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The Application of Computer Program for Determination of Fluid Properties and P-T Condition from Microthermometric Data on Fluid Inclusions

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ABSTRACT: Fluid inclusion has been widely used to study the origin and physiochemical conditions of ore deposits. However, it is difficult to get the accurate physiochemical data from fluid inclusion study due to the error of microthermometric data and the complexity of calculation of density and isochore of fluid inclusion. The computer programs HALWAT, CO_2, and CHNACL written by Nicholls and Crowford (1985) partly contributed to improve the accuracy of physiochemical data by using complicated equations. These programs are applied to determine the densities and isochores of fluid inclusions for the Cretaceous Keumhak mine using Choi and So's data (1992) and for the Jurassic Samhwanghak mine using Yun's data (1990). The estimated P-T for Keumhak mine from calculated isochores of coexisting fluid inclusions are 230°~290℃ and 500~800 bar which matche well to the P-T estimated by Choi and So (280°~360℃ and 500~800 bar, 1992). However, the P-T for Samwhanghak mine estimated in this study by combining the calculated isochores and sulfur isotope geothermometer data by Yun (1990) are about 4~7 kb at 329± 50°~344± 55℃ which are quite different form the P-T estimates by Yun (255°~294℃ and 1.2~1.9 kb, 1990). This discrepancy caused by misinterpretation of homogenization temperature (Th) of fluid inclusion and by application of inappropriate isochores. The application of homogenization temperature and/or inappropriately selected isochore to determine the trapping P-T condition of ore-deposits should be avoided, particularly for ore-deposits formed at pressures higher than 1~2 kb.

INTRODUCTION

Fluid inclusion has been used to study the origin and physiochemical conditions of ore deposit (Roedder, 1976 and 1979; Brown and Lamb, 1986; Choi and So, 1992). Fluid inclusion study can be also widely applied to the metamorphic, igneous, and diagenetic sedimentary studies (Touret, 1977; Crawford, 1981; Roedder, 1984). The applications of fluid inclusion are based on microthermometric data such as homogenization temperature, melting temperature and volume estimation of each phase of fluid inclusion. In spite of the broad application of fluid inclusion, it is difficult to get the accurate physiochemical data such as density, isochore, and P-T condition of fluid inclusion and mole fraction of each phase. The accuracy of physiochemical data is caused by the error of microthermometric data, particularly in volume estimation, and the complexity of calculation of density and isochore of fluid inclusion. Recently, the accuracy of physiochemical data was partly improved by the computer programs suggested by Bowers and Helgeson (1985) and Nicholls and Crowford (1985) in which very complicated equations are used to calculate the density and isochore of fluid inclusion.

The P-T conditions of Cretaceous and Jurassic vein deposits in Korea are still in argue due to the inaccurate physiochemical data from fluid inclusion study. In this paper, as reconnaissance study, Nicholls and Crowford's programs (1985) are applied to determine the P-T conditions of Cretaceous Keumhak and Jurassic Samhwanghak mines. The difference between the P-T estimates in this study and those in previous studies are discussed.

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PROGRAMS USED IN THIS STUDY

The programs used in this paper are HALWAT, CO_2, and CHNACL written by Nicholls and Crowford (1985) and the numbers 0.0174 in line 115 of the program CO_2 and in lines 127 and 270 of the

Table 1. Input data for three programs.

Program	Input Data
HALWAT	Th-H_2O
	Tm-Clath or TL-ice or TL-NaCl
CO_2	Th-CO_2
CHNACL	Th-CO_2
	whether the CO_2 phase homogenized to a liquid or gas
	Vol-CO_2
	The temperature at which Vol-CO_2 is estimated
	Whether a clathrate melting was observed
	Tm-Clath (if observed)

* Th-H_2O and ThCO_2; homogenization temperatures of H_2O and CO_2, Tm-Clath; melting temperature of clathrate, TL-ice; liquidus temperqture of ice, TL-NaCl; liquidus temperature of NaCl, and Vol-Co_2; volume fraction of CO_2.

program CHNACL are replaced by 0.0714 following the suggestion by Brown and Lamb (1986). HALWAT, CO_2, and CHNACL programs are used to calculate the densities and isochores of fluid inclusions consisting of H_2O+NaCl, CO_2, and $H_2O+CO_2\pm$NaCl, respectively. The input data for these programs are shown in Table 1.

HALWAT Program

The density and isochore are calculated by combining the data in Potter and Brown (1977) and lest-squares treatment. The salinity is calculated from a function of the melting temperature of the clathrate (Hendell and Hollister, 1981), the liquidus temperature of ice (Potter et al., 1978), or liquidus temperature of halite (Hass, 1976) depending on which phase is designated on input.

CO_2 Program

The density is calculated by the equation recommended by Angus et al. (1976). Isochore is calculated in two ways: one set is calculated by using IUPAC equation (International Union of Pure and Applied Chemistry equation; Angus et al. 1976) and the other set by the Redlich-Kwang form (Holloway, 1981). The isochore from the IUPAC equation is more accurate at pressures less than 1,000 bar and the isochore from the Redlich-Kwang form at pressures higher than 1, 000 bar.

CHNACL Program

The density of CO_2 phase is calculated from the IUPAC equation (Angus et al., 1976) or Keenan et al. data (1969) and the density of H_2O phase and H_2O-NaCl solution from data of Keenan et al. (1969) and Potter and Brown (1977). The isochore, for binary CO_2-H_2O compositions, are calculated with the Redlich-Kwang equation given by Holloway (19 81) or with the polynomial expressions for the compressibility factor for CO_2-H_2O mixtures (Greenwood, 1973). The isochore for ternary composition (H_2O-CO_2-NaCl) is calculated with the Redlich-Kwang equation of state devised by Bowers and Helgeson (1983, 1985). If clathrate melting is observed, the calculation is made with the following assumption: (1) NaCl is present in the inclusion; and (2) the calculations are made on the assumption that the NaCl is confined to the H_2O phases. As a result, clathrate melting temperature should be less than homogenization temperature and the volume estimate must be made above the homogenization temperature of the CO_2 phase and above the melting temperature of any solids.

APPLICATION

Keumhak mine is one of the Cretaceous mine (Choi and So, 1992) and Samhwanghak mine is one of the Jurassic mesothermal type mine (Yun, 1990). The data produced from Fluid inclusion studies on these mines by Yun (1990) and by Choi and So (1992) are good enough for estimating pressure condition of ore deposits by applying the introduced programs. The pressure conditions of these two mines are estimated and the results are compared to the pressure estimates by Yun (1990) and by Choi and So (1992).

Keumhak Mine

The Keumhak Cu-bearing hydrothermal vein-type deposit located within the northwest of the Gyeongsang Basin, was mineralized at Late Cretaceous (77.6± 1.6 Ma) and probably associated with Late Cretaceous granitic rocks; in Keumhak mine, three stages of mineralization occurred and four types of fluid inclusions are observed. At stage I mineralization, quartz+sulfides+hematite+sulfosalts are mineralized; and at Stage II and III, quartz+sulfides and barren calcite are formed respectively. The main ore mineralization, stage I, can be classified into three substages (pyritic, basemetal, and sufosalt substages). Four types of fluid inclusions are

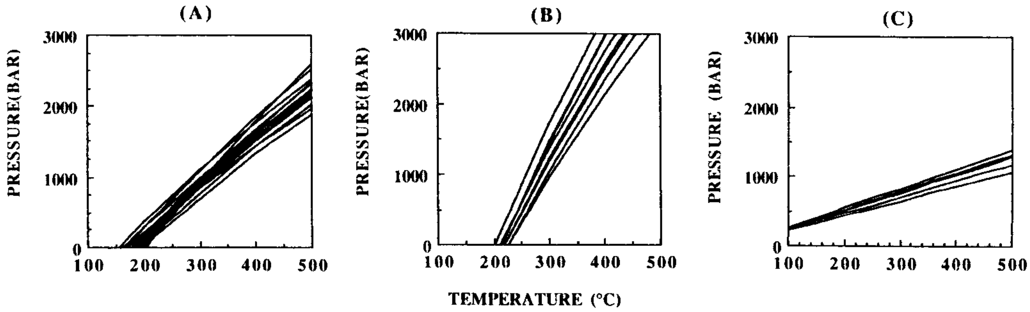

Fig. 1. The calculated isochores of Type IIIa (A), Type IIIb (B), and Type IV (C) fluid inclusions at Keumhak mine.

liquid-rich H_2O type (Type 1), vapor-rich H_2O type (Type II), CO_2-H_2O type (Type III) and CO_2 type (Type IV). All types of fluid inclusions have been observed in stage I vein quartz. Stage I sphalerite and fluorite, stage II vein quartz and stage III calcite sampes show Type I inclusions only. Type III fluid inclusions are classified into three subtypes; Type IIIa (CO_2-rich and homogenized to CO_2 phase), Type IIIb (H_2O-rich and homogenized to H_2O liquid phase) and Type IIIc (similar to type IIIb but homogenized to CO_2 phase). Choi and So (1992) suggested that Type IIIa and Type IIIb are coexisting fluid inclusions due to the fluid unmixing with decreasing temperature (380° to 280°C) of stage I and then, Type IIIc inclusions were resulted from the retrograde boiling of a H_2O fluid with decreasing pressure.

The estimated temperatures from sulfur isotope are 315± 55°C for pyritic stage, 244± 35°C for base-metal stage, and 131± 30°~218± 35°C for sulfosalt stage (Choi and So, 1992). The pressures for stage I mineralization were estimated by combining isochores of CO_2 inclusions (Type IV) and homogenization temperatures (281~365°C of CO_2-H_2O fluid inclusions (Type IIIa and IIIb) under the assumption that Type IIIa, Type IIIb, and Type IV inclusions coexisted during stage I mineralization. The estimated P-T condition of early part of stage I mineralization by Choi and So (1992) are 280°C<T<360° C and 500 bar<P<800 bar.

Isochores for Type IIIa, Type IIIb, and Type IV are calculated from published fluid inclusion data for Keumhak mine by Choi and So (1992). Fig. 1(A), 1(B) and 1(C) show three isochore sets calculated, respectively, for Type IIIa, Type IIIb and Type IV fluid inclusions. As these three fluid inclusions were suggested to coexist during stage I mineralization, the P-T area overlapped by the three sets of isochores represents the trapping P-T conditions for fluid inclusion which are equivalent to the P-T of stage I mineralization. The estimated P-T are 230°~290°C and 500~800 bar (Fig. 2) and match well to the P-T estimates by Choi and So (1992).

Samhwanghak Mine

Samhwanghak Au-bearing mine is Jurassic mesothermal-type gold deposit formed at about 146~151 Ma (So et al., 1989). Samhwanghak Au deposit has two paragenetic stages and three types of fluid inclusions (Yun, 1990). At paragenetic stage I, quartz +sulfide+economic concentrations of gold were deposited and at stage II, a post-ore barren calcite filled the newly opened faults and fractures. Pyrite, the earliest sulfide, rarely occurs and abundant sulfides are pyrrhotite, sphalerite and chalcopyrite. Galena and electrum were introduced after second fracturing and brecciation of earlier-deposited vein materials usually cementing the sphalerite and pyrrhotite. Gold occurs as small grains of electrum and native gold and gold grains frequently fill the fracture in sphalerite, galena and late quartz. Clear quartz, calcite and traces of pyrite and chalcopyrite occur within fractures of the third generation. Three types of fluid inclusions are Type I (CO_2-CH_4), Type II (CO_2-CH_4-H_2O± NaCl), and Type III (H_2O± NaCl). Type II inclusions can be subdivided into three types. Type IIa (vol% of carbonaceous phase> 55%), Type IIb (25%<vol% of carbonaceous phase<45 %), Type IIc (vol% of carbonaceous phase<10%). Type I and most Type II inclusions were considered to reflect the earliest fluids and the trapping P-T conditions of these fluid inclusions represent the P-T conditions of Samhwanghak ore-deposit. Type III were thought to be the latest fluid.

The estimated pressure conditions of Samhwanghak ore-deposit from the isochores of Type I fluid inclusions and homogenization temperatures of

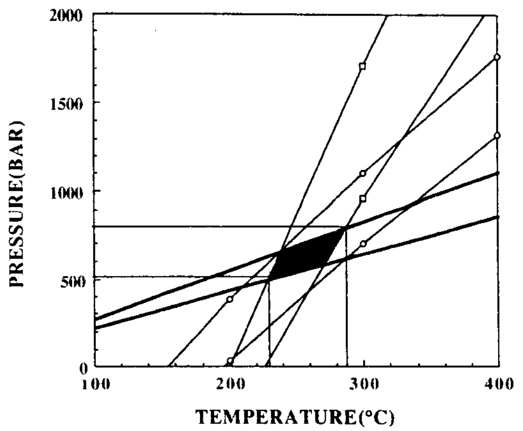

Fig. 2. The P-T conditions of stage I mineralization (filled P-T area) at Keumhak mine determined form isochores of coexisting Type IIIa, Type IIIb and Type IV fluid inclusions.

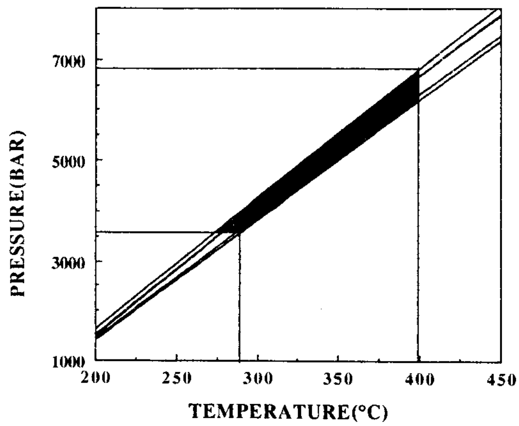

Fig. 3. The calculated isochores of Type IIc fluid inclusions at Samhwanghak mine and the P-T conditions of Samhwanghak mine (filled area) determined by combining calculated isochores and sulfur isotope geothermometric data.

Type IIc fluid inclusions (255°~294℃) are 1.2~1.9 kb; but the pressure estimated from sphalerite composition in the assemblage of sphalerite+pyrrhotite+pyrite is about 4.5 kb (Yun, 1990). For the pressure estimation, isochores of pure CO_2 instead of isochores of CO_2-CH_4 were used. The estimated temperature conditions of late mineralization of Samhwanghak ore-deposit from sulfur isotope are 327± 50°~344± 55℃ (Yun, 1990).

As Type I, Type IIa and Type IIb fluid inclusions contain CH_4, the isochores of the inclusions can not be claculated by the introduced programs. However, the CH_4 content of Type IIc inclusions is so low as to be neglected (less than 1 mole percent) in isochore calculation by using CHNACL program. The calculated isochores for Type IIc inclusions from the published data by Yun (1990) are shown in Fig. 3. The pressures estimated in this study by combining the isochores of Type IIc and sulfur isotope geothermometric data are about 4~7 kb and 327± 50°~344± 55℃. The estimated pressures are closer to the pressures estimated from sphalerite geothermometer than the pressures estimated from Type I isochores and homogenization temperatures of Type IIc fluid inclusions.

DISCUSSION

The pressure estimates for Keumhak mine in this study are well matched with the pressures estimated by Choi and So (1992) but the pressure estimates for Samhwanghak mine in this study are different from the pressures estimated by Yun (1990). This discrepancy in pressure estimation is caused by misinterpretation of homogenization temperature (Th) of fluid inclusion and by application of inappropriate isochores.

The homogenization temperature (Th) measured at 1 bar does not represent the trapping temperature (Tt) of fluid inclusion. For fluid inclusion filled with H_2O, CO_2, and H_2O+NaCl, the Th is the starting point of isochore and the difference between Th and Tt increases as trapping pressure condition increases due to the positive slope of isochore (Hollister, 1981; Bowers and Helgeson, 1985). For fluid inclusion of CO_2+H_2O+NaCl, each isochore has a fixed P-T condition for the transition from two phases (CO_2 +H_2O) into one phase (Roedder, 1984; Burruss, 1981). The final homogenization temperature for fluid inclusion of CO_2+H_2O+NaCl represents the transition point and is not the trapping temperature (Tt). Therefore, Th should not be used to estimated trapping pressure conditions. For the mesothermal type deposits such as Samhwanghak mine, the difference between Th ande Tt can cause a serious problem in estimating pressure. However, application of Th to pressure estimation in deposits formed at pressures less than 1 kb such as Keumhak mine causes a monor problem because of the small difference between Th and Tt resulted from low pressure conditions.

In most previous studies for fluid inclusions in Korea, the isochores are selected from published data

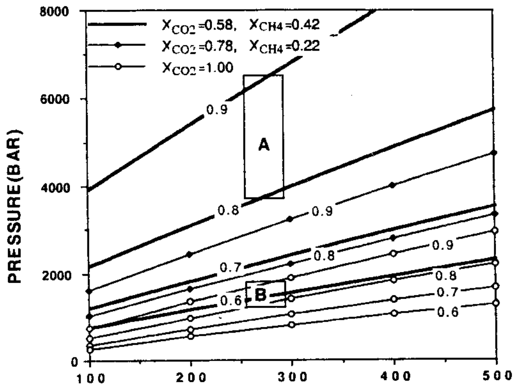

Fig. 4. The calculated isochores of CO_2-CH_4 fluid inclusions with densities of 0.9, 0.8, 0.7 and 0.6 for three different compositions (X_{CO2}=0.58, X_{CH4}=0.42; X_{CO2}= 0.78, X_{CH4}=0.22; Pure CO_2). A represent the P-T conditions estimated by using the isochores of CO_2-CH_4 fluid inclusions and B represents the P-T conditions estimated by using pure CO_2 fluid inclusions at 255°~294℃ from Samhwanghak mine.

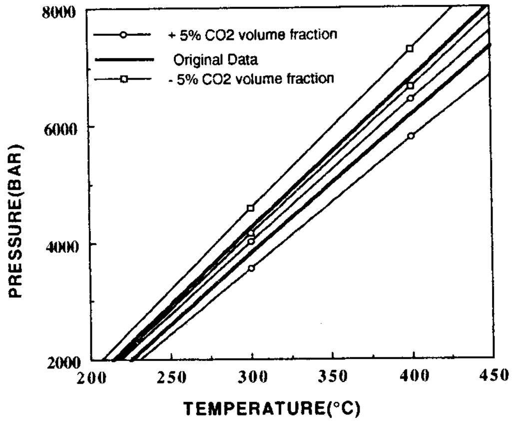

Fig. 5. The calculated isochores of Type IIc fluid inclusions at Samhwanghak mine with three different CO_2 volume fractions: (1); original CO_2 volume fraction, (2); CO_2 volume fraction increased by 5% compared to original one, and (3); CO_2 volume fraction decreased by 5% compared to original one.

instead of calculating isochores by using computer programs. The selection of isochore from limited published isochore data prohibits us from using adequate isochore. For example, in Yun's study (1990), the isochores of pure CO_2 used instead of the isochores of CO_2-CH_4 fluid inclusions to estimate the pressure of Samhwanghak mine. The difference between isochores for pure CO_2 inclusions and those for CO_2-CH_4 inclusions is evaluated in this study by calculating the isochores with density 0.9, 0.8, 0.7 and 0.6 for three fluid inclusions (X_{CO_2}=0.58, X_{CH_4}=0.42; X_{CO_2}=0.78, X_{CH_4}=0.22; X_{CO_2}=1.00) by using ISOCHORE program (Holloway, 1981). The calculated isochores of fluid inclusions filled with CO_2-CH_4 are quite different from those of the pure CO_2 fluid inclusions (Fig. 4). For example, at 300℃, the pressure for fluid inclusion with X_{CO_2}=0.58, X_{CH_4}= 0.42 and density of 0.9 is higher than that for pure CO_2 fluid inclusion by 4500 bar. As X_{CH_4} in CO_2-CH_4 fluid inclusion increases, the pressure condition for isochore increases. In Fig. 4. P-T region B represents the P-T estimates for Samhwanghak by using isochores of pure CO_2 fluid inclusions and P-T region A represents the P-T estimates for Samhwanghak by using isochores of CO_2-CH_4 fluid inclusions. The difference between the two P-T estimates is 2-5 kb. As the modification of isochore caused by the change of the mole fraction of CO_2, CH_4, and NaCl is very complicated (Bower and Holgeson, 1983 and 1985; Roedder, 1984), the application of inappropriate isochore always leaves a room for incorrect interpretation.

The error in volume estimation of fluid inclusion is a another obstacle to improve the accuracy of physiochemical data from fluid inclusion. To evaluate the effect of error in volume estimation on the isochore calculation, the isochores of Type IIc inclusions of Samhwanghak mine are calculated with different CO_2 volume fractions from original CO_2 volume fractions (Fig. 5). By increasing 5% of CO_2 volume fraction compared to the original data from Yun's study, the slopes of isochores decrease while the slopes of isochores increase by decreasing 5% of CO_2 volume fraction. The change of 10% CO_2 volume fraction causes 0.5~1 kb difference in pressure estimation at temperature range of 250°~400℃.

CONCLUSION

From this study the following conclusions are reached;

1. The P-T estimation for ore-deposits can be improved by using densities and isochores of fluid inclusions calculated by computer programs written by Nicholls and Crawford (1985); P-T estimation can be done by determining the P-T area where isochores of coexisting fluid inclusions intersect each other or by combining the calculated isochores and inde-

pendently determined pressures or temperatures.

2. For the determination of P-T conditions of ore-deposits, the application of homogenization temperature and/or inappropriately selected isochore should be avoided, particularly for ore-deposits formed at pressures highter than 1~2 kb.

3. The error in volume estimation by ±5% CO_2 volume fraction for fluid inclusions of Samhwanghak mine causes 0.5~1 kb diference in pressure estimation.

The computer programs (Nicholls and Crawford, 1985; Holloway, 1981) make it possible to calculate the physiochemical data of fluid inclusion not only more accurately but also much quickly. Therefore, it is strongly recommended to use the introduced programs to determine the physiochemical conditions for ore-deposits in Korea. The introduced programs can be also used for interpretation of metamorphic evolution, diagenesis and other petrological event.

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유체포유물의 생성시 온도-압력 조건과 유체포유물의 물리화학적 특성연구에 있어서의 컴퓨터 프로그램의 이용

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요 약: 유체포유물은 광상의 기원과 물리화학적인 환경 연구에 널리 사용되어져 왔다. 하지만 측정치의 오차와 유체포유물의 밀도와 isochore 등의 계산에 있어 그 과정의 복잡성 때문에 유체포유물에 내포한 물리화학적인 정보를 정확히 알아내는 것은 매우 힘들다. HALWAT, CO_2 그리고 CHNACL 등의 컴퓨터 프로그램들은 (Nicholl and Crowford, 1985) 유체포유물의 밀도와 isochore 등을 복잡한 공식을 이용하여 계산함으로서 유체포유물로 계산될 수 있는 물리화학적인 정보의 정확도를 개선하였다. 본 논문에서는 이들 프로그램을 백악기에 형성된 금학 광산에 대한 최상훈과 소칠섭 (1992)의 유체포유물 측정치와 쥬라기에 형성된 삼황학 광산에 대한 윤성택 (1990)의 유체포유물 측정치에 적용하여 보았다. 컴퓨터 프로그램을 이용하여 결정된 금학광산의 온도-압력조건은 230°~290℃, 500~800 bar로서 최상훈과 소칠섭 (1992)에 의하여 추정된 온도-압력조건인 280~360℃, 500~800 bar와 유사하다. 하지만 본 연구에서 결정된 삼황학 광산에 대한 온도-압력 조건은 대략 4~7 kb, 328± 50°~344± 55℃ 이며 이는 윤성택 (1990)에 의하여 추정된 온도-압력 조건인 1.2~1.9 kb, 255°~294℃ 와 차이를 보여준다. 삼황학 광산에서의 온도-압력 추정에 있어서의 차이는 광상의 온도-압력조건 추정에 있어 균일화 온도와 유체포유물의 포획온도를 동일시하고 적당치 못한 isochore의 사용함에 기인한다. 금학광상의 경우는 낮은 압력조건 (<1~2 kb)에서 균일화 온도와 유체포유물의 포획온도가 비슷하기 때문에 큰 차이를 보여 주지 않았다. 따라서 1~2 kb보다 높은 압력조건에서 형성된 광상에서는 광상의 온도-압력조건 추정에 있어 균일화 온도의 잘못된 적용과 적당치 못한 isochore의 사용을 피하여야 하며 이를 위하여 본 연구에서 소개된 컴퓨터 프로그램 이용을 추천한다.