

## THERMODYNAMIC PROPERTIES OF WATER IN THE CRITICAL REGION

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### ABSTRACT

The supercritical-water-cooled reactor (SCWR) is one of the nuclear reactor technologies selected for research and development under the Generation IV program. SCWRs offer the potential for high thermal efficiencies and considerable plant simplifications for improved economics. One of the main characteristics of critical water is the strong variations of its thermal-physical properties in the vicinity of the critical point. These large variations may result in an unusual heat transfer behavior. The 1967 IFC Formulation for Industrial Use, which until 1998 formed the basis of steam tables used in many areas of steam power industry throughout the world since the late 1960's, has been now replaced with the IAPWS IF-97 Formulation for the Thermodynamic Properties of Water and Steam for Industrial Use, adopted by the International Association for the Properties of Water and Steam (IAPWS) in 1997. An IAPWS release points out that this new formulation has some unsatisfactory features in the immediate vicinity of the critical point. In order to investigate this singular aspect, which is crucial to better understand the heat transfer mechanism in a SCWR system, predictions by the IAPWS-IF97 formulation will be compared with thermodynamic properties values predicted by an alternative crossover equation of state as well as with experimental data found in literature.

### 1. INTRODUCTION

The supercritical-water-cooled reactor (SCWR) is one of the six nuclear reactor technologies selected to Generation IV nuclear energy systems [1]. SCWRs are basically similar to light-water-cooled reactors (LWRs) that operate above the thermodynamic critical point of water (22.064 MPa and 647.064 K) with a direct once-through cycle. Operation above the critical pressure eliminates coolant boiling, since the supercritical primary coolant remains single-phase throughout the system. Due to the absence of two-phase flow in the reactor, no boiling crisis (i.e., departure from nucleate boiling or dry out) exists, thereby avoiding discontinuous heat transfer regimes within the reactor core. Recirculation pumps, steam generators, steam separators and dryers are eliminated. Besides being simpler plants with fewer major components, SCWRs offer increases in the thermal efficiency in comparison to current-generation LWRs. The efficiency of a SCWR can approach 44% compared to 35% for LWRs.

The heat transfer mechanisms at supercritical pressures are mainly characterized by the thermal-physical properties which may vary strongly, specially near the pseudo-critical curve. Thus, correct predictions of thermodynamic and transport properties of water at and near the pseudo-critical temperatures play a crucial role in the thermal-hydraulic design of a SCWR

system. Therefore, existing theoretical formulations for predicting the thermal-physical properties of water in the critical region must be evaluated for both precision and accuracy as well as for their ability to reproduce available experimental data. In this sense, the present work is intended to investigate to what extent the IAPWS Formulation 1997 [2,3] and the crossover equation of state proposed by Wyczalkowska and collaborators [4] can describe the thermodynamic properties of water in the critical region.

Official international formulations for water properties are developed and maintained by the International Association for the Properties of Water and Steam. IAPWS is an international non-profit association of national organizations concerned with the properties of water and steam, particularly thermophysical properties and other aspects of high-temperature steam, water and aqueous mixtures that are relevant to thermal power cycles and other industrial applications. IAPWS is organized as an association of member countries (or groups of countries). Current members are Argentina and Brazil, Britain and Ireland, Canada, the Czech Republic, Denmark, France, Germany, Greece, Italy, Japan, Russia, and the United States.

Since 1967 the 1967 IFC Formulation for Industrial Use (IFC-67) has been formally recognized to calculate thermodynamic properties of water and steam for any official use such as performance guarantee calculations of thermal power cycles. The IFC-67 formulation was developed by the International Formulation Committee (IFC) formed in 1963 by the Sixth International Conference on the Properties of Steam held in New York. The equations underlying IFC-67 are published for example by Meyer [5] and Schmidt [6]. IFC-67 has formed the basis of steam table [5, 6] and thermodynamic properties computer programs used in many fields of industry throughout the world.

In September 1997 the IAPWS adopted a new formulation for the thermodynamic properties of water and steam for industrial use. This new formulation, called IAPWS Industrial Formulation 1997 for the Thermodynamic Properties of Water and Steam [2], abbreviated to IAPWS-IF97, replaces the previous industrial formulation IFC-67. The IAPWS-IF97 formulation covers a wider range of pressure and temperature than the IFC-67 formulation and fits the current accepted best data within closer tolerances.

It should be emphasized that the IFC-67 formulation is still current in spite of the issuing of the IAPWS-IF97 formulation. The long period of IFC-67 use has resulted in all or parts of it being embedded in a wide range of software used by manufacturers of boilers, turbines, heat exchangers, and pumps; it has been used by utilities and private industrial power plant operators. Consequently, the change-over from IFC-67 to IAPWS-IF97 will not necessarily be either simple or inexpensive.

The IAPWS-IF97 formulation should not be confused with the IAPWS-95 formulation [4] adopted by IAPWS in 1995 for general and scientific use. The IAPWS-95 formulation provides a state-of-the-art, accurate representation of the thermodynamic properties of water and steam that is recommended for research use and general use outside the power industry, where computational speed is not an issue.

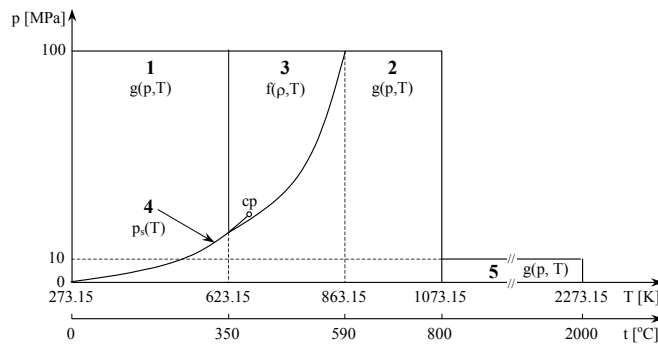
Crossover formulations for the description of the thermodynamic properties of fluids in the vicinity of the critical point have been developed by Chen and co-workers [9,10], van Pelt and Sengers [11], Kiselev and Friend [12], Wyczalkowska and Senger [13], Wyczalkowska and co-workers [4].

## 2. THEORETICAL FORMULATIONS

This section is devoted to describe briefly the IAPWS Formulation 1997 for the properties of water and steam as well as the crossover equation of state for steam in the critical region.

### 2.1. The IAPWS Formulation 1997

The IAPWS Industrial Formulation 1997 is applicable to the whole of the thermodynamic subdomain shown in Fig. 1 that extends in temperature from 273.15 K to 2273.15 K, and that extends in pressure from the ideal-gas zero pressure to an upper pressure limit. This upper pressure limit is 100 MPa for temperatures between 273.15 K and 1073.15 K, and 10 MPa for temperatures between 1073.15 K and 2273.15 K.



**Figure 1. Regions and Equations of IAPWS-IF97.**

The whole subdomain is divided into five regions, numbered 1 to 5. Regions 1, 2, 3, and 5 refers to the single-phase states of compressed water and superheated steam; region 4 corresponds to the conjugate saturation states, that is, the liquid-vapor saturation curve. The boundary between regions 2 and 3 is defined by a quadratic pressure-temperature relation. The values of pressure, temperature and density at the critical point (cp) are 647.096 K, 22.064 MPa, and  $322 \text{ kg m}^{-3}$ , respectively.

Regions 1, 2 and 5 are described by fundamental equations for the specific Gibbs free energy  $g(T,p)$ , and region 3 by a fundamental equation for the specific Helmholtz free energy  $f(\rho,T)$ . By using appropriate thermodynamic relations, all the thermodynamic properties may be derived directly by partial differentiation of these fundamental equations. Region 4 is described by an implicit bi-quadratic equation which can be solved directly for either saturation pressure  $p_s$  or saturation temperature  $T_s$ .

Further details about this formulation, including basic equations and their uncertainties, consistency along region boundaries, and computing-time results may be found in the article published by the working group responsible for the development of the IAPWS Industrial Formulation 1997.

## 2.2. The Crossover Equation of State

In crossover formulations [4, 8-13], the thermodynamic properties of fluids in the critical region are described in terms of scaling laws with universal critical exponents and universal scaling functions. As illustration, scaling law for isothermal compressibility  $\kappa$  along a critical isochore, pressure  $p$  along a critical isotherm, density  $\rho$  along the phase coexistence curve, and specific isochoric heat capacity  $c_v$  along a critical isochore are as follows [14]:

$$\kappa^* = \Gamma^\pm |\Delta T^*|^{-\gamma}, \quad |\Delta p^*| = D |\Lambda \rho^*|^\delta, \quad |\Delta \rho^*| = B |\Delta T^*|^\beta, \quad c_v^* = A^\pm |\Delta T^*|^{-\alpha}$$

where the asterisk (\*) denotes a property value made dimensionless by using appropriate combinations of the critical pressure  $p_c$ , temperature  $T_c$ , and density  $\rho_c$ , the symbol  $\Delta$  denotes the departure of a property from its critical value, and  $\alpha$ ,  $\beta$ ,  $\gamma$ , and  $\delta$  are the critical exponents. The constants  $\Gamma^\pm$ ,  $D$ ,  $B$ , and  $A^\pm$  are called critical amplitudes.

In the light of the crossover model proposed by Wyczalkowska and co-workers [4], which is considered in the present work, the specific Helmholtz free energy  $f$  may be expressed as

$$f = \frac{A}{\rho V} = \frac{p_c T_c}{\rho T_c} \tilde{A}, \quad (1)$$

where  $A$  is the Helmholtz free energy; and

$$\tilde{A} = \frac{T_c}{p_c} \frac{A}{VT} \quad (2)$$

defines the dimensionless Helmholtz free energy density. The Helmholtz free energy  $A$  per unit volume  $V$  is assumed to be a function of density  $\rho$  and temperature  $T$ .

The various thermodynamic quantities are made dimensionless with the aid of the critical point parameters  $\rho_c$ ,  $T_c$ , and  $p_c$  such that

$$\tilde{\rho} = \frac{\rho}{\rho_c}, \quad \tilde{T} = -\frac{T_c}{T}, \quad \text{and} \quad \tilde{p} = \frac{p T_c}{p_c T}. \quad (3)$$

In addition, the reduced density difference and the reduced temperature difference are defined by the relations

$$\Delta \tilde{\rho} = \tilde{\rho} - 1 \quad \text{and} \quad \Delta \tilde{T} = \tilde{T} + 1. \quad (4)$$

The Helmholtz free energy density is decomposed as

$$\tilde{A} = \Delta \tilde{A} + \tilde{\rho} \tilde{\mu}_0(\Delta \tilde{T}) + \tilde{A}_0(\Delta \tilde{T}), \quad (5)$$

where  $\Delta\tilde{A}$  is the part that takes into account the critical fluctuations;  $\tilde{\mu}_0(\Delta\tilde{T})$  and  $\tilde{A}_0(\Delta\tilde{T})$  are analytic background functions approximated by

$$\tilde{A}_0(\Delta\tilde{T}) = \sum_{k=0}^4 \tilde{A}_k (\Delta\tilde{T})^k \quad \text{and} \quad \tilde{\mu}_0(\Delta\tilde{T}) = \sum_{k=0}^3 \tilde{\mu}_k (\Delta\tilde{T})^k \quad (6)$$

with  $\tilde{A}_0 = -1$ . The remaining coefficients  $\tilde{A}_j$  and  $\tilde{\mu}_j$  are system-dependent and have to be determined from experimental data. Coefficients  $\tilde{\mu}_0$  and  $\tilde{\mu}_1$  are associated with the zero points of specific entropy and enthalpy. These two coefficients were here adjusted to give the entropy and enthalpy values calculated from the IAPWS-IF97 formulation at  $T = 500$  K and  $\rho = 500$  kg/m<sup>3</sup>. The system-dependent coefficients for water are presented in Table 1.

By introducing the following transformation of variables,

$$t = c_1 \Delta\tilde{T} + c \left( \frac{\partial \Delta\tilde{A}_x}{\partial M} \right)_t \quad \text{and} \quad M = c_p (\Delta\tilde{\rho} - d_1 \Delta\tilde{T}) + c \left( \frac{\partial \Delta\tilde{A}_x}{\partial t} \right)_M, \quad (7)$$

the critical part of the Helmholtz free energy density may be expressed as

$$\Delta\tilde{A} = \Delta\tilde{A}_x - c \left( \frac{\partial \Delta\tilde{A}_x}{\partial M} \right)_t \left( \frac{\partial \Delta\tilde{A}_x}{\partial t} \right)_M \quad (8)$$

On the base of a Landau six-term expansion [15], function  $\Delta\tilde{A}_x$  is approximated by [10]

$$\begin{aligned} \Delta\tilde{A}_x = & \frac{1}{2} t M^2 T D + \frac{u^* \bar{u} \Lambda}{4!} M^4 D^2 U + \frac{1}{5!} a_{05} M^5 D^{5/2} V U + \frac{1}{6!} a_{06} M^6 D^3 U^{3/2} \\ & + \frac{1}{4!} a_{14} t M^4 T D^2 U^{1/2} + \frac{1}{2!2!} a_{22} t^2 M^2 T^2 D U^{-1/2} - \frac{1}{2} t^2 K \end{aligned} \quad (9)$$

where  $T$ ,  $D$ ,  $U$ , and  $V$  are rescaling functions defined in terms of a crossover function  $Y$  as

$$T = Y^{(2\nu-1)/\Delta_S}, \quad D = Y^{-\eta/\Delta_S}, \quad U = Y^{\nu/\Delta_S}, \quad V = Y^{(\Delta_A-1/2)/\Delta_S}, \quad K = \frac{v}{\alpha \bar{u} \Lambda} (Y^{-\alpha/\Delta_S} - 1). \quad (10)$$

In these equations  $\nu$ ,  $\eta$ ,  $\alpha = 2 - 3\nu$ ,  $\Delta_S$  and  $\Delta_A$  are universal critical exponents. The values of the parameters occurring in Equations (9) and (10) are listed in Table 1.

The crossover function  $Y$  is to be determined iteratively from the following set of coupled algebraic equations

$$1 - (1 - \bar{u})Y = \bar{u} \left[ 1 + \left( \frac{\Lambda}{\kappa} \right)^2 \right]^{1/2} \quad (11)$$

and

$$\begin{aligned} \kappa^2 = & tT + \frac{u^* \bar{u} \Lambda}{2} M^2 DU + \frac{1}{3!} a_{05} M^3 D^{3/2} VU + \frac{1}{4!} a_{06} M^4 D^2 U^{3/2} \\ & + \frac{1}{2} a_{14} t M^2 TDU^{1/2} + \frac{1}{2} a_{22} t^2 T^2 U^{-1/2}. \end{aligned} \quad (12)$$

**Table 1. Parameters in the six-term crossover formulation for water [4].**

Parameter	Value	Parameter	Value
$T_c$	647,096 K	$a_{05}$	0.241 81
$p_c$	22.064 MPa	$a_{06}$	0.815 17
$\rho_c$	322 kg/m <sup>3</sup>	$a_{14}$	0.295 59
$\alpha$	0.110	$a_{22}$	0.737 32
$\eta$	0.033 333	$\tilde{A}_0$	-1.000 00
$v$	0.630	$\tilde{A}_1$	-6.810 75
$\Delta_S$	0.51	$\tilde{A}_2$	6.380 44
$\Delta_A$	1.32	$\tilde{A}_3$	8.267 78
$u^*$	0.472	$\tilde{A}_4$	-9.652 75
$\bar{u}$	0.351 85	$\tilde{\mu}_0$	-11.199 69*
$\Lambda$	2.373 59	$\tilde{\mu}_1$	-22.612 97*
$c_1$	2.513 03	$\tilde{\mu}_2$	-20.809 59
$c_p$	2.093 85	$\tilde{\mu}_3$	-14.473 83
$c$	-0.142 71		
$d_1$	-0.694 66		

\*Adjusted to reproduce entropy and enthalpy from IAPWS IF-97 Formulation.

All the thermodynamic properties can be derived directly by partial differentiation of the fundamental equations for the Helmholtz free energy. Various properties and first derivatives of thermodynamic interest are listed in Table 2 in terms of the functions  $f(\rho, T)$  and  $\tilde{A}(\tilde{\rho}, \tilde{T})$ .

The crossover equation of state here discussed is applicable to a temperature range

$$310 \text{ K} < T < 390 \text{ K} \quad \text{at} \quad \rho = \rho_c$$

and a density range

$$336 \text{ kg/m}^3 < \rho < 1170 \text{ kg/m}^3 \quad \text{at} \quad T = 310 \text{ K}.$$

This range of validity comprise a region bounded by  $\chi^{-1} = (p_c T / \rho_c^2 T_c) \chi \leq 2.0$ .

**Table 2. Thermodynamic properties expressed in terms of f and  $\tilde{A}$ .**

Thermodynamic Property	Helmholtz Relation	Crossover Model Relation
Pressure, p	$\rho^2 f_\rho$	$(p_c T / T_c) (\tilde{\rho} \tilde{A}_{\tilde{\rho}} - \tilde{A})$
Specific entropy, s	$-f_T$	$(p_c / \rho T_c) (\tilde{T} \tilde{A}_{\tilde{T}} - \tilde{A})$
Specific internal energy, $u = f + Ts$	$f - Tf_T$	$(p_c T / \rho T_c) (\tilde{T} \tilde{A}_{\tilde{T}})$
Specific enthalpy, $h = u + p / \rho$	$f + \rho f_\rho - Tf_T$	$(p_c T / \rho T_c) (\tilde{\rho} \tilde{A}_{\tilde{\rho}} + \tilde{T} \tilde{A}_{\tilde{T}} - \tilde{A})$
Specific chemical potential, $\mu = f + p / \rho$	$f + \rho f_\rho$	$(p_c T / \rho T_c) (\tilde{\rho} \tilde{A}_{\tilde{\rho}})$
Volume expansivity, $\beta = -(\partial \rho / \partial T)_p / \rho$	$\frac{f_{\rho T}}{\rho^3 f_{\rho\rho} + 2\rho^2 f_\rho}$	$\frac{1}{T} \left( \frac{-\tilde{\rho} \tilde{T} \tilde{A}_{\tilde{\rho}\tilde{T}} + \tilde{\rho} \tilde{A}_{\tilde{\rho}} + \tilde{T} \tilde{A}_{\tilde{T}} - \tilde{A}}{\tilde{\rho}^2 \tilde{A}_{\tilde{\rho}\tilde{\rho}}} \right)$
Isothermal compressibility, $\kappa = (\partial \rho / \partial p)_T / \rho$	$\frac{1}{\rho^3 f_{\rho\rho} + 2\rho^2 f_\rho}$	$-\frac{1}{p_c} \left( \frac{\tilde{T}}{\tilde{\rho}^2 \tilde{A}_{\tilde{\rho}\tilde{\rho}}} \right)$
Spec. isochoric heat capacity, $c_v = (\partial u / \partial T)_\rho$	$-Tf_{TT}$	$-(p_c / \rho T_c) (\tilde{T}^2 \tilde{A}_{\tilde{T}\tilde{T}})$
Spec. isobaric heat capacity, $c_p = (\partial h / \partial T)_p$	$c_v + \frac{\beta^2 T}{\rho \kappa}$	$\frac{p_c}{\rho T_c} \left[ \frac{(-\tilde{\rho} \tilde{T} \tilde{A}_{\tilde{\rho}\tilde{T}} + \tilde{\rho} \tilde{A}_{\tilde{\rho}} + \tilde{T} \tilde{A}_{\tilde{T}} - \tilde{A})^2}{\tilde{\rho}^2 \tilde{A}_{\tilde{\rho}\tilde{\rho}}} - T^2 \tilde{A}_{\tilde{T}\tilde{T}} \right]$
Isothermal susceptibility, $\chi = (\partial \rho / \partial \mu)_T$	$[2f_\rho + \rho f_{\rho\rho}]^{-1}$	$[(p_c T / \rho_c^2 T_c) \tilde{A}_{\tilde{\rho}\tilde{\rho}}]^{-1}$
Speed of sound (w), $w^2 = (\partial p / \partial \rho)_s$	$c_p \rho (2f_\rho + \rho f_{\rho\rho}) / c_v$	$(p_c T / \rho_c T_c) (\tilde{\rho} c_p \tilde{A}_{\tilde{\rho}\tilde{\rho}} / c_v)$

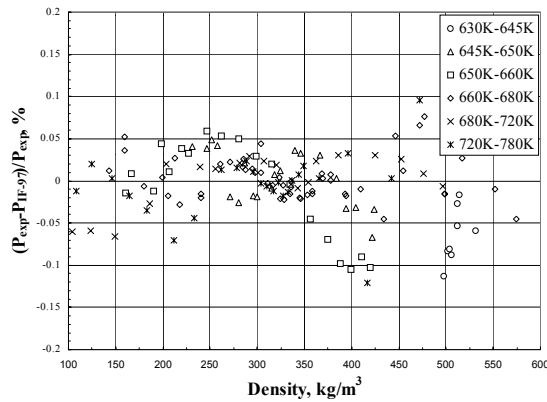
  

$f_\rho = \left( \frac{\partial f}{\partial \rho} \right)_T$	$f_T = \left( \frac{\partial f}{\partial T} \right)_\rho$	$f_{\rho\rho} = \left( \frac{\partial^2 f}{\partial \rho^2} \right)_T$	$f_{TT} = \left( \frac{\partial^2 f}{\partial T^2} \right)_\rho$	$f_{\rho T} = \left( \frac{\partial^2 f}{\partial \rho \partial T} \right)$
$\tilde{A}_{\tilde{\rho}} = \left( \frac{\partial \tilde{A}}{\partial \tilde{\rho}} \right)_{\tilde{T}}$	$\tilde{A}_{\tilde{T}} = \left( \frac{\partial \tilde{A}}{\partial \tilde{T}} \right)_{\tilde{\rho}}$	$\tilde{A}_{\tilde{\rho}\tilde{\rho}} = \left( \frac{\partial^2 \tilde{A}}{\partial \tilde{\rho}^2} \right)_{\tilde{T}}$	$\tilde{A}_{\tilde{T}\tilde{T}} = \left( \frac{\partial^2 \tilde{A}}{\partial \tilde{T}^2} \right)_{\tilde{\rho}}$	$\tilde{A}_{\tilde{\rho}\tilde{T}} = \left( \frac{\partial^2 \tilde{A}}{\partial \tilde{\rho} \partial \tilde{T}} \right)$

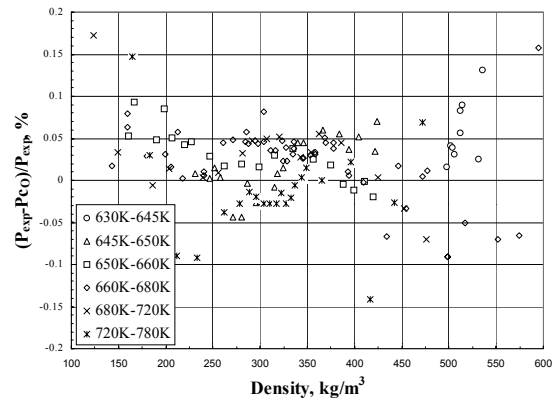
### 3. DISCUSSION OF RESULTS

In this section, the accuracy of the IAPWS-IF97 formulation will be discussed based on comparisons with selected experimental data and with values predicted by the crossover equation of state developed by Wyczalkowska et al. [4].

Comparisons between experimental critical  $p\rho T$  data reported by Rivkin and Akhundov [16,17], Rivkin and Troyanovskaia [18], and Rivkin et al. [19] and values predicted by the IAPWS IF-97 formulation and by the crossover equation of state are shown in Figs. 2 and 3, respectively. All the experimental data plotted in these figures are inside the range of validity of the crossover equation of state. The IAPWS IF-97 formulation describes the experimental data with a root mean square deviation (RMSD) of about 0.07%. The RMSD associated to the crossover equation of state was found to be about 0.18%. Due to the scattering of data points, no systematic tendency is obvious from these plots. One can point out that both the formulations reproduces the  $p\rho T$  data within the experimental uncertainties.

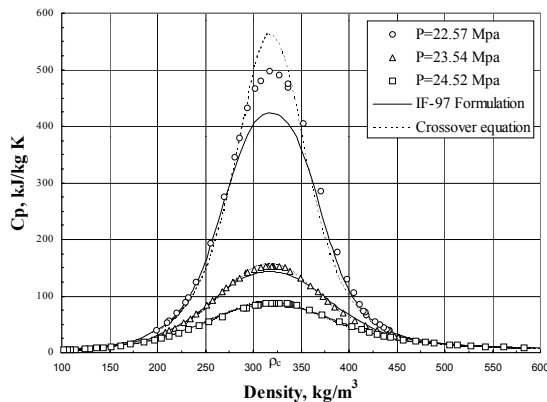


**Figure 2. Percentage deviations of experimental pressures for water [16-19] from values calculated with the IAPWS-IF97 formulation.**

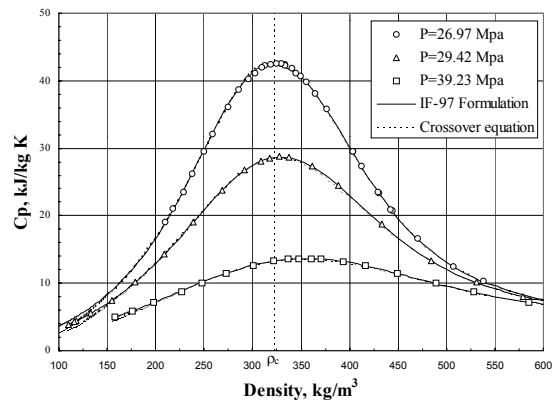


**Figure 3. Percentage deviations of experimental pressures for water [16-19] from values calculated with the crossover equation of state.**

The specific isobaric heat capacity for water along several representative isobars as a function of density is shown in Figs. 4 and 5. The solid curves indicate values calculated with the IAPWS-IF97 formulation, and the dashed curves correspond to values calculated with the crossover equation of state. Experimental data represented in the plots are those reported by Sirota and Mal'tsev [20] and by Sirota, Beljakova and Shrago [21]. Examination of these figures reveals a small offset in peak location between the experimental data and the predicted curves, as noticed earlier by Sengers et al. [14]. In the immediate vicinity of the critical point, the IAPWS-IF97 formulation tends to underestimate the specific isobaric heat capacity. Excluding the data points on the isobar  $p = 22.57$  Mpa, the IAPWS IF-97 formulation represents the remaining  $c_p$  data with a standard deviation of 2.2%, and the crossover equation with a standard deviation of 2.0%.

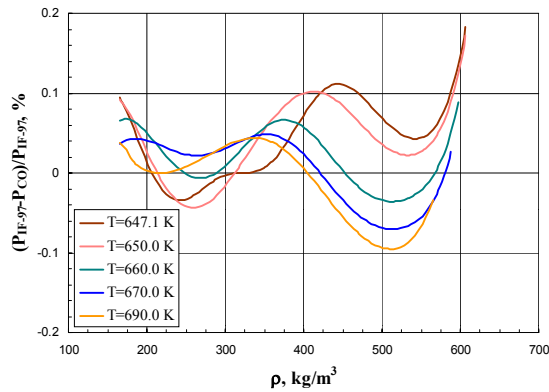


**Figure 4. Specific isobaric heat capacity of water in the vicinity of the critical point as a function of density along isobars.**

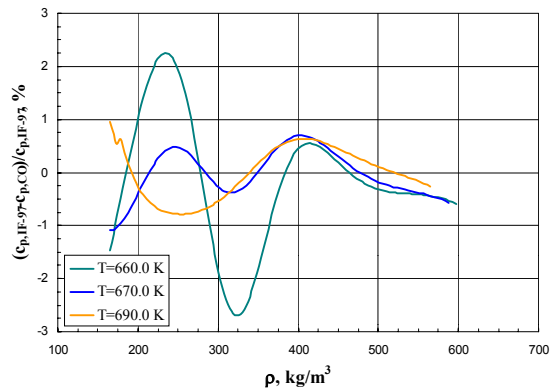


**Figure 5. Specific isobaric heat capacity of water in the extended critical region as a function of density along isobars.**





**Figure 6. Percentage differences between the values of pressure calculated with the IAPWS IF-97 formulation and with the crossover equation of state.**



**Figure 7. Percentage differences between the values of specific isobaric heat capacity calculated with the IAPWS IF-97 formulation and with the crossover equation of state.**

Comparisons between the IAPWS IF-97 formulation and the crossover equation of state along several isotherms as a function of density are shown in Figs. 6 and 7. The percentage differences between the values of pressure are represented in Fig. 6, and the percentage differences between the values of specific isobaric heat capacity are shown in Fig. 7. One can infer from Fig.7 that the specific isobaric heat capacity predicted by the IAPWS IF-97 formulation exhibits some unphysical behavior in the region very close to the critical point.

#### 4. CONCLUSIONS

Comparisons with experimental data and with values predicted by a crossover equation of state indicate that the IAPWS Industrial Formulation 1997 represents satisfactorily the thermodynamic properties of water in a large range of temperatures and densities in the critical region. However, this formulation tends to give an unphysical behavior for some of the derivative properties (here represented by the specific isobaric heat capacity) in the immediate vicinity of the critical point. Therefore, some caution is recommended when using this formulation to calculate derivative properties (e.g. isothermal compressibility, volume expansivity, isochoric and isobaric specific heat capacity, speed of sound, etc.) in such a region.

#### ACKNOWLEDGMENTS

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