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A SIMPLIFIED HARDNESS MODEL FOR WC–Co–CUBIC CEMENTED CARBIDES

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Abstract

Hardness is an essential mechanical indication of cemented carbides. The current work presents an approach to predict the hardness of three-phase WC–Co–Cubic cemented carbides, which establishes a relationship among composition, structure and mechanical performance. With the input of initial composition and grain sizes of carbides, structural parameters needed to predict the hardness can be calculated by thermodynamic calculations and diffusion simulations. The calculated hardness of a series of WC–Co–Cubic cemented carbides agrees reasonably with the experimental data. The present model is of reference to predict the hardness for multi-phase composites and design the new type of WC–Co-based cemented carbides.

Keywords: *Hardness model; WC–Co–Cubic cemented carbides; CALPHAD*

1. Introduction

Cemented carbides, which consist of hard transition metal carbides embedded in soft metal binders, are widely used for cutting, milling, and so forth [1]. With the increasing demand of the industry, more hard carbides, such as TiC, NbC, are introduced into cemented carbides to improve their comprehensive performance [2]. The hardness of cemented carbides is very important to the evaluation of the mechanical performance. A reliable model to predict the hardness of cemented carbides would make the development highly efficient.

Models developed to calculate the hardness of a single phase have been widely accepted [3, 4]. The present work focuses on one predictive model for multi-phase WC–Co–Cubic cemented carbides. There are several models reported in the literature, which can calculate the hardness of WC–Co two-phase cemented carbides [5-10]. The main differences among these models lie in different treatments for the deformation of the carbide phase and/or binder phase, and the selection of structural parameters which are believed to affect the hardness. The advantages and disadvantages of these models have been analyzed by Shatov et al. [11]. Among these models, Lee and Gurland's model (LG model) [5] relies on the Hall–Petch relation well-established for the hardness of polycrystals and has the smallest number of assumptions, and thus is most widely accepted [11]. The LG model is given as follows:

$$H_{CC} = H_{\mathrm{WC}} V_{\mathrm{WC}} C + H_{\mathrm{Co}} (1 - \mathrm{V_{WC}} \mathrm{C}) \tag{1}$$

$$H_{\mathrm{WC}} = 13.5 + \frac{7.2}{\sqrt{d_{\mathrm{WC}}}} (\mathrm{GPa}) \tag{2}$$

$$H_{\mathrm{Co}} = 2.98 + \frac{3.9}{\sqrt{\lambda}} (\mathrm{GPa}) \tag{3}$$

$$\lambda = d_{WC} \frac{V_{Co}}{(1 - V_{Co})(1 - C)} \tag{4}$$

where C is the WC/WC contiguity, d_{WC} the mean grain size of WC phase, V_{WC} the volume fraction of the WC phase, λ the mean free path of the binder phase, and H_{WC}, H_{Co} and H_{CC} are the hardness of WC, Co binder phase and the alloy, respectively. Both H_{WC} and H_{Co} follow the Hall-Petch relationship.

Xu and Ågren modified the LG model based on

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the observation that the binder and carbide phases have different ratios of hardness to the yield stress [7]. They added factors to the WC and Co terms in the LG model. Shatov et al. [10] provided an enhanced model based on the LG model which deals with the cemented carbides have WC tailored to the shape of flatter triangular prism, and explains the consequent increase of hardness. The LG model has also been applied to calculate the hardness of Ti(C, N)–Co/Ni alloys [12]. Lee et al. [13] extended the LG model to calculate the hardness of three-phase WC–Co–TiC cemented carbides. However, their model [14] is questionable according to the analysis by Shatov et al [11]. Their model simply treats WC–Co as one phase and TiC as another one. In addition, their model does not consider the contiguity of carbides, although the model is developed based on the LG model.

The aim of this work is to present a simplified approach to calculate the hardness of WC–Co–Cubic three-phase cemented carbides based on the LG model, since the cubic phases such as TiC, TaC, NbC, and Ti(C, N) are often introduced to cemented carbides. With the initial composition, the volume fractions of phases can be calculated from thermodynamic calculations and diffusion simulations. From the volume fractions of phases and grain sizes of carbides, other parameters needed to predict the hardness can be calculated using derived equations. As a result, the newly established hardness model for three-phase cemented carbides needs fewer parameters measured from experiment. With the calculated parameters, the new model allows one to predict the hardness of three-phase cemented carbides highly efficiently.

2. Development of the hardness model

The LG model is the most suitable one and chosen as the basis of this work, according to the analysis in the introduction, although it has the limitation that more parameters are used. For LG model, 4 parameters are needed to calculate the hardness of WC–Co, which are C, V_{WC}, d, and λ. According to Equation (4), the parameter λ can be expressed as a function of d, $V_{Co,}$ and C. It should be noted that the addition of cubic phase will affect the mean free path of the binder phase. The new equation for calculating λ can be derived based on stereological analysis as follows. Assume that the numbers for each kind of particle per unit amount of the alloy are Nwc, $N_{Cubic,}$ and N_{Co}, respectively. Then the total volume v_{WC}, v_{Cubic}, $v_{Co,}$ and total surface area S_{WC}, S_{Cubic}, and S_{Co} per unit amount of the alloy for each phase can be approximately expressed as shown in Equations (5) and (6), where d_{WC} and d_{Cubic} are the grain size of WC and cubic phase, respectively. According to the definition of contiguity, Equation (7) can be obtained. Assuming that the particles are spheres and combining equations (5)-(7), redundant terms can be eliminated and Equation (8) can be obtained through simple mathematical transformation. Actually, the shape of particles will affect the proportions in Equations (5) and (6). However, the proportions can be eliminated as long as the shapes of each kind of particle keep constant. The same method has been used for the WC-Co cemented carbides, and Equation 4 was the result. As shown in Equation (8), λ can be expressed as a function of C, V_{WC}, V_{Cubic}, V_{Co}, d_{WC} and d_{Cubic}, where V_{WC}, V_{Cubic}, V_{Co} are the volume fraction of each phase, C is the contiguity discussed below.

$$v_{WC} \sim {d_{WC}}^3 N_{WC} \quad v_{Cubic} \sim {d_{Cubic}}^3 N_{Cubic} \quad v_{Co} \sim \lambda^3 N_{Co} \tag{5}$$

$$S_{WC} \sim {d_{WC}}^2 N_{WC} \quad \mathrm{S}_{Cubic} \sim {d_{Cubic}}^2 N_{Cubic} \quad \mathrm{S}_{Co} \sim \lambda^2 N_{Co} \tag{6}$$

$$C = \frac{S_{WC} + \mathrm{S}_{Cubic} - \mathrm{S}_{Co}}{S_{WC} + \mathrm{S}_{Cubic}} = 1 - \frac{\mathrm{S}_{Co}}{S_{WC} + \mathrm{S}_{Cubic}} \tag{7}$$

$$\lambda = \frac{V_{Co} d_{WC} d_{Cubic}}{(d_{WC} V_{Cubic} + d_{Cubic} V_{WC})(1 - C)} \tag{8}$$

Lee and Gurland [5] assumed that WC forms a continuous skeleton in WC–Co and so does Co binder phase. However, new carbide-carbide contacts were created such as WC/Cubic and Cubic/Cubic by the addition of cubic carbide phase. It was confirmed that the carbide grains form a WC–Cubic- skeleton [14]. Since the LG model was used to calculate the hardness of Ti(C, N)–Co/Ni alloys successfully [12], we can assume that the contribution of WC–Cubic-skeleton to the hardness of cemented carbides can be treated in the same way as WC skeleton. Under this assumption, we could suggest that the contiguity C has a combined effect of $C_{WC/WC}$, $C_{WC/Cubic}$, $C_{Cubic/Cubic}$. Contiguity is the ratio of the grain boundary area to the total interface area of carbide grains according to its definition [11]. In WC-Co cemented carbides, it is determined by the WC/WC grain boundary and WC/Co interface areas. Two different approaches were developed to estimate the contiguity. One of them is the equilibrium approach which considers that the minimization of total interface energy drives the development of contacts. Another one is the geometrical approach which assumes that contacts are formed to fill a given volume where overlapping is necessary. From these points of view, many factors might be related to the contiguity of carbides, such as interface energy, the processing history, the shape of particles, and soforth. In German's work [15], the equilibrium approach was used to analyse the contiguity of carbides expressed as a function of grain size ratio, volume fraction of carbides, and dihedral angle (related to interfacial energies). The grains were assumed to be spherical with isotropic

interfacial energies, and the binder was considered as liquid in German's work [15]. The calculation is still complicated under this simplification and as concluded in the paper, anisotropic surface energy and shape accommodation effects should be considered in the future to estimate contiguity more accurately. However, the shrinkage during sintering would create a lot of contacts, which certainly affect the grain boundary area. Nevertheless, the preferential elimination of high-energy boundaries interface energy might also minimize the total interface energy, while the number of grain boundaries keeps the same. These factors make the analytical prediction of contiguity very hard. Luyckx and Love [16] gave another solution from the view of stereological method combined with empirical fitting to the experimental data. They found that the ratio of binder mean free path to carbide grain size can be expressed as a function of cobalt volume fraction. With the relationship given by Equation (4), the contiguity of carbides can be calculated only by cobalt content. Roebuck and Bennett [17] analyzed the phase size distribution in WC-Co cemented carbides and gave a similar conclusion - a reasonably accurate estimate of the cobalt mean free path can be obtained from WC grain size and the cobalt volume fraction. This means that there is a relationship between the ratio of mean free path of binder to grain size of carbide and volume fraction, which supports the results of Luyckx and Love. The calculated contiguity using the function proposed by Luyckx and Love agree with the experimental results reasonably, although the measured values often show large scatter. Following the method of Luyckx and Love, the contiguity can be expressed as Equation (9) through transformation of Equation (8). Then, we use the ratio of mean free path of binder to grain size of carbide given by Luyckx and Love, the volume fraction of each phase and the grain size ratio of WC to Cubic to estimate the contiguity. Bhaumik et al. [18] reported that the contiguity of carbides remains at almost the same level when the amounts of TiC are added. In their samples, the WC/TiC grain size ratio almost kept the same, as well as the volume fraction of the binder. This would support the present model.

$$C = 1 - \frac{V_{Co}}{(V_{Cubic}\, d_{WC}/d_{Cubic} + V_{WC})\,\lambda/d_{WC}} \tag{9}$$

At the same time, the contribution of carbide skeleton to the hardness should be modified following the rule of mixture since the carbide skeleton has cubic phase participates and both carbides have different grain sizes. The core-rim structure is often observed in the Cubic containing cemented carbides. The core-rim structure is formed by the precipitation of the (Ti,W)C from the liquid phase onto undissolved TiC grains during sintering [14, 19]. The core-rim boundary is purely compositional and the rim is essentially free of dislocations [20]. Of course, the core and the rim have different mechanical behavior at their local regions. However, from the view of macro level, the core and the rim can be seen as one particle. Thus, the core-rim structure of cubic phase is treated as a whole which include both the core and the rim, and the cubic grain is seen as homogeneous.

The effect of cubic phase on the hardness of Co binder needs to be considered. The Co binder has been reported to be a mixture of brittle hcp and ductile fcc phases [21]. The dissolved atom and residual stresses are probably the reasons why Co binder remains its high temperature form of fcc [22]. However, hcp Co has also been found in cemented carbides. As pointed out by Walbrühl et al. [22], the most prominent hardening effect of binder is the solid solution strengthening, and the fcc to hcp phase transformation can be neglected in their work of building a hardness model for the cemented carbides with alternative binders. It has been reported that the Co binder phase contained very little Ti [14]. Co has been used as major binder element in cemented carbides, and the processing conditions are usually very similar in the industrial environment [22]. Thus, different grades of cemented carbides potentially have similar binder hardness. Lee and Gurland reported the binder hardness with the Hall-Petch relation as a function of mean free path, and it was used in many works including Ti(C, N)-Co alloys [7, 12]. It seems that the hardness of the binder has been well established and might not need to be modified when considering those systems. As a result, the binder hardness reported by Lee and Gurland is used in this work.

As a result, we developed a new equation to calculate the hardness of WC–Co–Cubic, as shown in Equation (10). The hardness of cubic phase can be calculated from Equation (11), which is obtained from the literature [12].

$$H_{CC} = (H_{WC}V_{WC} + H_{Cubic}V_{Cubic})C + H_{Co}[1 - (V_{WC} + V_{Cubic})\,\mathrm{C}] \tag{10}$$

$$H_{Cubic} = 16.5 + \frac{1.8}{\sqrt{d_{Cubic}}}\,(\mathrm{GPa}) \tag{11}$$

According to the above equations, volume fraction of each phase and the grain sizes of carbides are needed to predict the hardness. Based on our thermodynamic and atomic mobility databases for cemented carbides [24, 25], the volume fraction of each phase can be calculated using Thermo-calc and DICTRA softwares as described by Zhang et al. [24]. Thus, we only need the initial compositions and grain sizes of carbides to calculate the hardness. The

presently developed model is consistent with the LG model. When the volume fraction of cubic phase is zero, Equation (10) reduces to Equation (1) of the LG model. Thus, we can say that the LG model is the application of the present model to two-phase WC–Co cemented carbides.

3. Results and discussion

To check the reliability of the presently developed hardness model, a series of calculations are made as shown in Figs. 1 a-d. The expected trends can be seen in Fig. 1 a-c: hardness decreases with the increase of cobalt volume fraction and the increase of grain sizes of WC and Cubic phases. Fig. 1 d shows that the addition of finer cubic phase will increase the hardness when the cobalt volume fraction remains constant, while the coarser cubic phase added the opposite.

Hardness of WC–Co–TiC cemented carbides was reported by Lee et al. [13]. The volume fraction of each phase can be calculated by using our thermodynamic database applied to the composition given in [13]. Haglund and Ågren [26] showed a good agreement for a computational assessment using DICTRA for substitutional W in Co binders with the experimental values of Hellsing [27]. Their results showed that the W solubility only changed minimally below 1000 °C. So, we assume that the microstructure of cemented carbides does not change noticeably below 1000 °C. The calculated values of phase fractions at 1000 °C are taken here to calculate the hardness of WC–TiC–Co cemented carbides. Figures 2-3 show the calculated results along with the experimental data, demonstrating that the calculated hardness can reproduce the experimental data [13] reasonably. Most of the calculated hardness values agree with the measured values within 10%. Fig. 3a shows the results of WC–Co cemented carbides. This is the situation where the volume fraction of cubic phase is zero. Fig. 3b-e show the results of WC–Co–TiC cemented carbides with different amounts of TiC added, indicating good agreements to the experimental results.

However, the use of the ratio λ/d_{WC} in WC-Co alloys to calculate the contiguity as described in section 2 may introduce a question whether the relationship still holds in WC-Cubic-Co. The basis of Luyckx and Love's work on the contiguity of WC is a hypothesis that two WC–Co grades with equal cobalt content but different grain size "x" and "ax" would appear the same in the micrographs at magnifications of "ay" and "y", respectively [16]. This basis still holds when WC/TiC grain size ratio is close to 1. When WC/TiC grain size ratio deviates from 1, the contiguity might not be influenced by the change of grain sizes at certain WC/TiC grain size ratio. However, the real value of ratio λ/d_{WC}

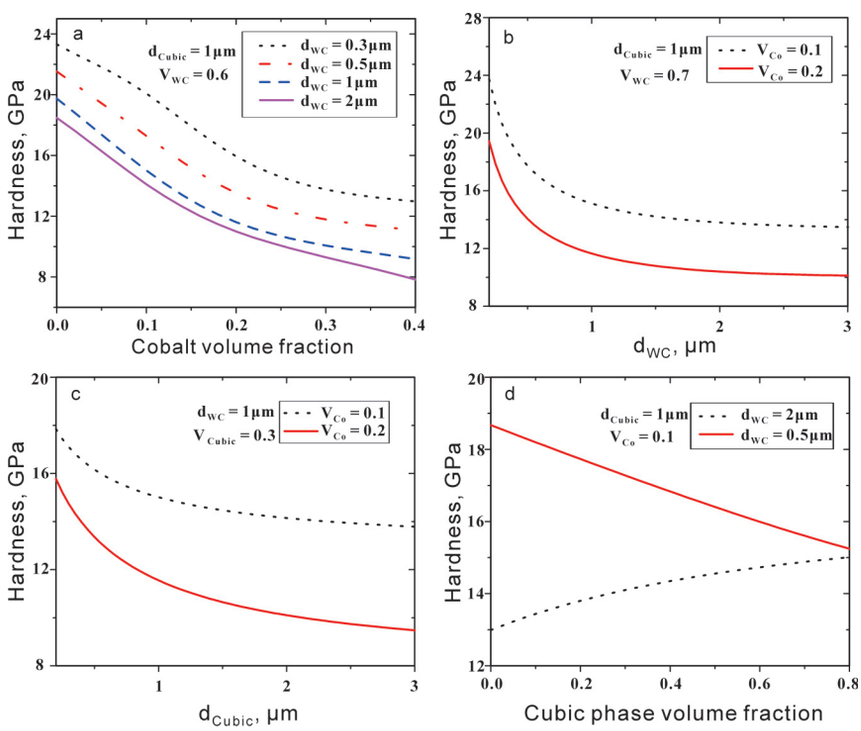

Figure 1. *Calculated hardness a) vs. cobalt volume fraction with a number of carbides grain sizes; b) vs. WC grain sizes with a number of volume fraction; c) vs. TiC grain sizes with a number of volume fraction; d) vs. carbides volume fraction with a number of carbides grain sizes*

might deviate from the calculation when WC/TiC grain size ratio deviates from 1. As can be seen in the reproduced figure 3d and 3e, the calculated hardness of samples with the WC/TiC grain size ratio of 0.5 is lower than the experimental data while the hardness of the samples with the ratio of 4 is higher. This is because small WC/TiC grain size ratio makes the calculated contiguity lower than real values when λ/d_{WC} is not adjusted, and the large ratio is the opposite. The higher cubic phase content, the more obvious the deviation. In spite of this, the introduction of WC/TiC grain size ratio to calculate the contiguity explained the phenomenon reported by K. H. Lee et al. [13] that when WC/TiC grain size ratio decreases to 0.5, the hardness increases much higher. The calculation reproduces the trend of experimental data successfully. Frankly speaking, it is arbitrary to calculate the contiguity of carbides in WC-Cubic-Co alloys using the ratio λ/d_{WC} reported by Luyckx and Love directly. Cautions should be taken when calculating the hardness of cemented carbides with WC/TiC grain size ratio deviates far from 1, especially when the TiC content is high.

The hardness distribution of a WC–Co–Ti(C, N) gradient cemented carbide is also calculated using the present model. The phase fraction distribution is simulated by DICTRA using the thermodynamic and diffusion databases, CSUTDCC1 [23] and CSUDDCC1 [24] developed in our group, which were successfully used to simulate the formation of gradient zone in many cemented carbides. With the calculated phase fraction distribution [25] and measured grain size of carbides, the predicted hardness of gradient cemented carbides vs. distance is obtained, as shown in Fig 4. The model predicted hardness results agree with the experimental data [25] reasonably.

As shown in Figures 2 and 3, several of the model-predicted hardness values are in some disagreement with the experimental values. The reason may be that a real WC–Co–TiC system has much more features than assumed by the present model, and the simplified process would introduce certain errors. To improve the accuracy of the presently proposed model, the composition of the cubic phase should be considered since the solution of tungsten atoms may affect the hardness of the cubic phase. Another aspect is that the suggested contiguity C which has a combined effect of $C_{WC/WC}$, $C_{WC/Cubic}$, $C_{Cubic/Cubic}$ is calculated approximately. There should be more experimental and analytical work focus on this point to improve the understanding and calculation of contiguity in the future. We suggest that a more reliable relationship between phase volume fractions and λ/d_{WC} should be built based on plenty of experimental data, which will deepen the understanding of contiguity. A comprehensive analytical model on the contiguity of carbides is still in need. Recently, the phase field simulation of structure evaluation in cemented carbides has been reported [28]. The phase field simulation may improve the calculation of parameters needed to predict the hardness, such as contiguity and phase volume fractions.

The present model should be used with caution when the process of manufacturing cemented carbides is tailored from conventional methods. For example, it has been reported that the extremely fine Co_3W nanoparticles precipitated in the cobalt binder would increase the hardness and strength of cemented carbides dramatically [29]. Another example reported by Shatov et al. [10] is that the addition of TiC to WC-Ni cemented carbides make the shape of WC crystals turn into a flatter triangular prism. When the shape of carbides is tailored, the present model might need to be modified before being used. Of course, it should also be noted that this model is valid when the carbide skeleton is maintained, since this is the basic assumption of the present model. Besides, care should be taken when the WC/TiC grain size ratio deviates far from 1, especially when the TiC content is higher than 20 wt. %. However, the present model can provide a reasonable prediction on the hardness of WC–Co–Cubic cemented carbides with limited parameters obtained from experiments, thus helping the design of the new type of cemented carbides.

Table 1. *The hardness of individual phases used in the present model*

Phase	Hardness (GPa)	Reference
Co	$2.98+\frac{3.9}{\sqrt{\lambda}}$	[5]
WC	$13.5+\frac{7.2}{\sqrt{d_{WC}}}$	[5]
Cubic	$16.5+\frac{1.8}{\sqrt{d_{Cubic}}}$	[12]

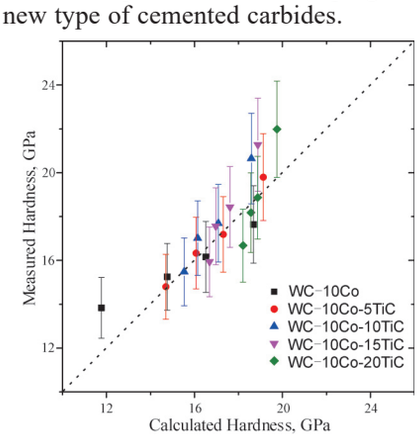

Figure 2. *Calculated hardness of WC–Co–TiC cemented carbides compared with the experimental data [13]*

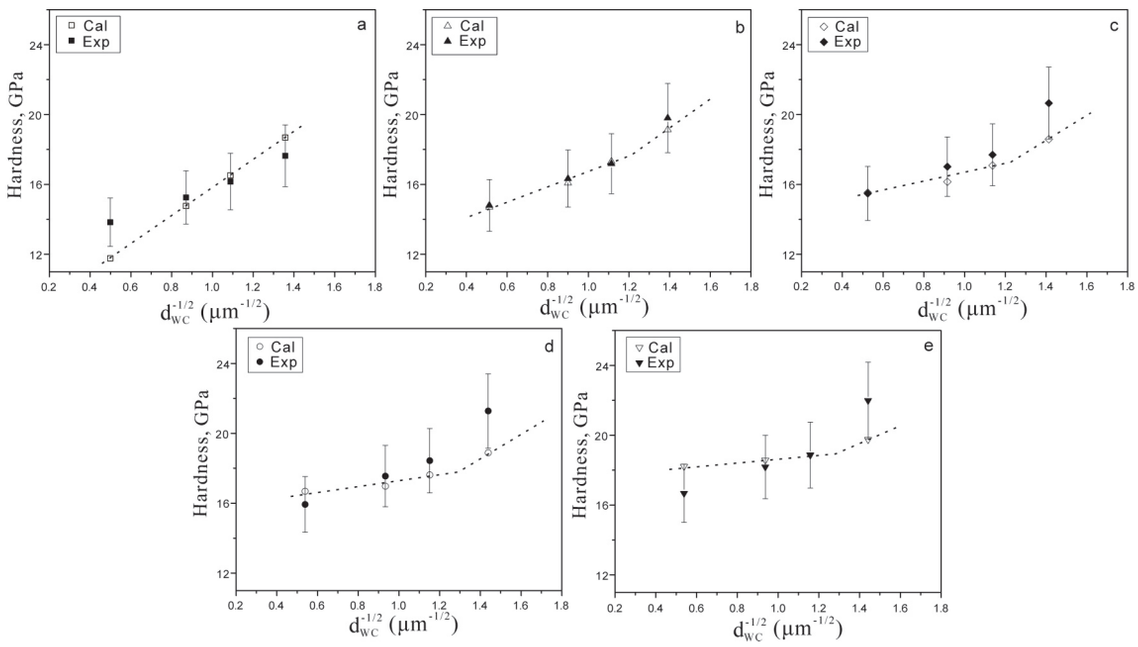

Figure 3. *Calculated hardness of WC–10 wt. %Co–x wt. %TiC cemented carbides compared with the experimental data [13]: (a) x=0; (b) x=5; (c) x=10; (d) x=15; (e) x=20*

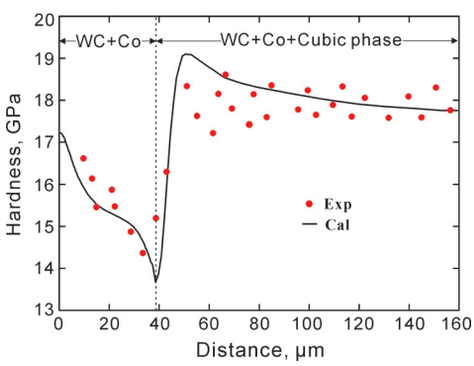

Figure 4. *The presently calculated hardness against distance along with the experimental data [24]*

4. Conclusion

In summary, we have developed a model, which can calculate the hardness of WC–Co–Cubic cemented carbides. With the volume fractions of the phases from CALPHAD calculations, we only need the original composition and the grain size of carbides to predict the hardness of cemented carbides with three phases. The present model is used to calculate the hardness of a series of WC–Co–TiC alloys as well as WC–Co–Ti(C, N) gradient cemented carbides. The calculated results show that the present model can predict the hardness of three-phase cemented carbides reasonably. It is expected that the presently developed model can be used as a reference to predict the hardness for multi-phase composites and design the new type of WC–Co-based cemented carbides efficiently.

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UPROŠĆENI MODEL TVRDOĆE WC–Co-KUBNIH CEMENTIRANIH KARBIDA

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Apstrakt

Tvrdoća je bitan mehanički pokazatelj cementiranih karbida. U ovom radu predstavljen je pristup za predviđanje tvrdoće trofaznih WC–Co-kubnih cementiranih karbida koji utvrđuje odnos između sastava, strukture, i mehaničkih performansi. Sa unosom inicijalnog sastava i veličine zrna karbida, a uz pomoć termodinamičkih proračuna i simulacija difuzije, mogu se izračunati strukturni parametri potrebni da se predvidi tvrdoća. Proračunata tvrdoća niza WC–Co–kubnih cementiranih karbida se prilično slaže sa eksperimentalnim podacima. Ovaj model je referentan za predviđanje tvrdoće višefaznih kompozita i dizajniranje novih tipova WC–Co cementiranih karbida.

Ključne reči: *Model tvrdoće; WC–Co–kubni cementirani karbidi; CALPHAD*