text{N}\equiv\text{U}\equiv\text{N}$ (3.081), $\text{N}\equiv\text{U}=\text{NH}$ (3.087), and $\text{N}\equiv\text{U}=\text{O}$ (3.238). Relatively strong double bonds were found to be $\text{U}=\text{NH}$ in $\text{U}\equiv\text{N}$, $\text{N}\equiv\text{U}=\text{NH}$, $\text{N}\equiv\text{U}=\text{O}$, and $\text{U}=\text{O}$ in UO_2 . While the $1\Phi_u$ singlet state of excited UO_2 exhibited longer and weaker $\text{U}=\text{O}$ interactions, the triplet $3\Phi_u$ ground state of UO_2 corresponded to the $\text{U}=\text{O}$ of $\text{N}\equiv\text{U}=\text{O}$. The relationships between BSO values and $H(\text{rb})$ and k are shown in Figs 4 and 5.

Table 1. The spin multiplicity and oxidation number of uranium for $N\equiv U\equiv N$, $U\equiv N$, $N\equiv U=NH$, $N\equiv U=O$, and UO_2 (a) were computed at the NESCaU/PBE0//cc-pwCVTZ-X2C (uranium) and NESCaU/PBE0//cc-pVTZ (N,O,H) levels of theory. Bond lengths in Å, k and in mDyn/Å, BSO and H(rb), in Hartree/Å³. [19]

$U\equiv N$ $U=L$; $L=N,O$ U State	r	k_a	BSO	H(rb)	r	k_a	BSO	H(rb)
$N\equiv U\equiv N$	1.713	8.085	3.081	-0.437	-	-	-	-
$N\equiv U$	1.732	7.772	2.976	-0.398	-	-	-	-
$N\equiv U=NH$	1.695	8.077	3.078	-0.474	1.828	4.918	1.989	-0.222
$N\equiv U=O$	1.697	8.554	3.238	-0.470	1.761	7.824	2.066	-0.319
UO_2	-	-	-	-	1.836	5.883	1.689	-0.239
UO_2	-	-	-	-	1.775	7.288	1.966	-0.310

Note that for UO_2 , the $1\Phi_u$ state is shown in the first column and the $3\Phi_u$ state in the second. In the table, dashes (-) represent missing data.

All investigated UN and UO bonds have a covalent nature, as shown in Table 1 and Fig. 1, supporting the general assumption that strong bonds are highly covalent. Energy density is measured only at a point along the bond path, while local potential constants are reflected by the electronic environment encountered no relationship. [19]

Fig 4. BSO values of UN bonds for the chosen uranium nitrides were obtained using the previously mentioned power relationship to get from k a values. Levels of theory NESCaU/PBE0//cc-pwCVTZ-X2C (uranium) and NESCaU/PBE0//cc-pVTZ (N,O,H) [20]

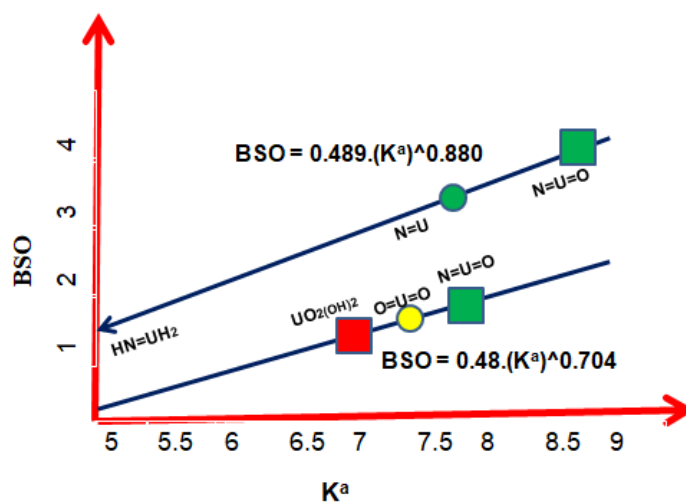
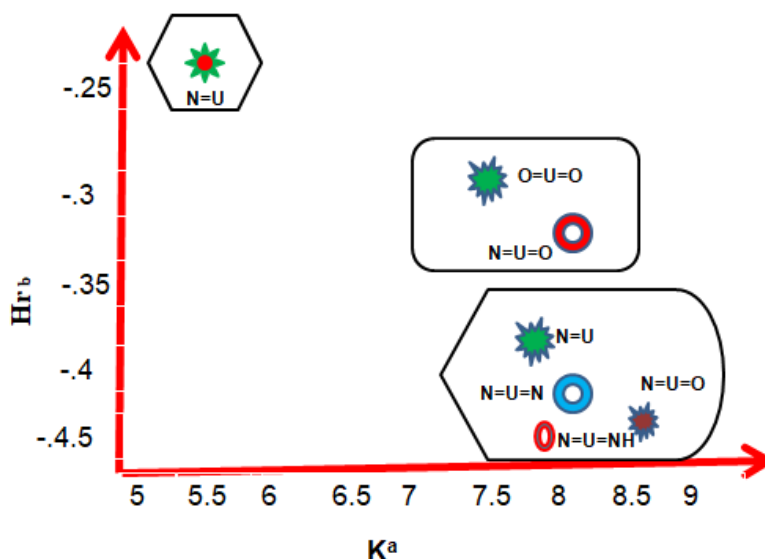


Fig 5. $H(r_b)$ and k_a correlation for UN and UO bonds. Levels of theory NESCau/PBE0//cc-pwCVTZ-X2C (uranium) and NESCau/PBE0//cc-pVTZ (N,O,H) [20]



Conclusions

This comprehensive review brings together the most current developments in the nuclear fuel production process using uranium, including chemical health concerns and recycling. It highlights the various reactivities of uranium and raises interest in its potential applications in enrichment. Emerging U(II) complexes with uranium nitrides are examples of unique characteristics of potential advances towards safe and effective nuclear fuel. Uranium triple bonds UN used vibrational spectroscopy in quantum chemical analysis to determine triple bond characteristics, especially in some nitrides. (II) Focus on evaluating the chemical properties and covalent properties of uranyl compounds. This effort is aimed at improving nuclear waste reduction from reactor fuel, and developing uranium-based raw materials has been good. The research seeks to stimulate collaboration between theory and experiment in order to fully realise the promise of uranium chemistry and foster innovation and improvement in the field.

References:

1. Tsoureas, N.; Mansikkamäki, A.; Layfield, R.A. Uranium(IV) cyclobutadienyl sandwich compounds: Synthesis, structure and chemical bonding. *Chem. Commun.* 2020, 56, 944–947. [CrossRef] [PubMed]
2. Xin, X.; Douair, I.; Zhao, Y.; Wang, S.; Maron, L.; Zhu, C. Dinitrogen Cleavage by a Heterometallic Cluster Featuring Multiple Uranium-Rhodium Bonds. *J. Am. Chem. Soc.* 2020, 142, 15004–15011. [CrossRef] [PubMed]
3. Boronski, J.T.; Liddle, S.T. The Emergence of Actinide Cyclobutadienyl Chemistry. *Eur. J. Inorg. Chem.* 2020, 2020, 2851–2861. [CrossRef]

4. Geist, A.; Adnet, J.M.; Bourg, S.; Ekberg, C.; Galan, H.; Guilbaud, P.; Miguirditchian, M.; Modolo, G.; Rhodes, C.; Taylor, R. An overview of solvent extraction processes developed in Europe for advanced nuclear fuel recycling, part 1—Heterogeneous recycling. *Sep. Sci. Technol.* 2021, 56, 1866–1881. [CrossRef]
5. Loveland, W.D.; Morrissey, D.J.; Seaborg, G.T. *Modern Nuclear Chemistry*; John Wiley & Sons: New York, NY, USA, 2017.
6. Crossland, I. *Nuclear Fuel Cycle Science and Engineering*; Elsevier: Amsterdam, The Netherlands, 2012.
7. Prabhat, P.; Rao, A.; Mishra, V.G.; Shah, D.J.; Kumar, P.; Tomar, B.S. Direct extraction of uranium from yellow cake and ore matrices using supercritical carbon dioxide. *Radiochim. Acta* 2020, 108, 769–777. [CrossRef]
8. International Atomic Energy Agency. (2006). **The nuclear fuel cycle**. IAEA.
9. Cotton, S. Binary Compounds of the Actinides. In *Lanthanide and Actinide Chemistry*; John Wiley & Sons, Ltd.: West Sussex, UK, 2006; Chapter 10, pp. 155–172.
10. Peluzo, B.M.; Galvão, B.R. Theoretical study on the structure and reactions of uranium fluorides. *J. Mol. Model.* 2018, 24, 197. [CrossRef] [PubMed]
11. Ali, K.; Ghosh, P.; Arya, A. A DFT study of structural, elastic and lattice dynamical properties of FeZr and FeZr₂ intermetallics. *J. Alloys Compd.* 2017, 723, 611–619. [CrossRef]
12. Streit, M.; Ingold, F. Nitrides as a nuclear fuel option. *J. Eur. Ceram. Soc.* 2005, 25, 2687–2692. [CrossRef]
13. Liu, G.; Zhang, C.; Ciborowski, S.M.; Asthana, A.; Cheng, L.; Bowen, K.H. Mapping the electronic structure of the uranium (VI) dinitride molecule, UN₂. *J. Phys. Chem. A* 2020, 124, 6486–6492. [CrossRef] [PubMed]
14. Wang, X.; Andrews, L.; Vlaisavljevich, B.; Gagliardi, L. Combined Triple and Double Bonds to Uranium: The N≡U=N-H Uranimine Nitride Molecule Prepared in Solid Argon. *Inorg. Chem.* 2011, 50, 3826–3831. [CrossRef]
15. Malin, S.A.; Alexis-Martin, B. Assessing the state of uranium research: Environmental justice, health, and extraction. *Extr. Ind. Soc.* 2020, 7, 512–516. [CrossRef]
16. Bhattacharyya, A.; Mohapatra, P.K. Separation of trivalent actinides and lanthanides using various ‘N’, ‘S’ and mixed ‘N, O’ donor ligands: A review. *Radiochim. Acta* 2019, 107, 931–949. [CrossRef]
17. Windorff, C.J.; MacDonald, M.R.; Meihaus, K.R.; Ziller, J.W.; Long, J.R.; Evans, W.J. Expanding the Chemistry of Molecular U(2+) Complexes: Synthesis, Characterization, and Reactivity of the {[C₅H₃(SiMe₃)₂]₃U}(-) Anion. *Chem. Eur. J.* 2016, 22, 772–782. [CrossRef] [PubMed]
18. Mayer, I. Bond order and valence indices: A personal account. *J. Comput. Chem.* 2007, 28, 204–221. [CrossRef]

19. Mako's, M.Z.; Zou, W.; Freindorf, M.; Kraka, E. Metal-Ring Interactions in Actinide Sandwich Compounds: A Combined Normalized Elimination of the Small Component and Local Vibrational Mode Study. *Mol. Phys.* 2020, 118, e1768314. [CrossRef
20. Wang, Y., Zhou, W., Wu, J., & Li, S. (2021). Theoretical investigation of the structure and properties of uranium nitrides UN_x (x = 1-7). *The Journal of Physical Chemistry C*, 125(47), 26230-26239.