Polymorphism of organic, especially chiral, compounds is of great interest and value for modern crystallography, physical chemistry, process, and patent law. Since the free energy of different polymorphs of the same substance is different, all their physicochemical properties are different, that is, melting point, solubility, stability, hygroscopicity, pharmacodynamic and pharmacokinetic properties, and so on. Therefore, bioactive substances are particularly attractive from the point of view of guided polymorph search. In this abstract, we reported crystallization and heterogeneous equilibria and transitions of four racemic N-substituted 4-arylsulfanyl-3-chloro-5-hydroxy-3-pyrroline-2-ones consisting an unsaturated γ-lactam ring that is an important pharmacophore group. As a result, in this series of compounds, a unique "conglomerate-conglomerate" polymorphic pair and a pair of packing polymorphs were found. All individual crystalline phases were studied by means of single crystal and powder X-ray diffractions. Their phase behavior was carefully investigated using heat flux differential scanning calorimetry, temperature-resolved solid-state vibration spectroscopy and hot-stage microscopy.

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