Electronic Supplementary Material (ESI) for RSC Advances.

$U@C_{36}$. Is There room enough for a second uranium?

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Supporting Information.

Content:

Table S1. Energy decomposition analysis of the uranium-fullerene interaction. Values in kcal/mol.

	$U@C_{28}$		$U@C_{36}$		$U_2@C_{36}$		$U_2@C_{36}^{2+}$	
Δ_{Orb}	-1634.4	60.9%	-1242.7	60.0%	-2043.7	54.8%	-1711.3	64.9%
Δ_{Elst}	-1049.0	39.1%	-829.8	40.0%	-1685.9	45.2%	-927.5	35.1%
Δ_{Pauli}	2163.1		1741.1		3480.8		2335.4	
Δ_{Int}	-520.3		-331.4		-248.7		-303.3	

Table S2. Thermodynamic parameters for $U@C_{36}$, $U_2@C_{36}$ and $U_2@C_{36}^{2+}$, related to the encapsulation of the uranium atoms. Values in kcal/mol.

	ΔG	ΔH
$U@C_{36}$	-1804.1	-1826.5
$U_2@C_{36}$	-1850.9	-1708.9
$U_2@C_{36}^{2+}$	-2028.0	-1901.3

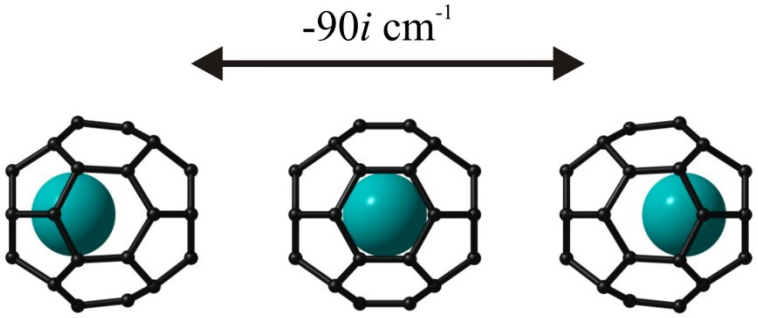

Figure S1. Representation for the single negative frequency of the Hessian found for D_{6h}-[$U@C_{36}$].

NCI Analysis for steric repulsion regions.

In order to account for steric interactions between the uranium and the cage, further analysis of the electron density ($\rho(r)$) is informative for locate repulsive regions. In this sense the non-covalent index based on the reduced density gradient ($s(r)$) at low-density regions has been introduced. The character of the non-bonding interaction can be addressed by the product between the second eigenvalue sign for the electron density Hessian (λ_2) and $\rho(r)$, denoting: stabilizing ($\lambda_2 < 0$), weak ($\lambda_2 \approx 0$) or repulsive interactions ($\lambda_2 > 0$) [S1,S2]. From such analysis, the main steric repulsion is located the six-membered rings from the middle of the prolate cage, which is larger in the bimetallic counterpart. The noncovalent interaction (NCI) analysis was carried out by using the NCIPLOT program developed by Weitao Yang and co-workers [S1.S2] and the NCI Milano program developed by Saleh and co-workers [S3,S4], both based on the analysis of electron density descriptors.

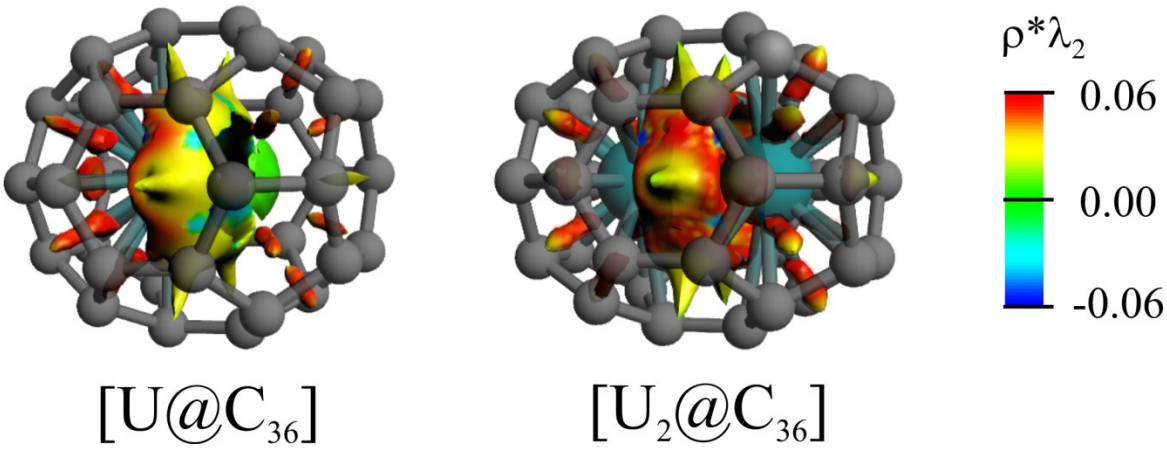

Figure S2. NCI analysis for $U@C_{36}$ and $U_2@C_{36}$.

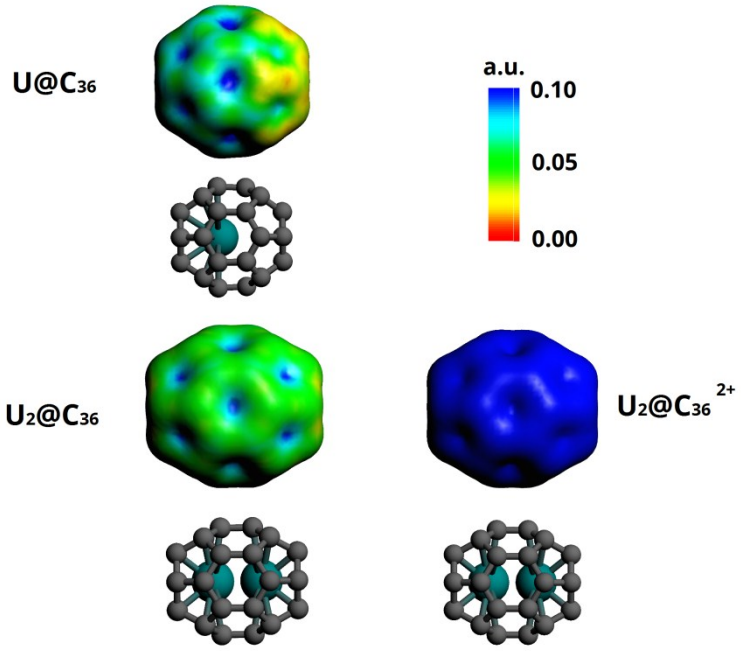

Figure S3. Electrostatic potential surface for $U@C_{36}$, $U_2@C_{36}$ and $U_2@C_{36}^{2+}$.

References

[S1] Johnson, E. R.; Keinan, S.; Mori-Sanchez, P.; Contreras-García, J.; Cohen, A. J.; Yang, W. Revealing Noncovalent Interactions J. Am. Chem. Soc. 2010, 132, 6498– 6506.

[S2] Contreras-García, J.; Johnson, E. R.; Keinan, S.; Chaudret, R.; Piquemal, J.; Beratan, D. N.; Yang, W.NCIPLOT: A Program for Plotting Noncovalent Interaction Regions J. Chem. Theory Comput. 2011, 7, 625– 632.

[S3] Saleh, G.; Gatti, C.; Lo Presti, L. Non-covalent Interaction via the Reduced Density Gradient: Independent Atom Model vs Experimental Multipolar Electron Densities Comput. Theor. Chem. 2012, 998, 148– 163.

[S4] Saleh, G.; Gatti, C.; Lo Presti, L.; Contreras-García, J.Revealing Non-covalent Interactions in Molecular Crystals through Their Experimental Electron Densities Chem.–Eur. J. 2012, 18, 15523– 15536.