QUANTUM-CHEMICAL CALCULATIONS OF THE ENTHALPY OF FORMATION OF ISOMERIC 5/6/5 TRICYCLIC TETRAZOLOTETRAZINE DERIVATIVES ANNELATED WITH NITROAZOLES

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Development of high-energy components of rocket fuels in FRC PCP MC RAS

- FRC PCP MC RAS in collaboration with other RAS institutes for more than 60 years has been developing high-energy substances.
- In recent years more attention is paid to quantum calculations of physical and chemical properties (Gaussian).
- Computer design of substances not yet synthesized.

High-energy density materials (HEDM)

military affairs

mineral exploration

rocket and space technology

construction

metallurgy

mining

Enthalpy of formation

$$\Delta_f H^0_{298}$$

Quantum-chemical ab initio calculations

Stationary Schrödinger Equation

$$\hat{H}(\vec{r},\vec{R})\psi(\vec{r},\vec{R}) = E\psi(\vec{r},\vec{R})$$

Hamiltonian of a polyatomic molecule

$$\hat{H} = \sum_i \hat{T}_i + \sum_\alpha \hat{T}_\alpha + \sum_{i>k} V_{ik} + \sum_{i,\alpha} V_{i\alpha} + \sum_{\alpha>\beta} V_{\alpha\beta} + V_{so}$$

Electronic and nuclear kinetic energies

Electronic and nuclear Coulomb interaction energies

Spin-orbital interaction

Approximations of ab initio calculations

- Adiabatic approximation - the kinetic energy of nuclei is neglected
- Self-consistent field - the electron moves in the effective field of the remaining electrons
- Rectification of the electron correlation

Quantum-chemical calculation methods

GAUSSIAN 09

- DFT:
  - M062X with the 6-311G+(2d,p) basis
  - ωB97XD with the aug-cc-pVTZ basis
- CBS methods: CBS-4M, CBS-QB3, CBS-APNO
- G4 and G4(MP2)

Calculating the enthalpy of formation

The atomization reaction

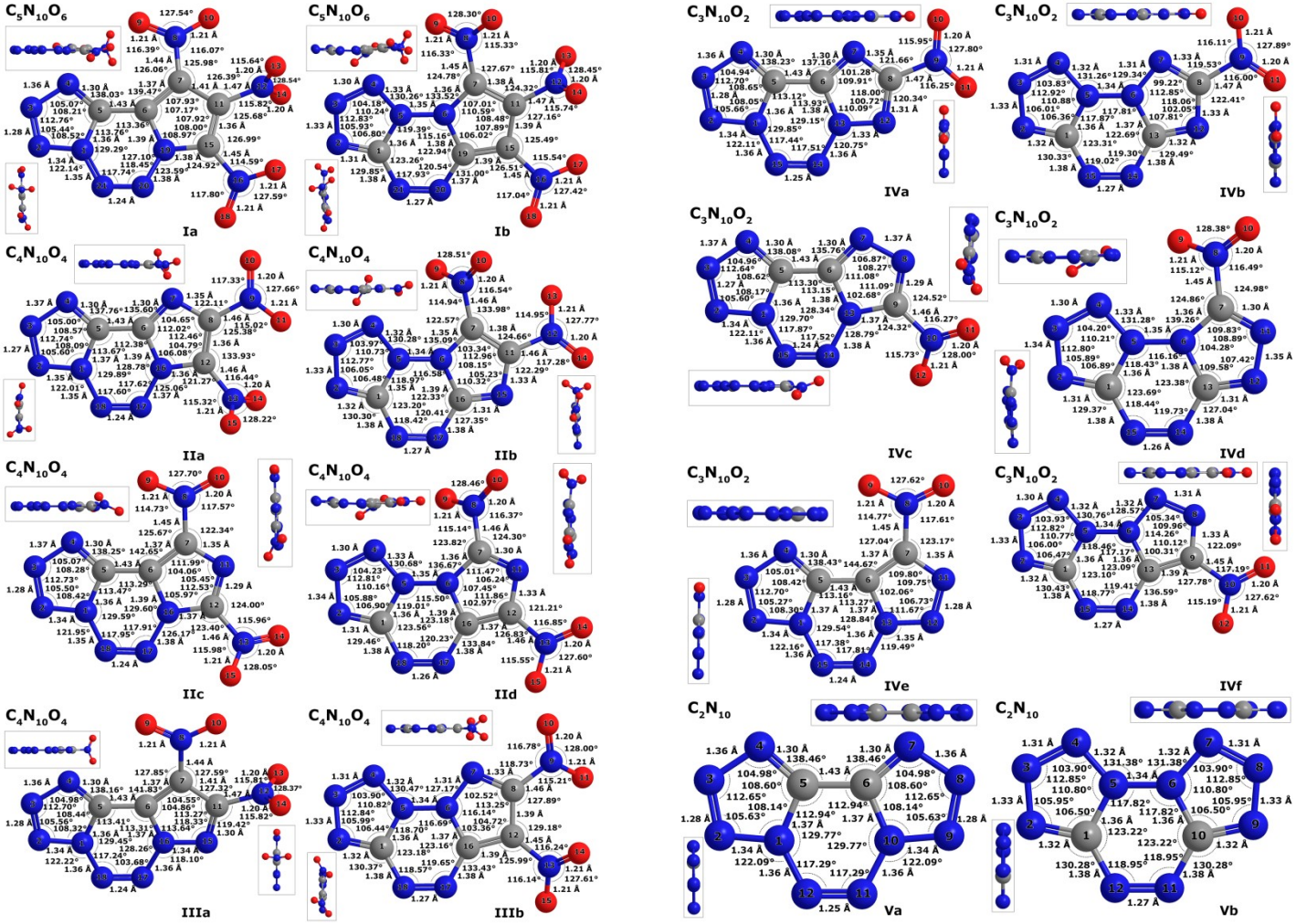
$$\Delta \quad (\quad): \quad (\quad) + \quad (\quad) + \quad (\quad) = \quad (\quad)$$

The formation reaction from simple substances

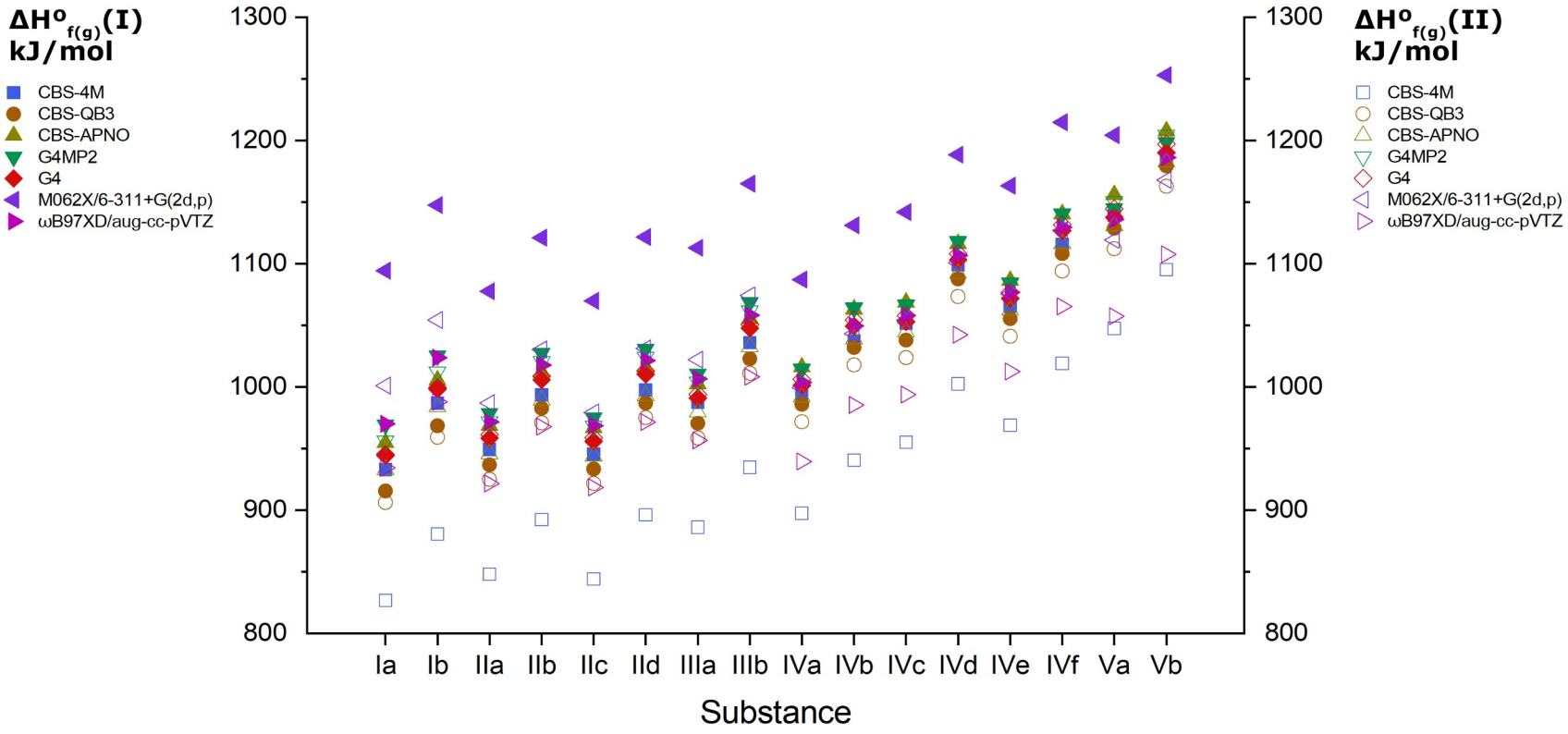
$$\Delta \quad (\quad): \quad (\quad h \quad) + \frac{}{2} \quad {}_2(\quad) + \frac{}{2} \quad {}_2(\quad) = \quad (\quad)$$

Optimized geometry

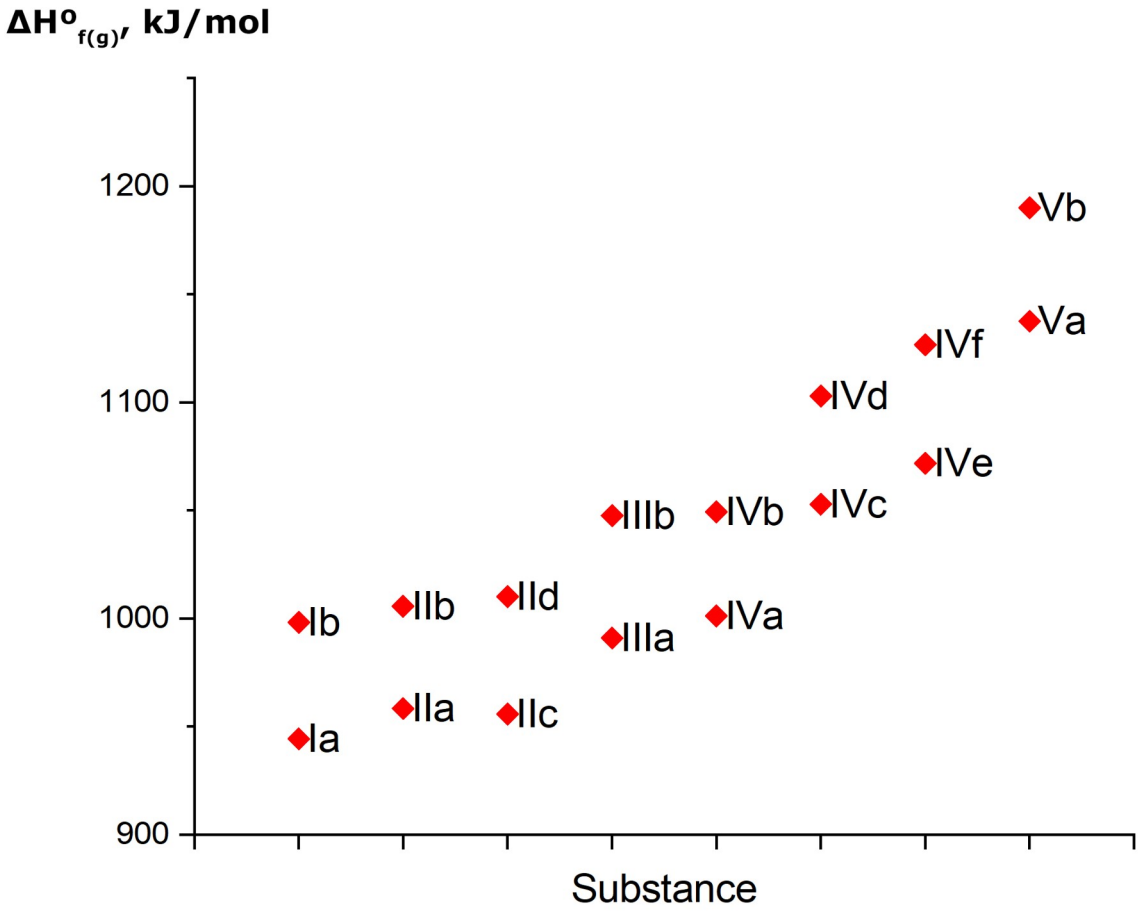
Calculation level: **M062X/6-311+G(2d,p)**



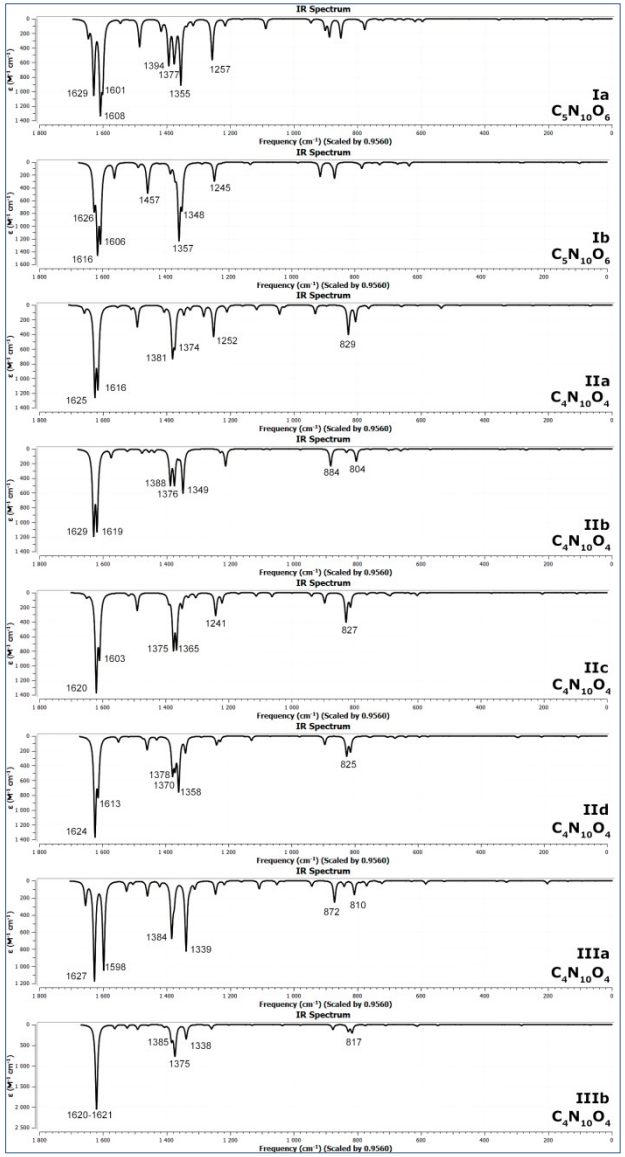
Enthalpy of formation

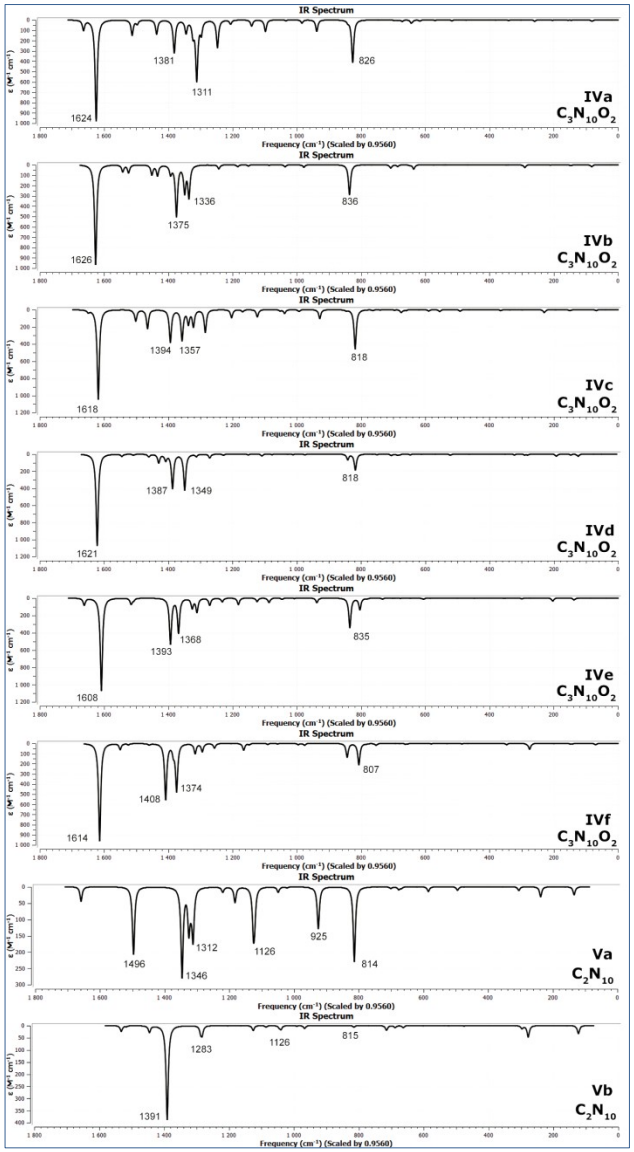


Enthalpy of formation (G4)



The IR absorption spectra





ωB97XD/ aug-cc-pVTZ 0.956

Computational details

Volta2 (Lomonosov-2)
Intel Xeon Gold 6240 processors (18 cores, 2.60GHz - 1497.6 GFlop/s) with Nvidia Tesla V100 graphics accelerators (900-2G500-0010-000, 1246 MHz, 7 TFlop/s)
Time limit 48 hours

FRC PCP MC RAS
(Intel(R) Xeon(R) Gold 6140 CPU @ 2.30 GHz, RAM 259 Gb, 20Tb disk space)
8 cores per task
20 minutes (CBS-4M for substances IIa-d and IIIa,b)
3.5 months (CBS-APNO for substances Ia,b).

Conclusions

- Geometric and thermochemical parameters of a number of 5/6/5 of tricyclic compounds based on 1,2,3,4- and 1,2,4,5-tetrazoletetrazines, annelated with nitroazoles, have been obtained by ab initio quantum-chemical methods (Gaussian 09).
- For a known substance, the results of our calculations showed a slight deviation (2-8% depending on a method) from the literature data, and for new, not yet synthesized tricycles, the data were obtained for the first time.
- The dependence of the enthalpy of formation on the structure of the compounds has been established.
- The methods were compared in terms of accuracy and time costs. The least demanding method is CBS-4M, the most demanding one is CBS-APNO.

The work was performed using the equipment of the Center for Collective Use of Super High-Performance Computing Resources of the Lomonosov Moscow State University.

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THANK YOU FOR THE ATTENTION!